Los Alamos

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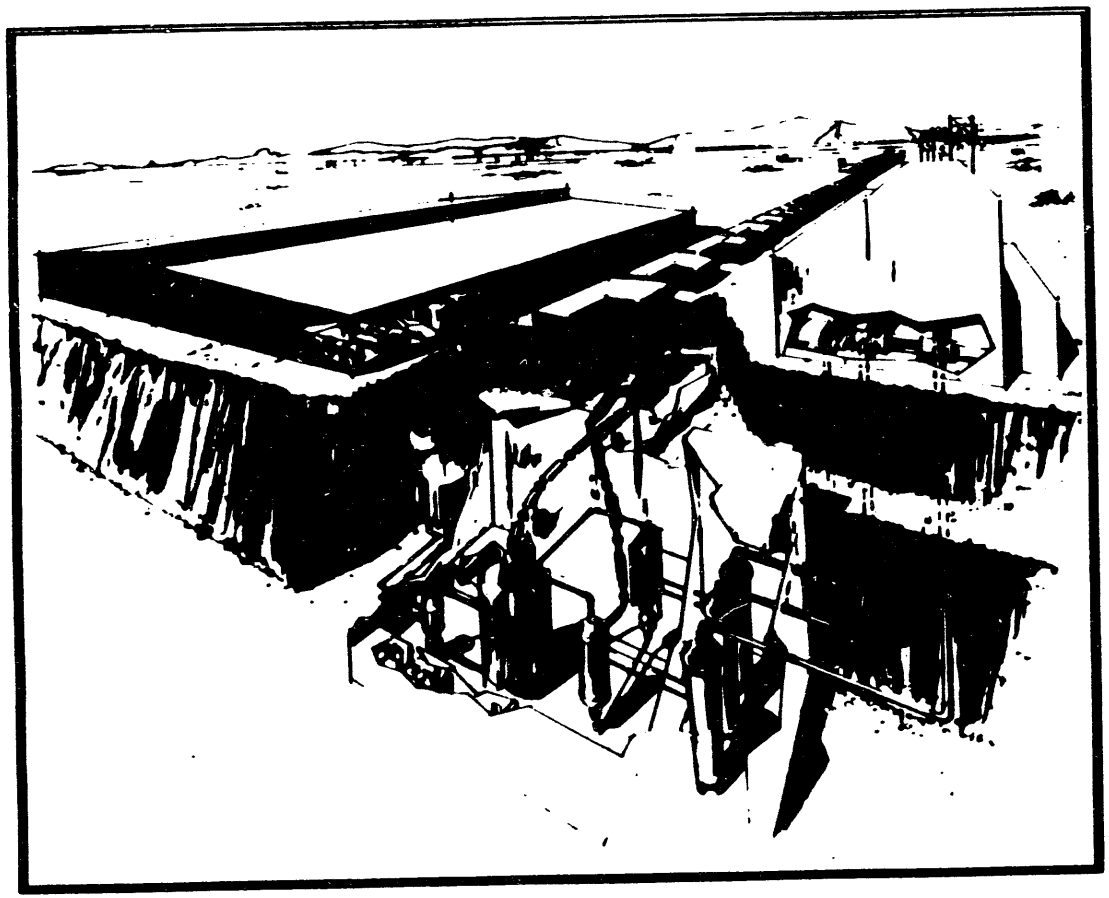

A Los Alamos Concept for Accelerator Transmutation of Waste and Energy Production (ATW)

December 10 - 12, 1990
External Review

ATW External Review
December 10-12, 1990
Hilltop House, Taos/Tyuonyi Room

Agenda

Monday, December 10, 1990 (1:00 p.m. - 5:00 p.m.)

CHARGE TO THE COMMITTEE

1:00 - 1:30	Charge to the Committee	Stan Schriber / Mike Stevenson
1:30 - 1:45	Committee Meeting	Committee Members Only

OVERVIEW OF THE CONCEPT

1:45 - 3:45	A New Los Alamos Concept for Nuclear Waste Transmutation and Fission Energy	Ed Arthur
	Near Term Application of ATW High-Level Defense Wastes — Hanford	Ed Arthur
3:45 - 4:00	*Break*	

LINAC TECHNOLOGY

4:00 - 5:00	Accelerator Technology for the ATW System	George Lawrence
5:00 - 5:30	Discussion	All
5:30 - 6:15	No-Host Reception (Taos/Tyuonyi)	
6:15 - 7:30	No-Host Dinner/Informal Discussions (Hilltop House)	By Invitation Only
8:00 - 9:30	Committee Meeting (Taos Room)	Committee Members Only

Tuesday, December 11, 1990 (8:00 a.m. - 5:15 p.m.)

8:00 - 8:30	Committee Comments/Agenda Discussions	

NEAR-TERM APPLICATION – HIGH-LEVEL DEFENSE WASTES

Intense Thermal Neutron Source

8:30 - 9:15	ATW Neutron Production	Paul Lisowski
9:15 - 9:45	ATW Target/Blanket Design: Application to Hanford Defense Waste	Mike Cappiello
9:45-10:15	*Break*	

CHEMISTRY AND MATERIALS

10:15 - 11:15	Aqueous Chemical Processing for the Tc and Tc/Np Transmutation Blankets	Steve Yarbro
11:15 - 12:00	ATW Materials Issues	Karl Staudhammer
12:00 - 1:30	*Lunch*	

ADVANCED APPLICATION OF THE ATW CONCEPT – FISSION ENERGY WITHOUT A HIGH-LEVEL WASTE STREAM

Overview

1:30 - 2:00	Overview of Advanced Application	Ed Arthur

Advanced Technology

2:00 - 3:15	Fission Energy Without a High-Level Waste Stream	Charlie Bowman
3:15 - 3:30	*Break*	

Advanced Chemistry

3:30 - 4:15	Molten Salt Chemistry for the Advanced ATW Concept	Scott Kinkead

SUMMARY

4:15 - 5:00	Summary and Research Issues	Ed Arthur
5:00 - 7:00	No-Host Dinner (Hilltop House)	Committee Members Only
7:00 - 9:30	Committee Meeting (Taos Room)	Committee Members Only

Wednesday, December 12, 1990 (8:00 a.m.- 10:00 a.m.)

8:00 - 10:00	Feedback from Review Team	

ATW External Review Team

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Charge to Committee
Accelerator-Based Transmutation of Waste (ATW)
1990 December 10-12
Hilltop House, Taos/Tyuonyi Room

Los Alamos National Laboratory has been working on a radioactive waste transmutation process based on spallation neutrons from a high energy proton accelerator target system. Very little funding has been available for this activity, therefore only pre-conceptual studies have been pursued. The Laboratory did fund an overall systems study mid-year 1990 that determined the feasibility of this accelerator-based process in the context of DOE defense waste applications.

The external review is considered a pre-conceptual design review and the review committee is asked to consider the following items in its deliberations.

1) Assess the state of the technology and the viability of this approach for having an impact on several potential application areas.
   — DOE Defense Waste
   — Nuclear Power Production

2) Determine areas requiring further study and make recommendations on actions/activities that should be pursued— including those not presently covered. Cover both technical and systems issues.

3) Make recommendations on actions the Laboratory should take:
   — Internal
   — Outside Community (technical, political, etc.)

OVERVIEW OF THE CONCEPT

A New Los Alamos Concept For Nuclear Waste Transmutation and Fission Energy

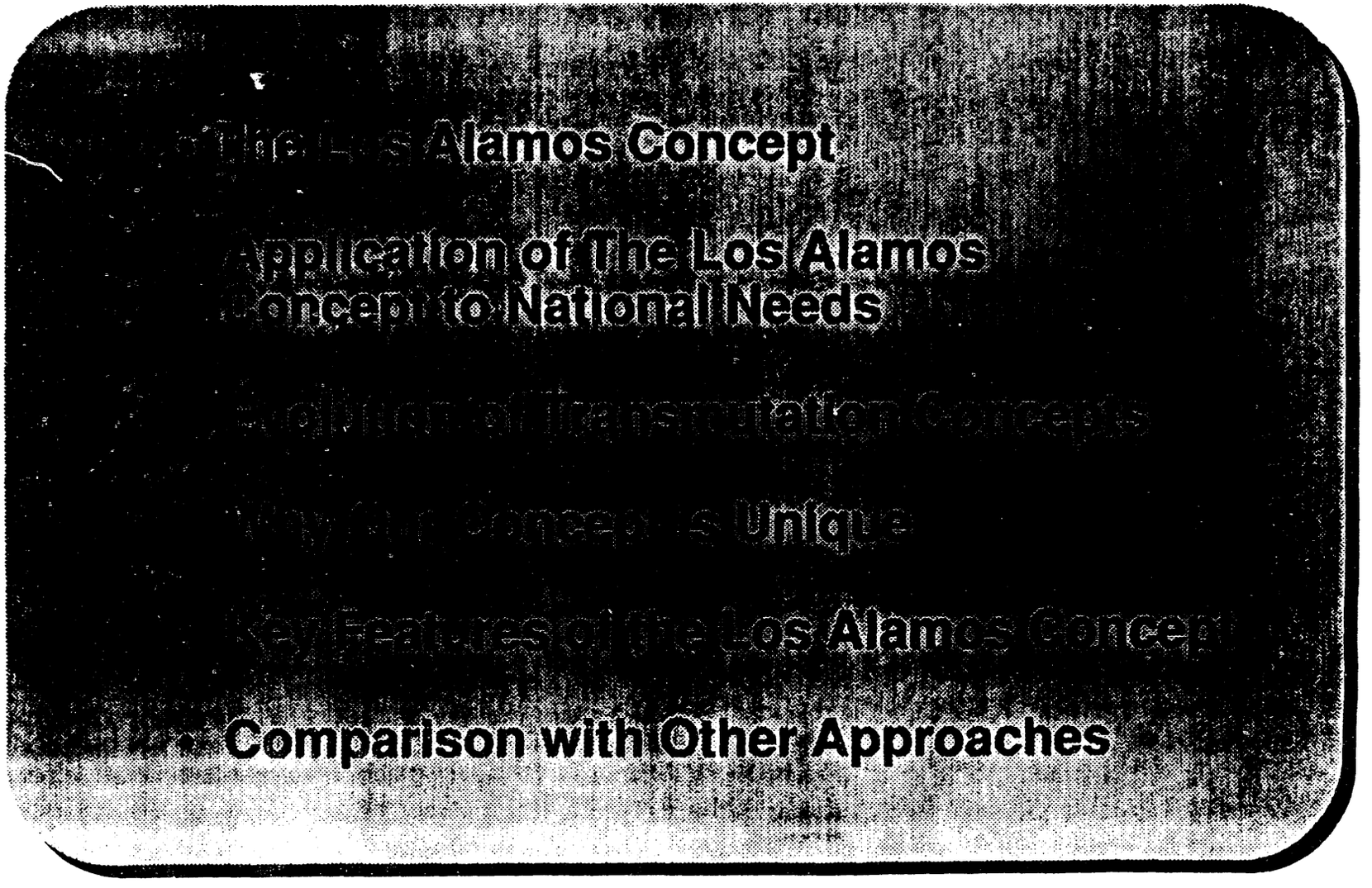
Edward Arthur
Theoretical Division

A New Los Alamos Concept for Nuclear Waste Transmutation and Fission Energy

Edward D. Arthur
Los Alamos National Laboratory

December 10, 1990

Overview


The Los Alamos Concept
Application of The Los Alamos Concept to National Needs
Unique
Comparison with Other Approaches

The Los Alamos Concept

We envision two focus areas for the Los Alamos Concept for Accelerator Transmutation of Waste and Fission Energy Production. The first is aimed at problems associated with high level defense wastes stored at several Department of Energy Sites. A more advanced, and farther term, effort is aimed at investigating the potential of such an accelerator-driven system on the production of fission energy with a minimal high-level, long-lived nuclear waste stream.

We often refer to our combination of concepts as ATW.

The Los Alamos Concept

INTEGRATED TECHNOLOGY
ATW

NUCLEAR WASTE STRATEGIES

- **DOE High Level Defense Waste Cleanup**
- **DOE Complex Reconfiguration**

ENERGY STRATEGIES

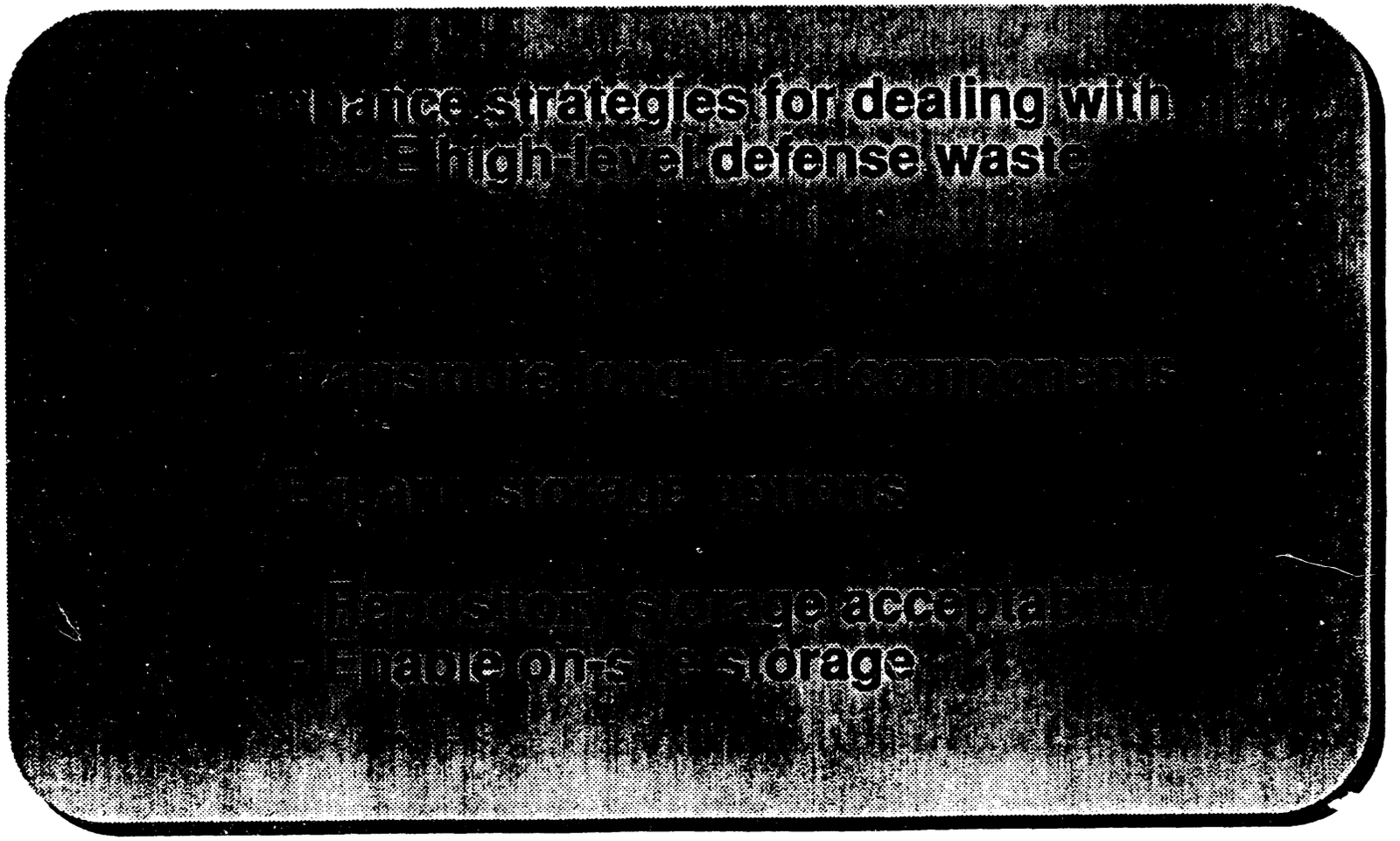
- **Fission Energy With a Minimal Long Term, High-Level Waste Stream**

What Can This Concept Do To Address National Problems

The near-term application of the Los Alamos Accelerator Transmutation Concept is directed at the problem area of DOE defense high-level wastes. These are stored at sites such as Hanford, Savannah River, and Idaho Falls. Our system could enhance strategies for dealing with these wastes by transmuting all long-lived fission products and actinides that comprise them. Specifically our system is aimed, in the near term, at transmutation of the long-lived species ^{99}Tc and ^{129}I which are fission products, as well as higher actinides such as ^{237}Np and $^{241,243}Am$.

Partitioning or chemical separation of these isotopes from waste forms stored at DOE sites, followed by transmutation, could achieve two results. By removing long-lived fission products (^{99}Tc in particular) that are the main contributors to long-term dose concerns, repository acceptability may be enhanced. Likewise, their removal may also permit on-site storage of the residue that remains after partitioning and transmutation have occurred.

What Can This Concept Do To Address National Problems?



The Los Alamos Concept Can Impact Energy Strategies

The features inherent in the Los Alamos Concept can have significant implication for new approaches to fission energy production. Specifically we are investigating the feasibility of a system that can produce fission energy efficiently, while burning its own fission product wastes. The end result could be a fission energy source with a long-term, high-level waste stream that is dramatically reduced when compared to present nuclear power reactor waste environments. This exciting possibility forms the longer term, advanced component of our concept.

As we describe later, the use of an accelerator-driven spallation neutron source increases substantially the effective number of neutrons per fission that can occur in an accelerator/multiplying blanket system. This feature could permit efficient generation of electricity, while providing a neutron-rich environment that can be used to transmute all fission products having half lives greater than 11 years.

The Los Alamos Concept Can Impact Energy Strategies

- Significant implications for fission energy production
- Near absence of long-term high level waste stream

Evolution of Transmutation Concepts

The Los Alamos concept for accelerator transmutation of nuclear waste is dramatically different than past or other current transmutation concepts.

Past approaches to transmutation have centered around two principal systems. The first, thermal reactor systems having low neutron flux levels, were considered for transmutation and rejected. This occurred because such systems could not fission higher actinide waste components that have small fission cross sections at low neutron energies. Also because of low flux levels, these systems could not transmute well fission products having very low capture cross sections. Likewise, these systems had very small volumes where higher flux levels were available.

In order to avoid the problems of such thermal systems, most transmutation concepts have centered around use of a fast neutron system. Such systems can be either reactor or accelerator driven. These systems burn higher actinides effectively because of higher cross sections that occur for fast neutron energies. However such systems require large material inventories to achieve suitable performance levels, and suffer from significant reactivity swings as fissile material is burned. Examples of current concepts based on the use of fast neutrons for transmutation include the Integrated Fast Reactor Project at Argonne National Laboratory, and the Japanese Omega Project.

Evolution of Transmutation Concepts

Low-flux thermal systems

- Low burn rate for small cross section fission products
- Low fission destruction rate
- Insufficient high-flux volumes

Fast neutron systems (reactor or accelerator driven)

- Difficulty in fission product transmutation
- Large inventories required
- Reactivity control concerns

We Utilize an Intense Thermal Neutron Flux in Our Concept

Our concept is quite different than previous approaches that have been proposed during the past several decades. With a high-current, high energy accelerator we can produce intense fluxes of thermal neutrons. The existence of an intense thermal neutron flux allowed us to find a new physical operating regime where higher actinides can be transmuted efficiently using low-energy neutrons. The existence of high fluxes also allows us to rapidly transmute waste products having low transmutation cross sections. Similarly, the availability of a high intensity neutron source has allowed us to identify a unique, low-inventory operating regime. Because our system has dramatically reduced material inventories, the chemistry requirements and options that we can pursue are simpler than other systems requiring large material inventories.

We Utilize An Intense Thermal Neutron Flux in Our Concept

This permits

- **New approach to actinide burning**
- **Rapid burning of low cross section waste products**
- **Miniscule waste inventory with high throughput**
- **Simpler process chemistry options**

Unique Physics and Operating Features

The unique physics and operating features of our concept are listed below. Again, they are enabled by the intense thermal neutron flux feature. Not only does this intense flux have significant implications for the physics and operating regime of the system, but the demands and features of the required chemical processing systems are changed quite dramatically over other concepts.

These features are discussed in more detail in the parts that follow in the presentation.

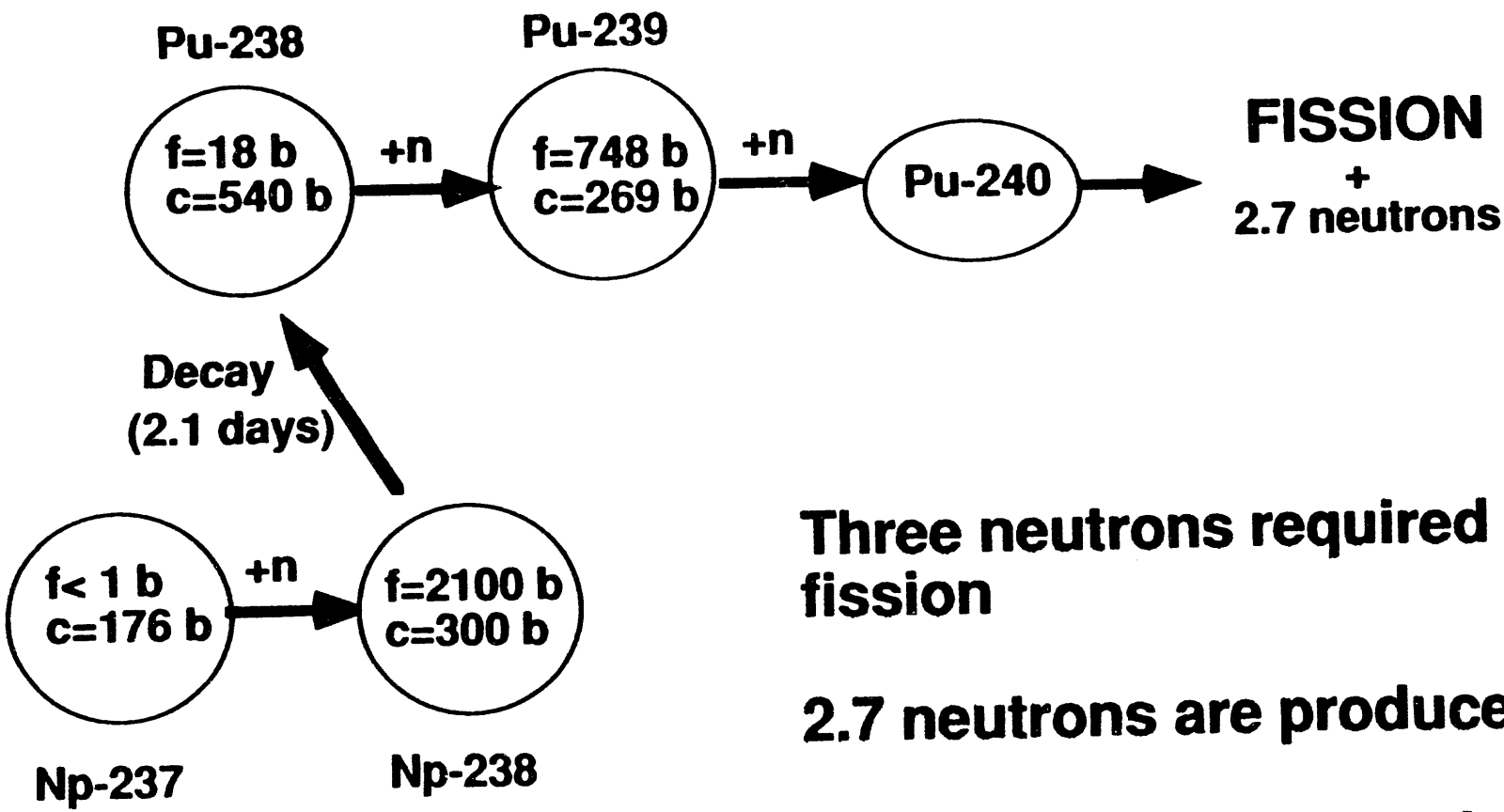
Unique Physics and Operating Features

- Two-step fission in an intense thermal flux
- New, dilute inventory operating regime
- Continuous material flow and low inventories → New chemical processing options

Low Intensity Thermal Neutron Flux- Difficult to Fission Higher Actinides

Before describing the process by which higher actinides are fissioned in an intense thermal neutron flux, it is useful to review the process occurring in a low-flux thermal environment. Higher actinides, such as ^{237}Np, have fission cross sections that exhibit low-energy thresholds. For ^{237}Np the fission cross section at thermal energies is quite small. Neutrons reacting with ^{237}Np are captured to produce ^{238}Np which decays in 2.1 days to ^{238}Pu. Likewise, ^{238}Pu has a relatively small fission cross section for thermal neutrons, so in most cases a neutron will be captured to produce ^{239}Pu. If a neutron then interacts with ^{239}Pu, it will most likely fission since its cross section is large. In the normal process that occurs in a thermal reactor about 2.7 neutrons are produced for each fission event. However, in this instance it has taken three neutrons to produce fission. Therefore, in a low thermal flux environment, ^{237}Np acts as a net absorber of neutrons.

Low Intensity Thermal Neutron Flux-Difficult to Fission Higher Actinides



Three neutrons required for fission

2.7 neutrons are produced

Neptunium is a net neutron absorber

$f = \sigma_f$

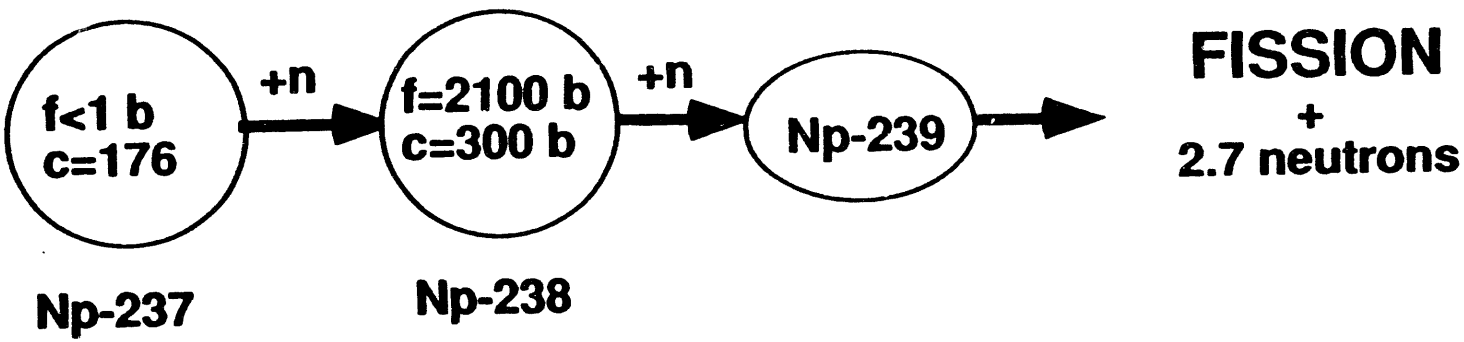
$c = \sigma_c$

A High Thermal Neutron Flux Allows Efficient Fission of Higher Actinides

In our high-flux thermal neutron environment, a quite different process occurs. After interaction of a neutron with a ^{237}Np target, ^{238}Np is produced. However because of the high flux, a second neutron is available to interact before decay occurs. Because the fission cross section for ^{238}Np is so large, such an interaction will most often lead to fission where again 2.7 neutrons are produced. In this case, two neutrons were required to produce fission. Therefore, ^{237}Np acts as a net producer of neutrons in this intense flux environment.

A similar situation holds for other threshold fission actinides such as $^{241,243}Am$ and ^{238}U.

A High Thermal Neutron Flux Allows Efficient Fission of Higher Actinides



Two neutrons produce fission

2.7 neutrons are produced

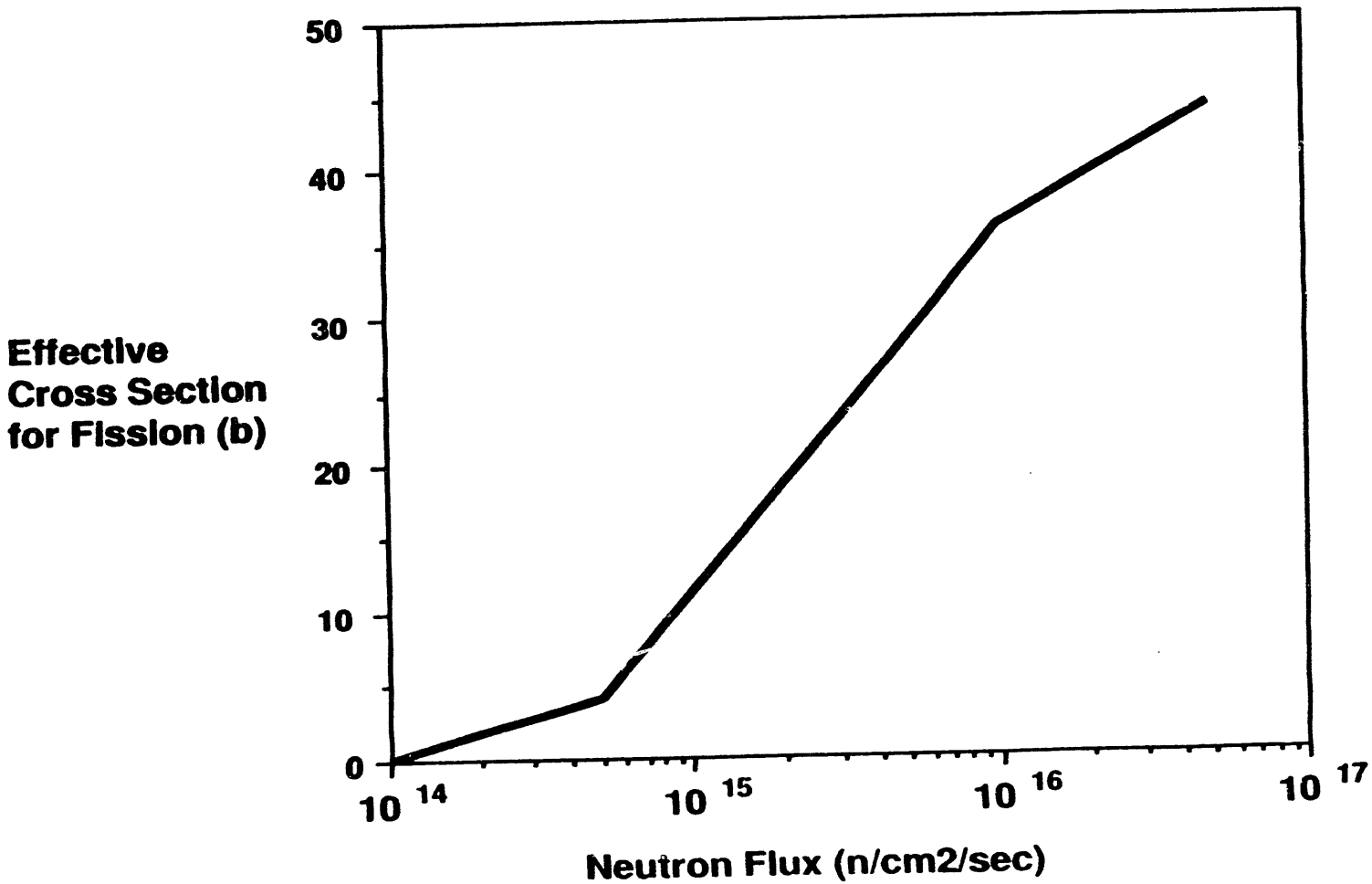
Net neutron production

Neptunium Fission in an Intense Thermal Neutron Flux

Another feature of neptunium fission in a high flux thermal neutron environment is the increase in the effective cross section for fission. This occurs because of the increasing probability for ^{238}Np fission that occurs at high flux levels. Here we have determined an effective neptunium fission cross section by determining concentrations of ^{237}Np and ^{238}Np appropriate for a given flux level, and multiplying them by a spectrum weighted fission cross section for our system.

For comparison, the fission cross section of ^{237}Np in a fast neutron environment is about 1.5 to 2 barns.

Neptunium Fission in An Intense Thermal Neutron Flux



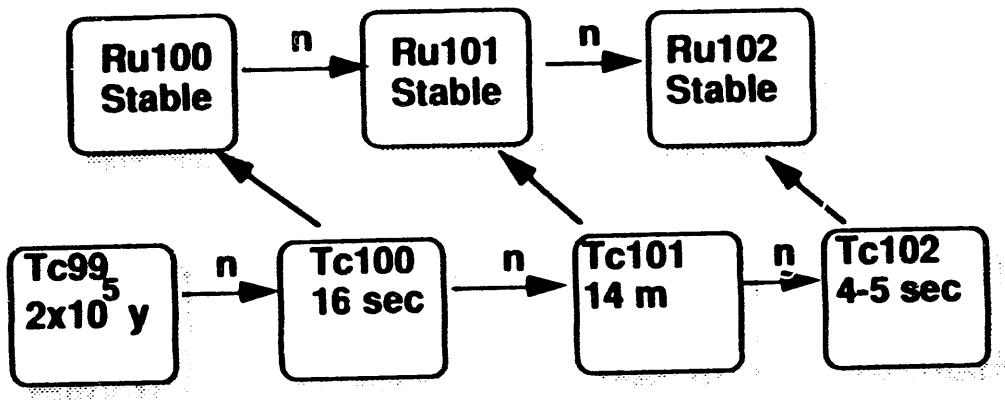
An Intense Thermal Flux Provides Rapid Fission Product Transmutation

The use of a thermal flux in our system also allows very efficient transmutation of fission products. In the example shown, ^{99}Tc captures a neutron to produce ^{100}Tc which decays after 16 seconds to the stable product, ruthenium. As illustrated, even if several successive neutron captures occur, the end result is production of a stable end product. Such a reaction chain also holds for the other major fission products of interest, ^{129}I, ^{90}Sr, and ^{137}Cs.

At the bottom part of the page is a comparison of the cross section for transmutation for thermal and fast neutron energy regions. At thermal energies, fission product transmutation cross sections are uniformly about an order of magnitude higher than at fast neutron energies.

An Intense Thermal Flux Provides Rapid Fission Product Transmutation

Tc^{99}



Isotope	σ (thermal)	σ (fast)
Tc99	20 barns	0.2 barns
I129	31	0.2
Sr90	1	(0.1)
Cs137	0.25	(<0.01?)

High Flux and Cross Sections Allow Significant Inventory Reduction

The combination of high neutron flux values and large cross sections means that we can achieve large transmutation rates in our system with very small resident material inventories. Specifically, cross sections for transmutation in our system are about 10 times higher than fast systems, and our flux level is 10 to 100 times higher than other thermal or fast reactor systems. The net result is that we can lower our inventory by a factor of 100 and still achieve very high transmutation rates.

High Flux and Cross Sections Allow Significant Inventory Reduction

10 times higher cross section

10 to 100 times higher flux

Transmutation Rate

$$T_R = M\ \sigma_T \Phi$$

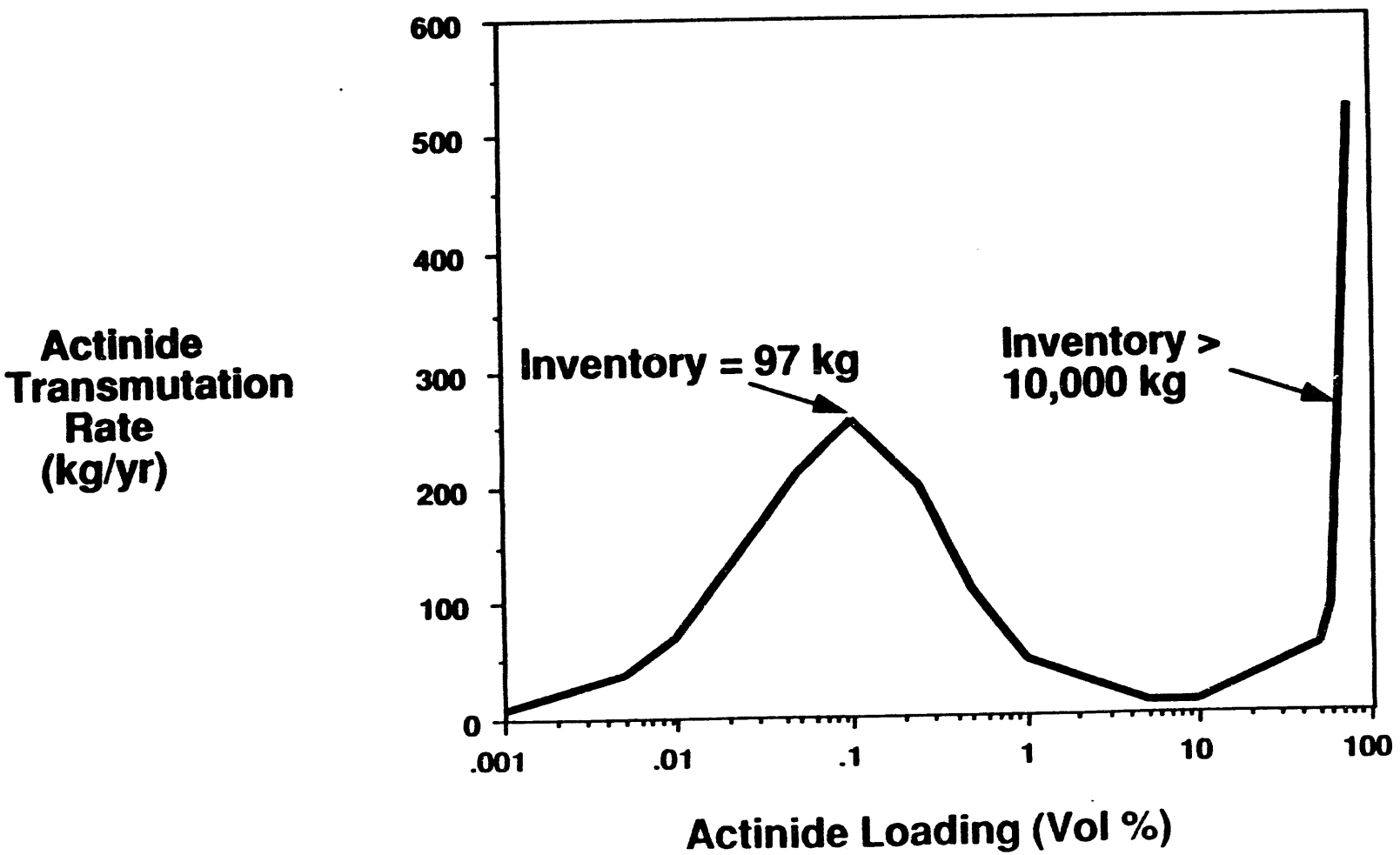
Reduction of mass inventories by > 100

A Unique, Dilute Operating Regime Characterizes Our System

The striking effect of our high flux, large cross section neutron environment is illustrated here. In a series of initial one-dimensional neutronics calculations, we were able to identify a unique operating regime that exists at very dilute loadings (by volume percent). For this particular series of calculations we achieved a peak in actinide transmutation rates at about a 0.1% loading value. For the calculated transmutation performance of 250 kg/yr of material transmuted, the required instantaneous inventory was less than 100 kilograms. In contrast, a more conventional system where loadings were greater than fifty percent by volume, would require more than 10,000 kilograms to achieve the same performance. This unique, dilute loading feature has not been recognized before, and occurs primarily because of the two-step actinide burning process described earlier and because of the high flux levels in our system.

The features of the plot can be described as follows. At very small volume percent loadings there is not enough material for very large transmutation rates to occur. As the concentration is increased to 0.1%, the amount of material becomes larger, and a significant contribution to transmutation occurs from the production of ^{238}Np and its subsequent fission. As more and more material is loaded (volume percents in the range from 1 to 10) the flux level is depressed and the two-step actinide burn process decreases. Finally, as more material is added, the transmutation rate increases again because of the dominance of the material mass term in the transmutation rate equation shown earlier.

A Unique, Dilute Operating Regime Characterizes Our System



A Dramatically Reduced Materials Inventory Environment

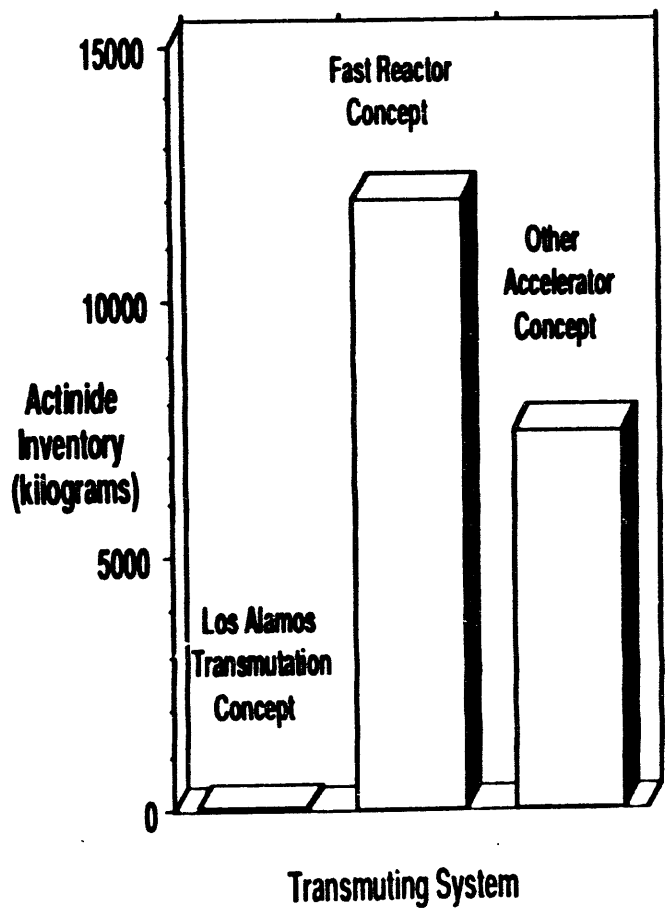
The design characteristics of our transmutation concept lead to a drastic reduction in the inventory of waste material that must be contained within the irradiation system. This follows from the system's intense neutron flux (10^{16} n/cm^2 /sec) and because of the efficiency of the thermal neutron actinide fission process described earlier. Hence a dilute system where material loadings are less than one percent of the total system volume is achieved. Our system transmutes at much higher rates than other proposed concepts and has actinide loadings of less than 100 kilograms. Other more conventional systems require as much as 10,000 kilograms of materials to achieve the same performance.

Our system is driven by an external source (the accelerator) and operates far below regions of criticality concern (our k_{eff} range is 0.2-0.8 depending upon the application). This fact, combined with the drastic reduction in actinide inventory, provides important safety enhancements. Criticality safety concerns are reduced considerably because of these design characteristics. They also should lead to reduced concerns in areas pertaining to nuclear safeguards, security, and environmental issues as compared with conventional nuclear energy systems. Such features can provide increased flexibility in meeting siting requirements for a facility based on our concept.

Finally, the reduced inventory of waste material in our system results in two important additional features. For transmutation applications much less hazardous material must be assembled in our system in order to begin efficient operation. The environmental mortgage of our system will be much smaller than other transmutation approaches since it would leave untransmuted residues of less than tens of kilograms. In contrast, other reactor or accelerator systems could leave upwards of thousands of kilograms of core material that must be encapsulated and placed in long-term storage.

A Dramatically Reduced Materials Inventory Environment

- 100 times smaller fissile material inventory than other systems
- No criticality issues ($k_{eff} \rightarrow$ 0.2 to 0.8)
- Reduced environmental legacy



The Low Material Inventory Enables New Chemistry Options

The unique high-flux, low-inventory features of our concept have striking implications on the chemistry technology needed for it. Because of high transmutation rates, continuous material flow loops are more desirable than solid form transmutation targets. There are several options for carrier media that will be discussed in the context of this concept. They are (1) aqueous slurries, (2) aqueous flows containing waste products dissolved in them, and (3) molten salts such as LiF/BeF_2 which are desirable for designs where power production is desired.

A direct result of the low inventory feature is the reduced requirement on the processing capacity of the chemical processing system. In our system, material inventories are in the range of tens of kilograms, so that the overall chemical system size is only slightly greater than laboratory scale systems. Because of the carrier media choice and the small capacities involved, advanced chemistry processing technologies can be investigated that may not be practical for larger system applications. Examples include use of ion-exchange columns in aqueous processing and use of fluorine chemistry in the case of molten salt carriers.

The Low Material Inventory Enables New Chemistry Options

- Continuous material flow
- Several carrier media options
- Small capacities required for chemical processing system (tens of kg)
- Advanced chemical separations technologies can be used

The Accelerator Transmutation of Waste System (ATW)

Several key features that comprise our concept are summarized here. The cornerstone is an advanced, high proton current linear accelerator - a result of SDI-supported and DOE-initiated research. This machine delivers a 1.6 GeV proton beam to a lead-bismuth spallation neutron production target. A range of proton currents appropriate to the specific application can be used. A heavy-water moderator surrounds the spallation target to slow the neutrons down to thermal energies. The result is a high intensity thermal neutron flux - greater than 10^{16} n/cm^2-sec. This intense neutron source - up to 100 times greater than that occurring in a thermal power reactor system - enables several exceptional features for our concept that we have previously discussed. They are summarized again here.

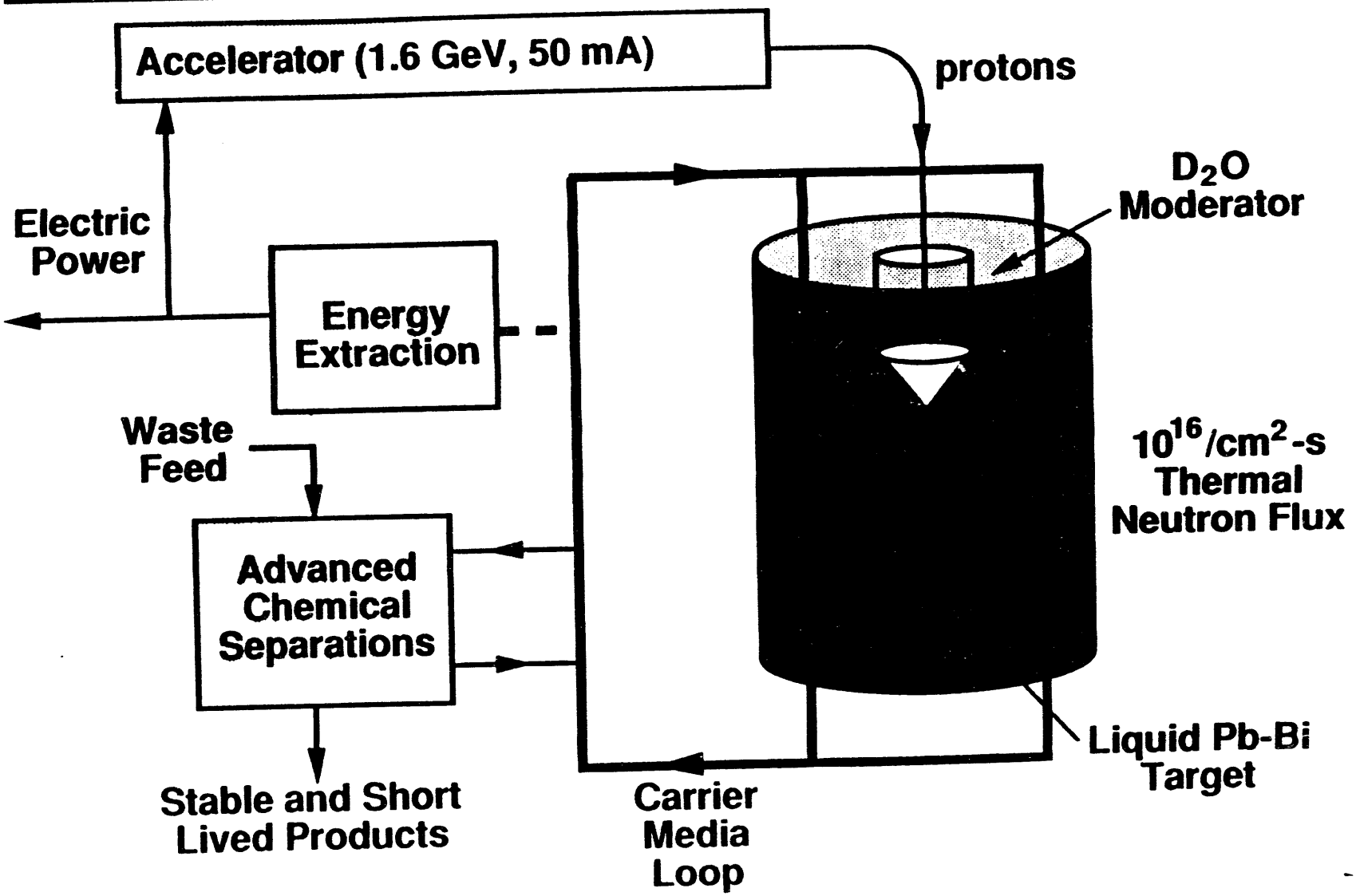
1.) The system can efficiently fission higher actinides that normally cannot be transmuted using a thermal neutron flux. Previous approaches rely exclusively upon use of fast neutrons. Our concept utilizes a completely different approach based on an intense thermal neutron source to achieve superior efficiencies in higher-actinide and fission product transmutation.

2.) There is a large reduction in resident material required for our system to perform effectively. This results not only from the high efficiency thermal fission process but also from the fact that we can effectively trade our hundredfold higher neutron flux for a similar reduction in resident inventory materials.

3.) The intense neutron flux allows design of a dilute inventory system coupled with continuous material feed. We can choose among several options for carrier materials as well as the chemical partitioning approach that will lead to highest decontamination factors.

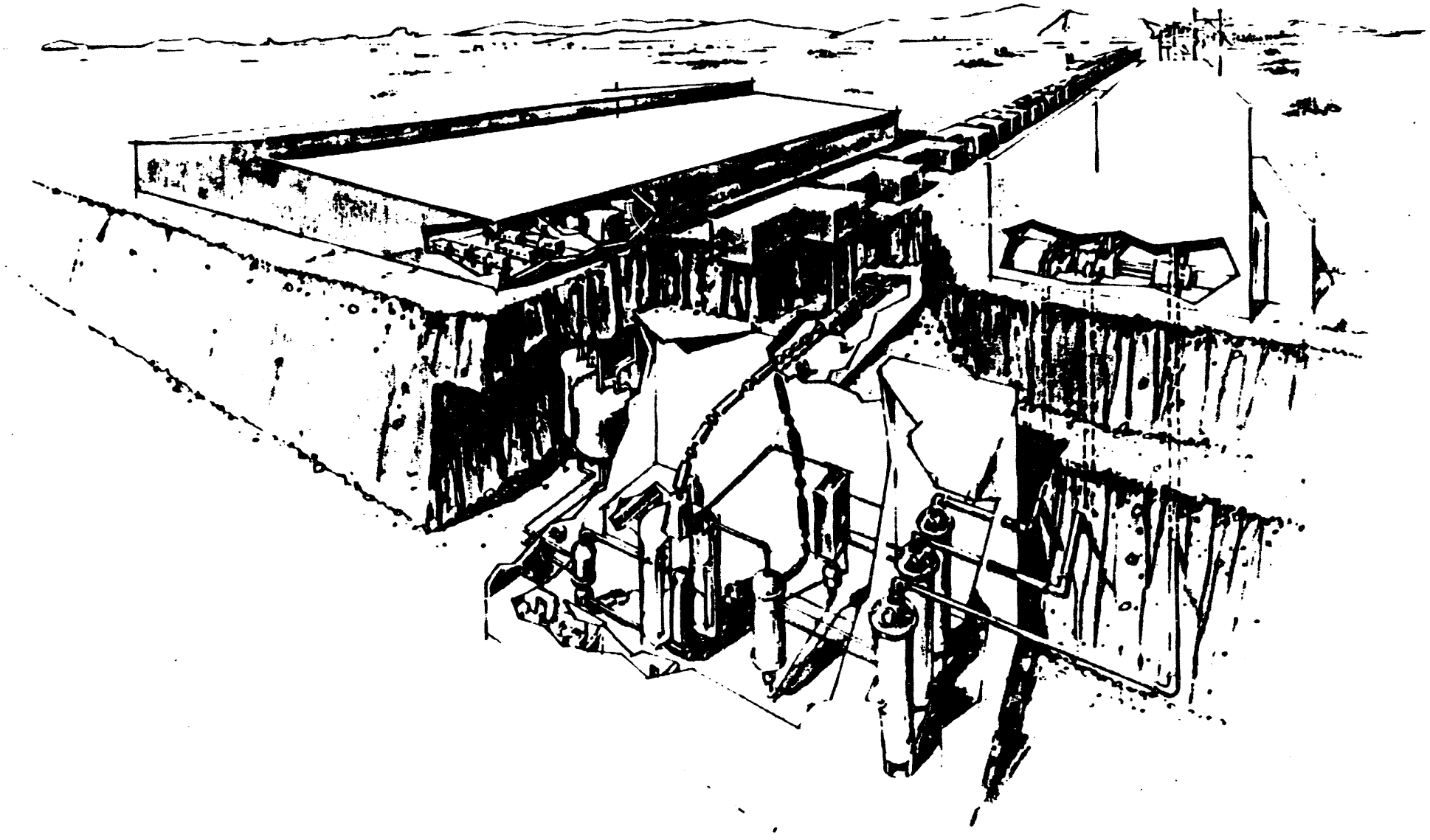
Finally, when the carrier material is a molten salt, such as lithium-beryllium-fluoride, we can extract electric power efficiently because of the salt's high thermal-to-electric conversion capabilities. Its choice also enables a new spectrum of advanced fluoride chemical separations to be utilized.

The Accelerator Transmutation of Waste System (ATW)



An Artist's Rendering of the ATW System

An artist's rendering of the ATW system shows the scale and main components. Components dealing with the accelerator, target blanket, chemistry processing, and power production are shown. For the near-term application, the complexity of the chemical processing requirements is reduced and the power production requirement is gone.



Transmutation Concepts

Several concepts for transmutation have been proposed during the recent past. Here we compare the features of our concept to them. Specifically, the concepts that are shown are the Integrated Fast Reactor (IFR) concept from Argonne, the Phoenix concept from Brookhaven, and the Clean Use Of Reactor Energy (CURE) concept from Hanford.

The IFR is based upon a fast reactor system, producing from about 0.4 to 1.4 gigawatts electric while burning actinide waste. Required inventories for the system extend to over 10,000 kilograms. A chemical partitioning approach based on pyrochemical methods would be used to separate actinides to be burned from spent fuel elements. The system is aimed solely at transmutation of commercial power reactor waste.

The Phoenix concept uses a 100 mA, 1600 MeV accelerator driving a multiplying assembly operating at a k_{eff} of 0.9. This system is aimed at transmuting higher actinide waste and would require target inventories up to 24,000 kilograms. This system uses the organically-based PUREX and TRUEX aqueous processing technologies for extraction and separation of higher actinides from spent fuel elements. This system is aimed solely at commercial waste applications.

The CURE concept would use cleanup fast reactors (CFRs) operating in the range of 400 MW_E to transmute both actinide and long-lived fission product wastes. Versions of CURE exist for both defense and commercial waste arenas. It too uses PUREX and TRUEX chemistry processing and partitioning technologies.

Our ATW concept transmutes both fission products and higher actinide waste components. It is aimed primarily at defense wastes. Because of its high flux environment, the ratio of material transmuted per year to instantaneous inventory is greater than 1.5 to 1. It uses advanced aqueous chemical processing techniques.

Transmutation Concepts

	ATW	IFR	Phoenix	CURE
Actinides	Y	Y	Y	Y
Fission Products	Y	N	N	Y
Waste Type	Defense	Commercial	Commercial	Defense Commercial
Fraction Burn: Inventory	>1.5:1	<0.05:1	<0.1:1	<0.05:1
Keff	0. - 0.2	1.	0.95	1.
Chemistry	Aqueous	Pyro	PUREX, TRUEX	PUREX, TRUEX

Summary

Our ATW concept utilizes a new approach to transmutation. Several apparent advances and advantages have arisen from our investigations up to now. We are focusing on two applications - a near term one aimed at defense wastes, and an advanced concept aimed at fission energy with a minimal long-lived, high level waste stream.

Summary

- Our ATW concept is a new approach to transmutation that employs an intense thermal neutron source

- The intense thermal neutron source enables
  - Efficient actinide and fission product burn
  - Small material inventories
  - Options for advanced chemical processing

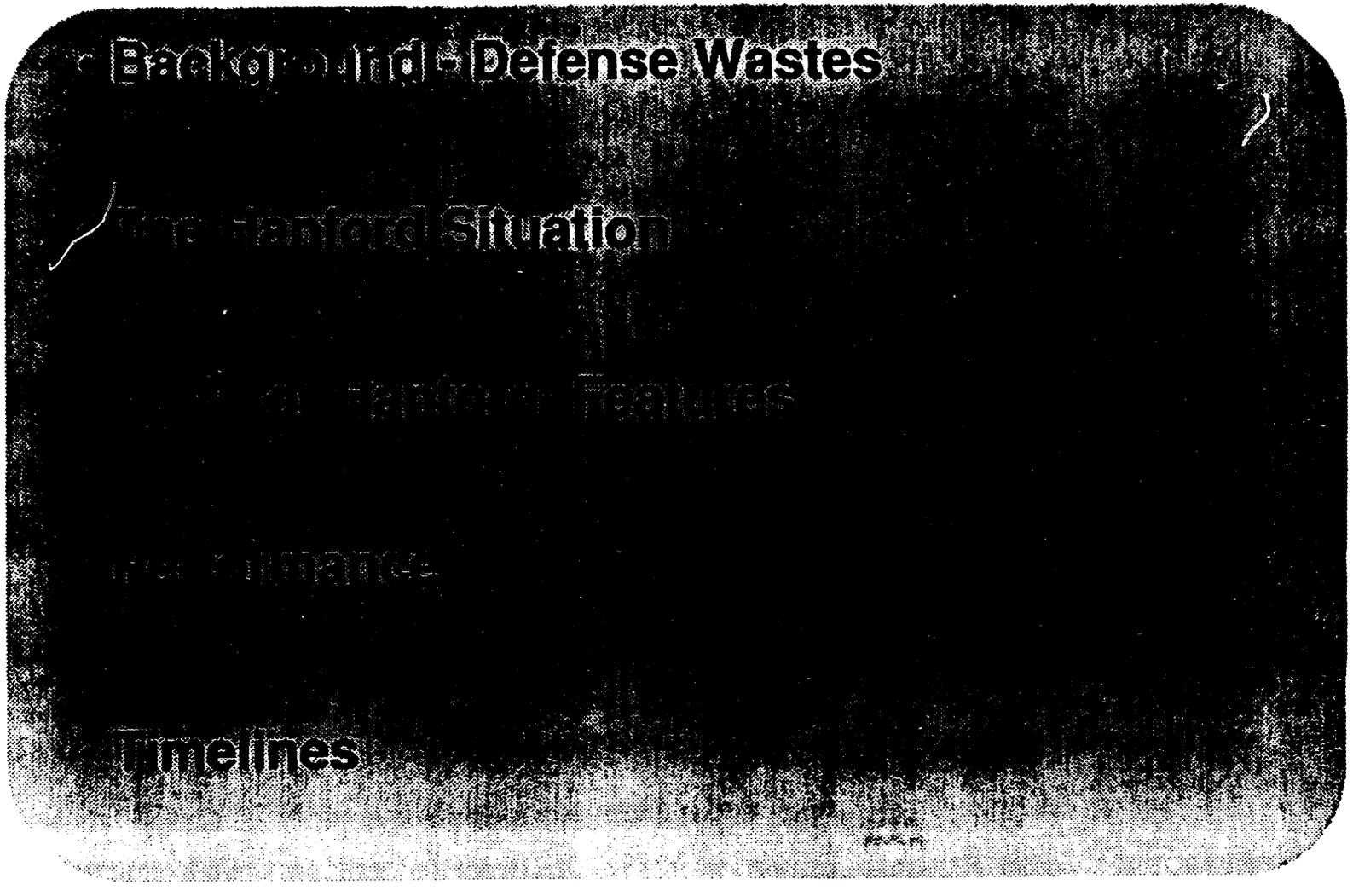
- We are focusing two applications
  - Near term - Defense wastes
  - Advanced - Fission energy with a minimal long-term HLW stream

OVERVIEW

Near Term Application of ATW
High-Level Defense Wastes
Hanford

Edward Arthur
Theoretical Division

Contents



Defense High-Level Wastes are Mainly Fission Products

The total inventory of long-lived, high-level DOE defense wastes present at Hanford, Savannah River, and Idaho is about eighteen metric tons, most of which are fission products (^{99}Tc and ^{129}I). This figure does not include ^{90}Sr and $^{135,137}Cs$ which account for another 6 to 8 metric tons.

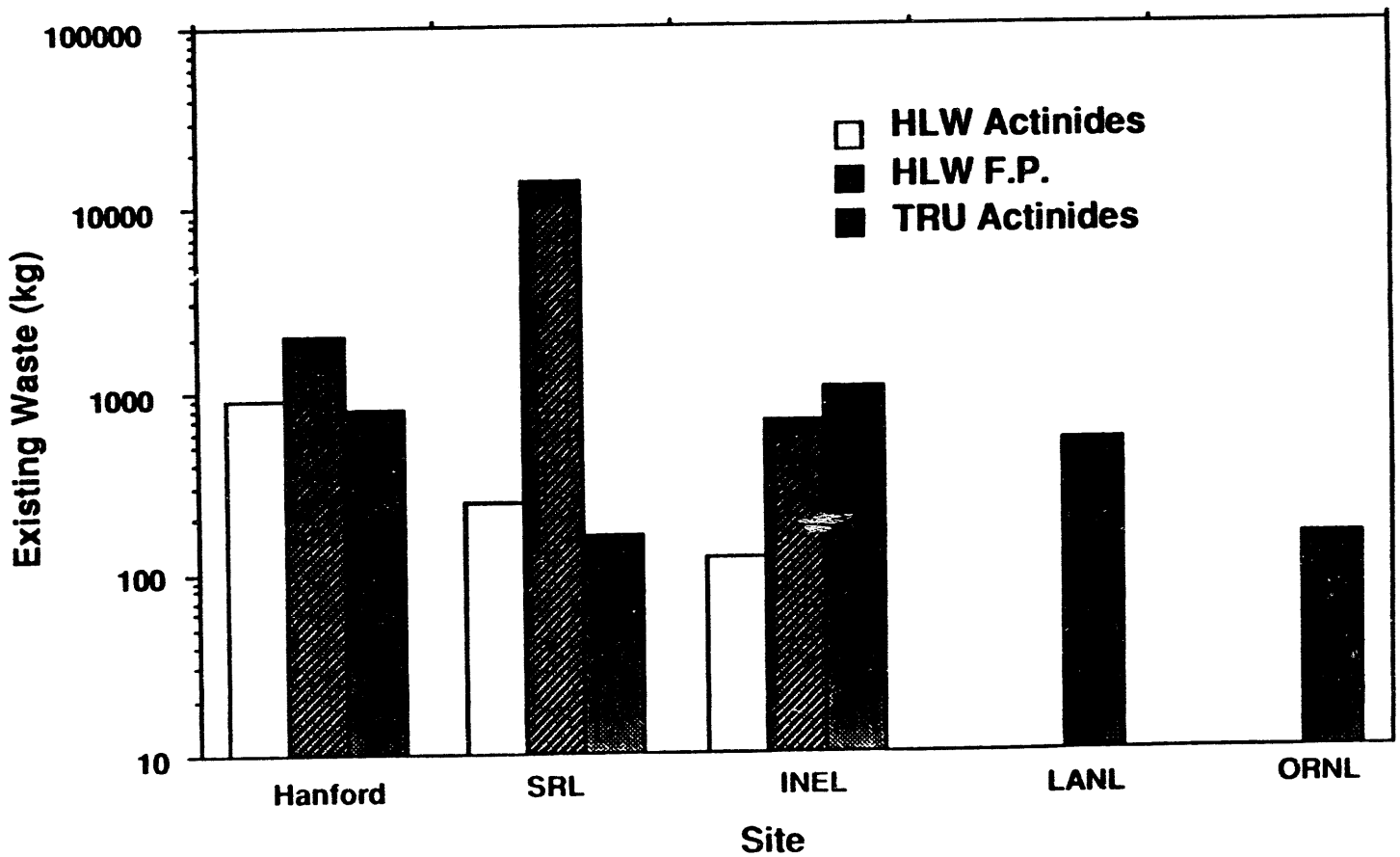
Defense High-Level Wastes Are Mainly Fission Products

Inventory Summary	Tonnes
Actinides (Np, Am)	1.3
Fission Products (^{99}Tc, ^{129}I)	16.8
TOTAL	18.1

Defense Waste Inventories

The distribution of defense waste as a function of DOE site is illustrated. Most of the HLW resides at Hanford and Savannah River. In addition, TRU waste exists at Los Alamos and at Oak Ridge.

Defense Waste Inventories



Two Specification Criteria Can be Utilized for the ATW Defense Waste Application

There are two environments in which performance of a partitioning/transmutation system can be assessed. The first assumes that the fission products ^{99}Tc and ^{129}I are the major drivers because of their long half lives and significantly larger chemical mobilities, as compared to actinides.

The second environment addresses wastes from a toxicity perspective so that an overall reduction in the toxicity of the wastes is the relevant measure. In this scenario, actinides contribute mainly to toxicity levels.

Since the ATW System transmutes both fission products and actinides, it can meet criteria associated with both of these problem-definition environments.

Two Specification Criteria Can Be Utilized for the ATW Defense Waste Application

1.) Tc, I are major long-term storage drivers

or

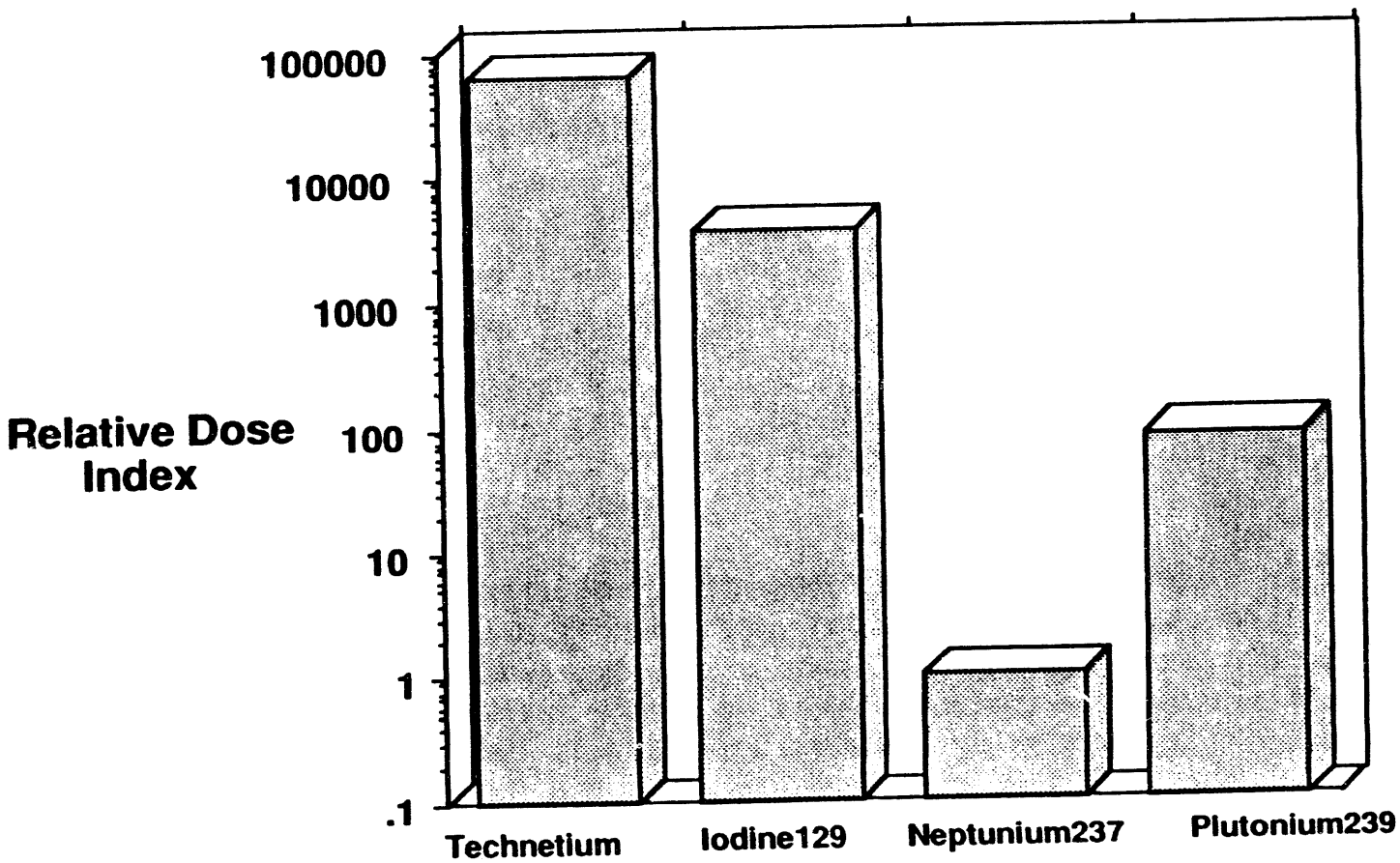
2.) Overall reduction in toxicity of HLW is the relevant measure

The ATW system transmutes <u>both</u> fission products and actinides and therefore could meet 1) or 2)

Technetium Dominates Long-Term Risk in a Geologic Repository

This comparison illustrates the importance of technetium and iodine in assessing the long-term risk associated with repository storage. The relative dose index is determined from a product of the amount of the waste present, its half life, and its relative mobility. The mobility of technetium and iodine is significantly greater than that of actinides such as neptunium and plutonium. Thus, their relative dose indices are orders of magnitude greater.

Technetium Dominates Long-Term Risk in a Geologic Repository



Overall Toxicity Measure Approach

In the toxicity measure approach, the goal of partitioning/transmutation is the reduction of the toxicity of the final waste form to a level below that of the uranium burned to create the waste. In the context of defense wastes, the present HLW toxicity is about 3 x$10^{11}$$m^3$ H_2O (a measure of the amount of water needed to dilute the waste to levels that will meet disposal purity requirements). The toxicity of the uranium consumed to produce tritium, plutonium, and naval fuels is 3 x $10^7$$m^3$ H_2O. Thus, a decontamination factor of about 10^4 is suggested by use of this criteria.

Overall Toxicity Measure Approach

Through partitioning/transmutation reduce toxicity to less than that of original uranium

For Defense Wastes

Present HLW Toxicity	**3×10^{11} m^3 H_2O**
Toxicity of uranium consumed in Pu and T production, naval fuels	**3×10^{7} m^3 H_2O**

The Defense Waste Application Produces these Technology Requirements

Application of either of these criteria produce several technology requirements. The system must be able to transmute both long-term fission products and actinides. It must achieve high efficiencies in chemical separation (1 part in 100 to 10,000). Waste streams must be minimal and understood. Finally the technology used should be based on reasonable extrapolation of current technologies so that operating systems can be ready on timescales relevant to defense waste cleanup.

The Defense Waste Application Produces These Technology Requirements

- Transmute long-term fission products and higher actinides

  (Defense waste mix - 90% F.P. ; 10% higher actinides)

- Achieve high-efficiency factors in chemical separations

- Waste streams must be minimal and understood

- Reasonable extrapolation of present capabilities

The Hanford HLW Environment

We now turn to the specific environment of the proposed near-term ATW application, which is the defense waste problem at Hanford. There large volumes of radioactive waste is stored in double-shell and single-shell tanks. The double-shell tanks contain mostly liquid, which in turn contains most of the technetium estimated to be in waste there. The single-shell tanks contain mostly sludge and saltcake where most actinide waste components should reside. The estimated inventory of fission product and higher actinide waste is about 2000 kg and 400 kg respectively.

The Hanford HLW Environment

- **Double-Shell Tanks** — 47590 m^3

  Contain mostly liquid
  Contain majority of Tc

- **Single-Shell Tanks** — 140,100 m^3

  Sludge — 47,700 m^3
  Saltcake — 92,400 m^3

~ 2000 kg Tc, I
~ 400 kg Np, Am

A System for Hanford Application

In order to produce a reference point design for Hanford, a 30-year operating scenario was assumed. To meet this schedule requires that between 60 and 70 kg of technetium be transmuted per year. Similarly about 10-15 kg of actinides would be transmuted. An initial estimate of the accelerator requirements is a 1600 MeV, 50-60 mA machine consuming about 200 MW_E. The chemistry processing system would be aqueous-based and would require a capacity of 50 kg for technetium separation loops and less than 10 kg for actinides.

A System for Hanford Application

- 30-year operation scenario
- Requires 66 kg Tc,I transmuted/yr and 10-15 kg/yr of Np
- Translates into 50-60 mA accelerator @ 1600 MeV ($200\ MW_E$)
- Chemical processing system mass inventory requirements - 50 kg Tc, <10 kg actinide

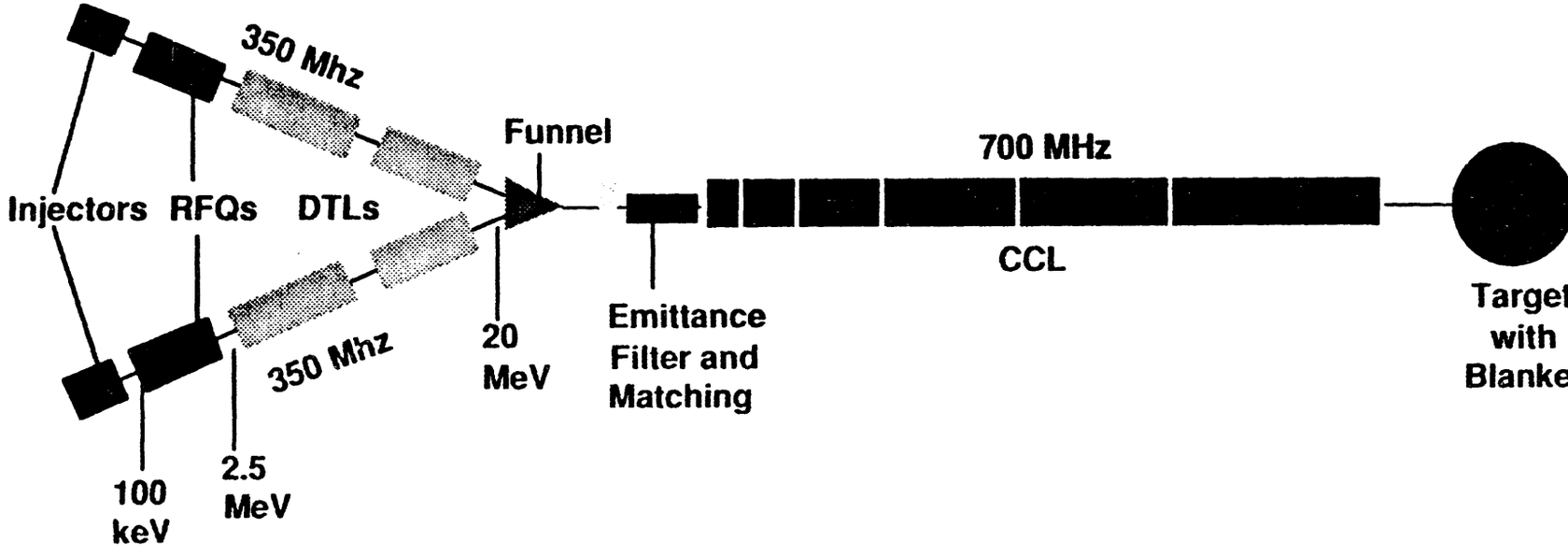
Accelerator Requirements for the Hanford Application

The transmutation accelerator is a high-power, radio-frequency, continuous beam linear accelerator (linac) that generates a 1.6-GeV proton beam with an average current of 50 to 60 mA. The configuration shown here consists of a 700-MHz coupled-cavity linac (CCL) injected at 20 MeV by a funneled beam-launcher. The latter is made up of two 100-keV injectors each containing 140-mA proton sources, two radio-frequency quadrupole linacs (RFQs), and two drift-tube linacs (DTLs). A funnel similar to that tested recently at Los Alamos is used to combine the two beams which are then accelerated to 1.6 GeV in a coupled-cavity linac. The CCL then makes up most of the accelerator.

This design takes full advantage of the considerable progress in high current linear accelerator technology that has occurred during the past decade, initiated by DOE support and advanced by the SDI Neutral Particle Beam program. These advances allow sizable improvements in the generation, acceleration, and handling of low-energy beams within the accelerator structure. Optimization of the low-energy performance is key to the successful creation of a reliable, high-current, high-energy accelerator system.

The accelerator front end is optimized to prepare a high-current, low-emittance, well-controlled beam. The CCL parameters are chosen to assure extremely low beam loss while maintaining a high conversion efficiency of radio-frequency power to beam power. We also select a very large ratio of the accelerating-structure aperture to beam diameter to assure low enough beam loss for "hands-on" maintenance. Beam performance has been confirmed by sophisticated multi-particle computer simulations. The overall accelerator design concept was reviewed earlier this year by a DOE Energy Research Advisory Board panel, which evaluated it as technically sound with no physics uncertainties.

Accelerator Requirements for the Hanford Application

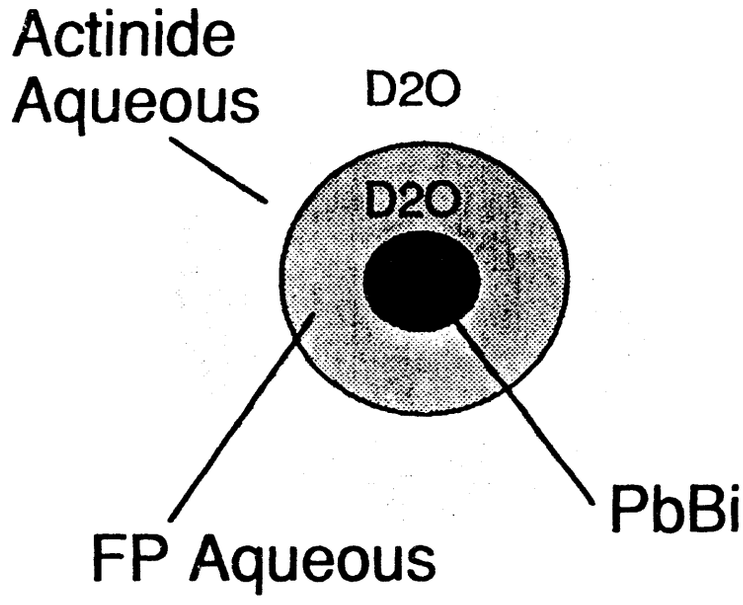


- 1600 MeV
- 50 to 60 mA
- 200 MW_E
- 75% availability

Blanket Configuration

The target/blanket configuration is illustrated schematically. In the center is a PbBi neutron production target. Surrounding this is a heavy water moderator region. The moderator would be divided into two regions, an inner one in which a slurry would contain fission products such as technetium. In the outer region a slurry would contain actinides in an oxide form.

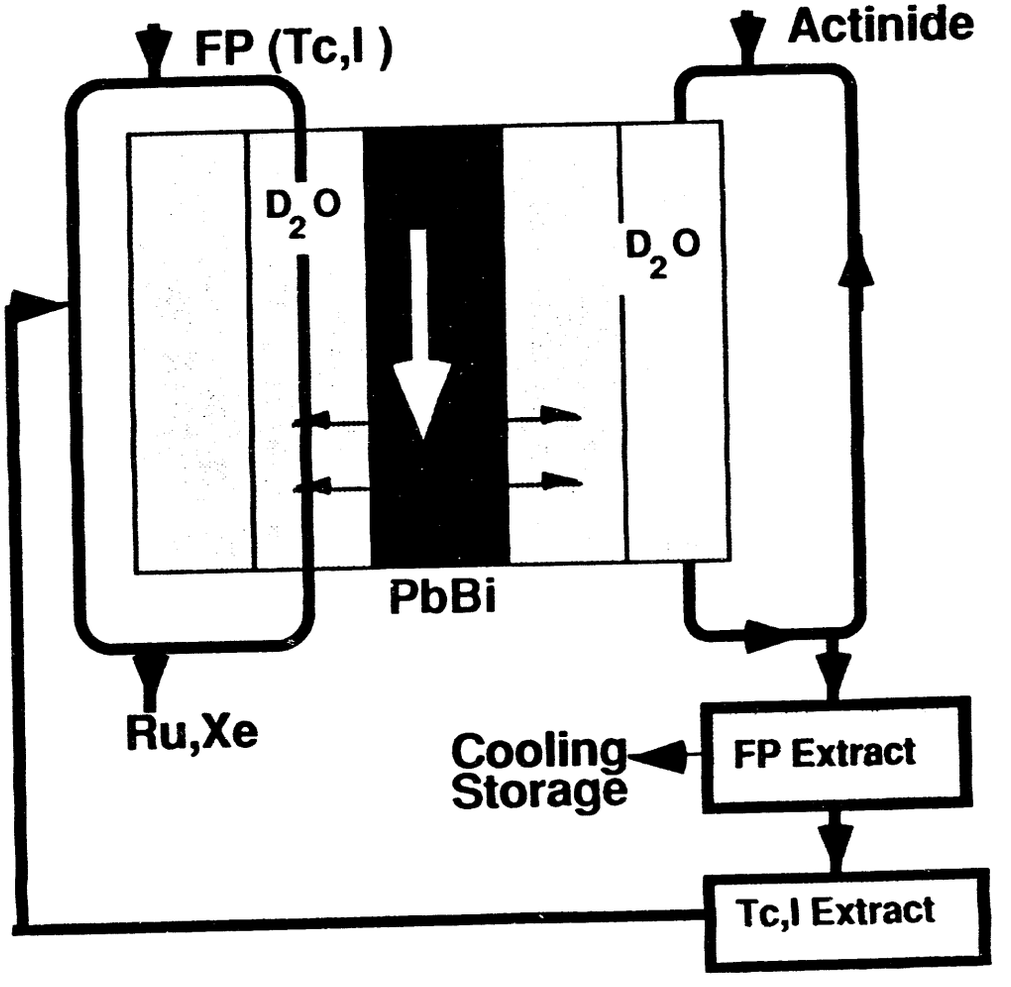
Blanket Configuration



Aqueous Processing Loops

Two aqueous loops are required for the near-term application. An actinide loop contains an actinide such as neptunium which is fissioned. As the slurry containing the actinide flows through the transmuter, a small slipstream is used to extract fission products produced during transmutation. From this fission product lump, most are sent to short-term cooling and storage. The long-lived technetium and iodine are separated from this lump and are introduced into the fission product loop shown at left. This loop also has a feed, fission products arising from direct tank waste extraction. In the aqueous loops shown, technetium is transmuted to produce ruthenium while iodine produces xenon. The ruthenium and xenon are then separated from the loop for final disposal.

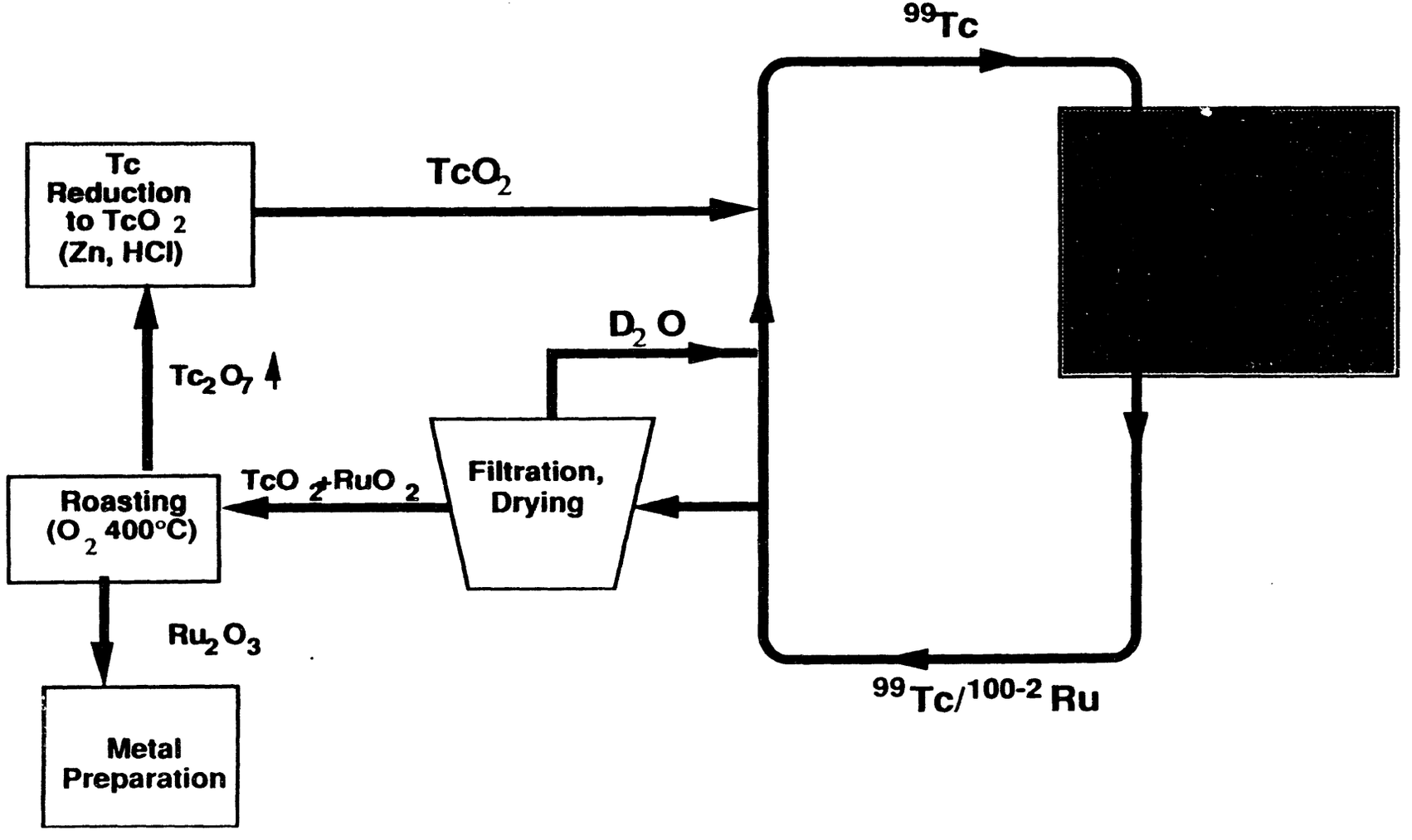
Aqueous Processing Loops



Straightforward Chemistry Concept for Tc/Ru Separation

A particular example of a processing loop appropriate for the near-term system is illustrated. Here technetium is introduced into the transmuter, and after transmutation occurs, ruthenium is produced. To separate the two components, the mixture is oxidized and roasted. The compound Tc_2O_7 comes off in a volatile form and is reduced back into a form to be reintroduced into the system. The ruthenium precipitates out as Ru_2O_3 and is prepared into metal for disposal.

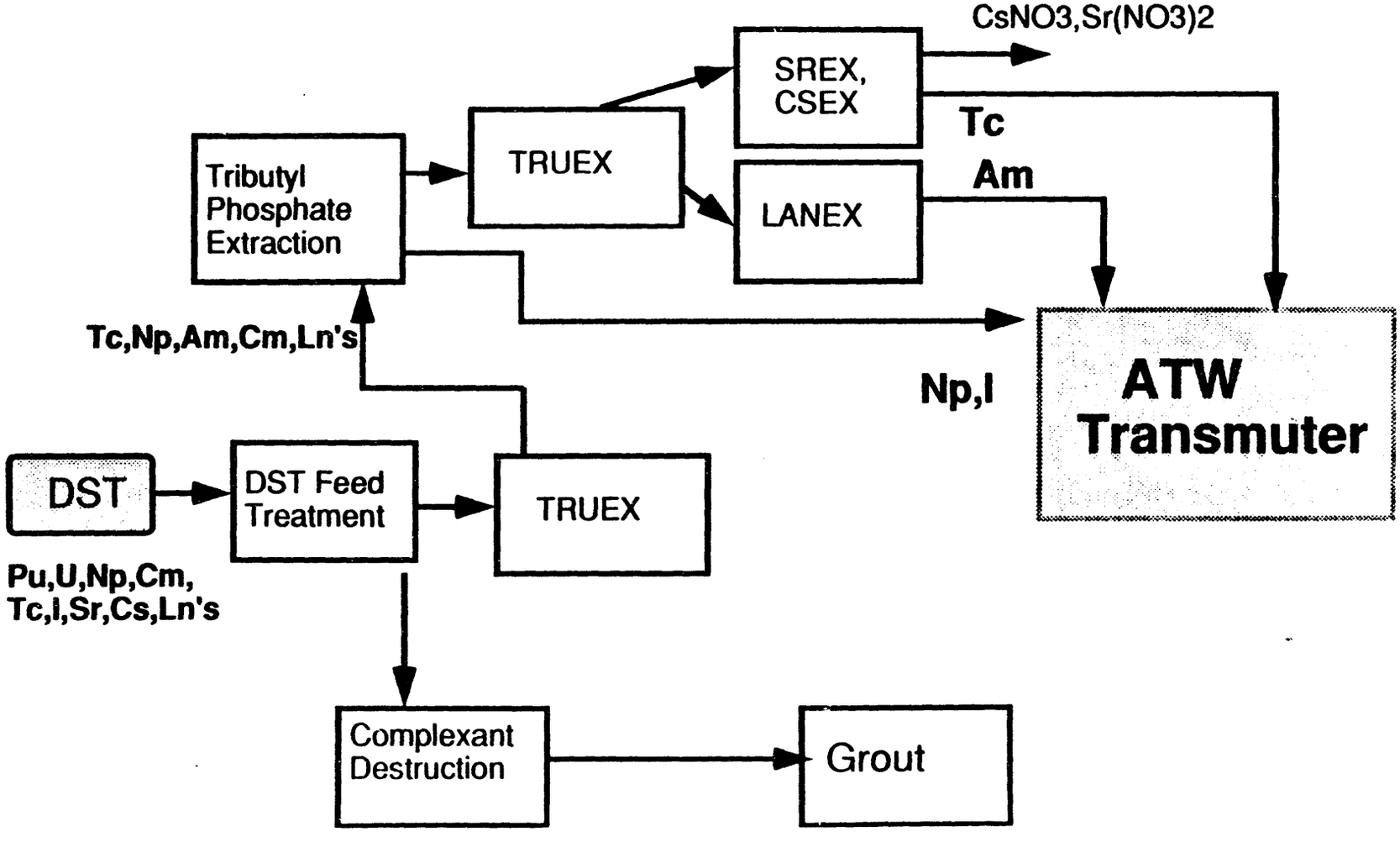
Straightforward Chemistry Concept for Tc/Ru Separation



Possible Hanford Chemical Partitioning is Consistent with ATW Requirements

A system for partitioning of tank waste contents has been proposed as an alternative means for waste cleanup. In it an organically-based extraction process such as TRUEX or LANEX is used to separate the actinides and fission products shown. Under this system these waste constituents could be fed directly into the ATW transmuter.

Possible Hanford Chemical Partitioning Is Consistent with ATW Requirements

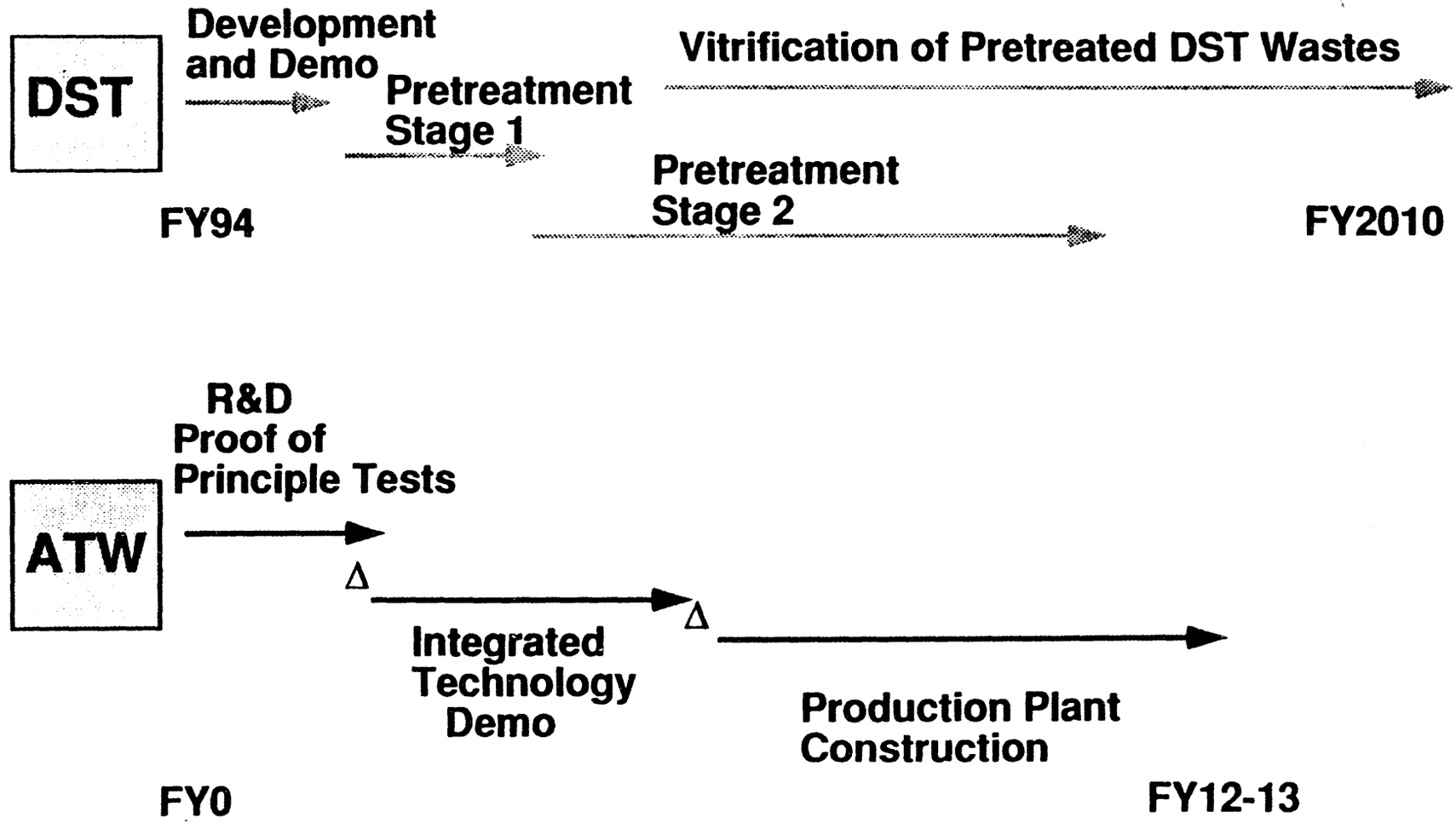


ATW Timelines are Consistent with Possible Hanford Cleanup Scenarios

We believe the timescales necessary to develop the ATW system are consistent with schedules associated with Hanford cleanup. At the top part of the figure we show timelines associated with a Hanford proposal that involves a significant degree of chemical partitioning of wastes. The timescales are also consistent with current plans for Hanford cleanup.

The ATW system can be developed and made operational in a similar timeframe. The first part of the development process would be several years aimed at R&D. At this point a decision about stopping or proceeding can be made. This is followed by a period of integrated technology demonstrations or proof of principle experiments. Finally, if previous results were successful, a production plant could be built in a period of 6 to 7 years. Thus we envision, from a technology perspective, a time to plant availability of about 12 years.

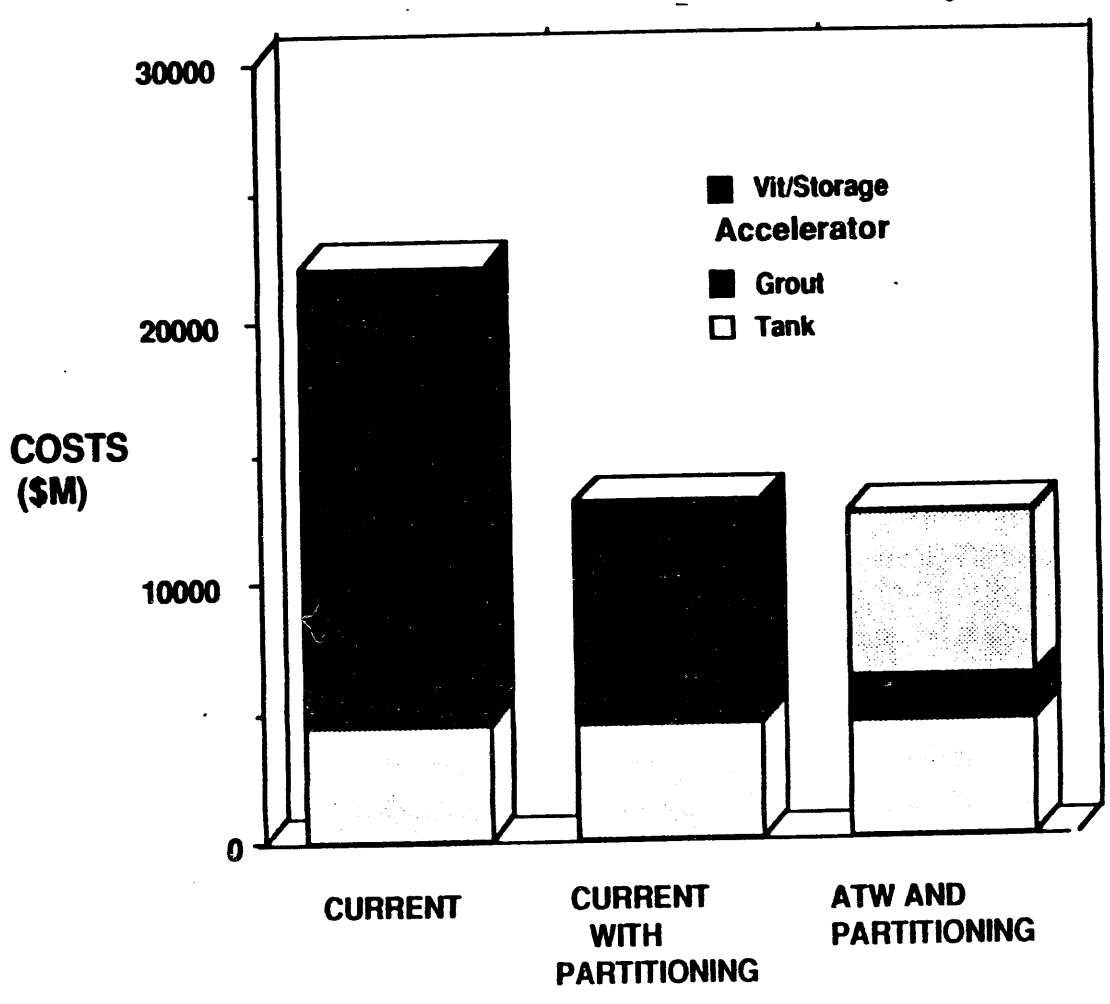
ATW Timelines Are Consistent with Possible Hanford Cleanup Scenarios



Preliminary Cost Estimates are Comparable with Other Alternatives

We have made a very preliminary estimate of costs that may be associated with an ATW application at Hanford. These estimates are for scoping purposes only and illustrate that our total costs may be cheaper than those associated with present Hanford scenarios and comparable to those involving partitioning followed by geologic storage. In our scenario, storage would be accommodated using on-site facilities.

Preliminary Cost Estimates Are Comparable with Other Alternatives



Preliminary Results: Long-Lived Isotope Inventories After 30 Years Operation

We have made an initial, and preliminary assessment, of the long-lived species that might be produced during the period of operation of a transmuter at Hanford. During a 30 year period we transmute 2400 kg of long-lived species. According to our initial calculations we produce about 60 kg of a relatively few long-lived species in the PbBi target as well as stainless steel components in the moderator tank.

We have begun to develop strategies to lower this figure further. One straightforward method is the reduction of Pb in the system. We believe the present design mass can be decreased by a factor of two. Likewise we believe a target could be made which is comprised solely of lead. In this manner, contributions from long-lived bismuth species could be eliminated. Finally alternatives such as depleted uranium may be used in a supplemental fashion to further reduce the lead mass.

Preliminary Results: Long-Lived Isotope Inventories After 30 Years Operation

HLW Components Transmuted

Np237	Tc99	I129
2×10^6 y	2×10^5 y	1.6×10^7 y

(2400 kg)

Long-Lived PbBi and SS Components Produced

Bi210	Bi208	Pb205	Mn53
3×10^6 y	4×10^5 y	1.5×10^7 y	3.7×10^6 y

60 kg

Further Reduction of Long-Lived Species

1.) Reduce PbBi volume by 2 → 32 kg

2.) Eliminate Bi → 19 kg

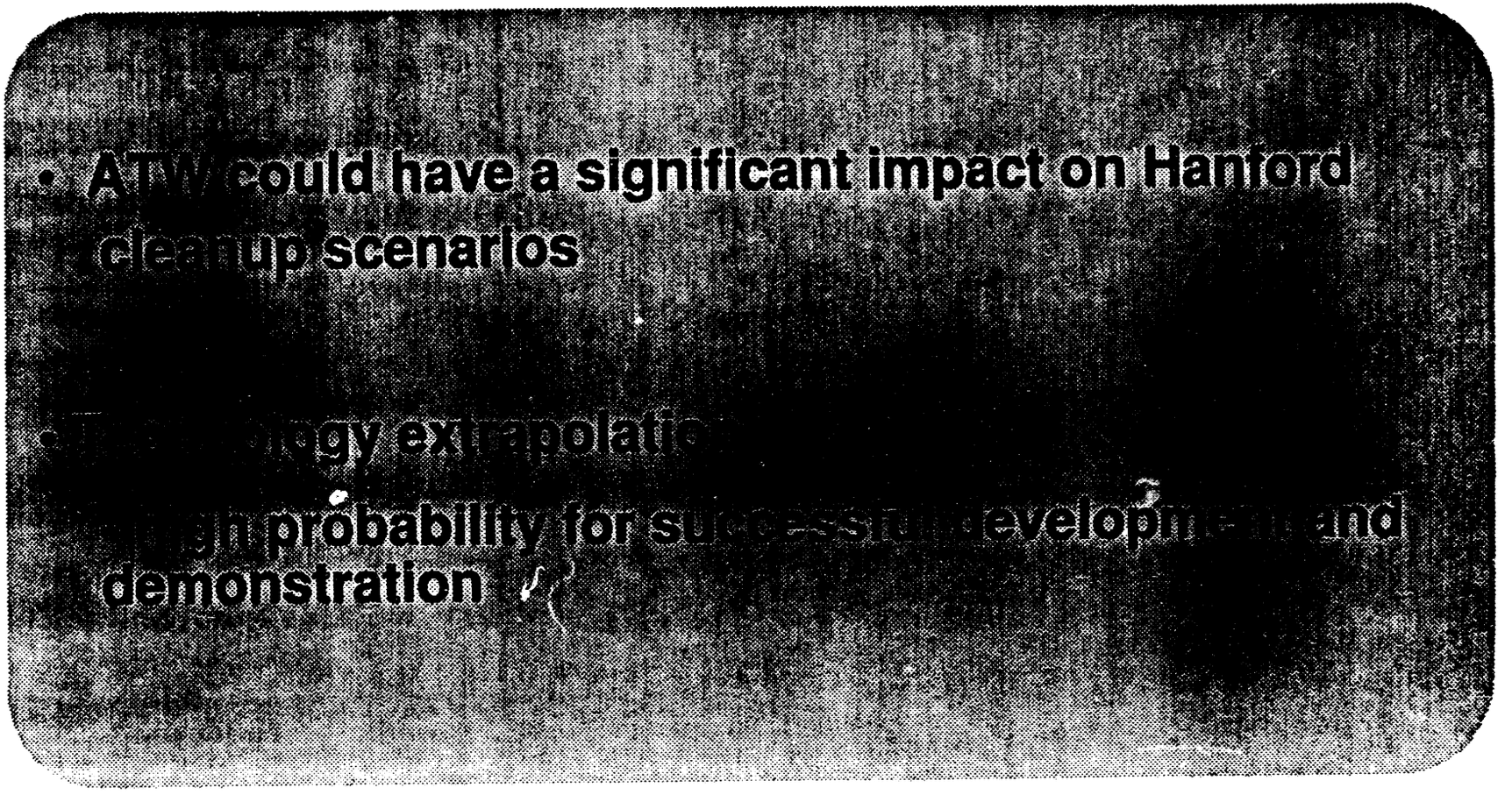
3.) Supplemental use of alternative materials → ?

Summary

In summary, an ATW system could have a significant impact on Hanford scenarios. It would provide a low inventory environment that is highly effective for transmutation. An ATW system can also provide a potential for on-site storage of waste with very small residuals left after transmutation is complete.

The system envisioned for Hanford is a straightforward extrapolation of several current technology areas at Los Alamos. Thus, we believe there is a very high probability for successful development and demonstration.

Summary


• ATW could have a significant impact on Hanford
cleanup scenarios
demonstration

LINAC TECHNOLOGY

Accelerator Technology for the ATW System

George Lawrence
Accelerator Technology Division

Outline

- History – high power linac concept – APT
- Accelerator technology advances
- APT linac conceptual design
- Present technology base
- Technical issues
- ERAB Panel evaluation
- Application to ATW requirements
- Research and development needs

High-Power Linac Concept Origins and Application to ATW Requirements

The design concept for a high-power proton linac suitable for waste transmutation applications originated as part of a 1989 Los Alamos/Brookhaven study that examined the feasibility of accelerator production of tritium (APT). Annual tritium production requirements established the reference beam parameters for the accelerator as 1.6 GeV proton energy, average current of 250 mA, and >75% availability. A Los Alamos team worked out a detailed pre-conceptual design for a high-power proton linac and beam transport system that would meet these performance requirements, while Brookhaven developed a spallation-target/production-matrix concept. It was clear from the beginning that an RF linac is the only practical machine that could satisfy such a high current requirement.

The APT study was given a thorough review by the D[illegible]nergy Research Advisory Board (ERAB) in the fall of 1989. An important finding included in the ERAB Report to the Secretary of Energy (February, 1990) was that the accelerator concept was evaluated as technically sound.

Near-term nuclear waste transmutation applications presently visualized at Los Alamos generally call for considerably lower beam current specifications than the 250 mA used in the APT design. A transmuter that could handle the technetium in the accumulated defense wastes would require about 55 mA average current. An advanced concept being considered for production of fission energy in an accelerator-driven fertile-to-fissile fuel converter (with no high-level waste stream) may require still lower current levels (30 mA).

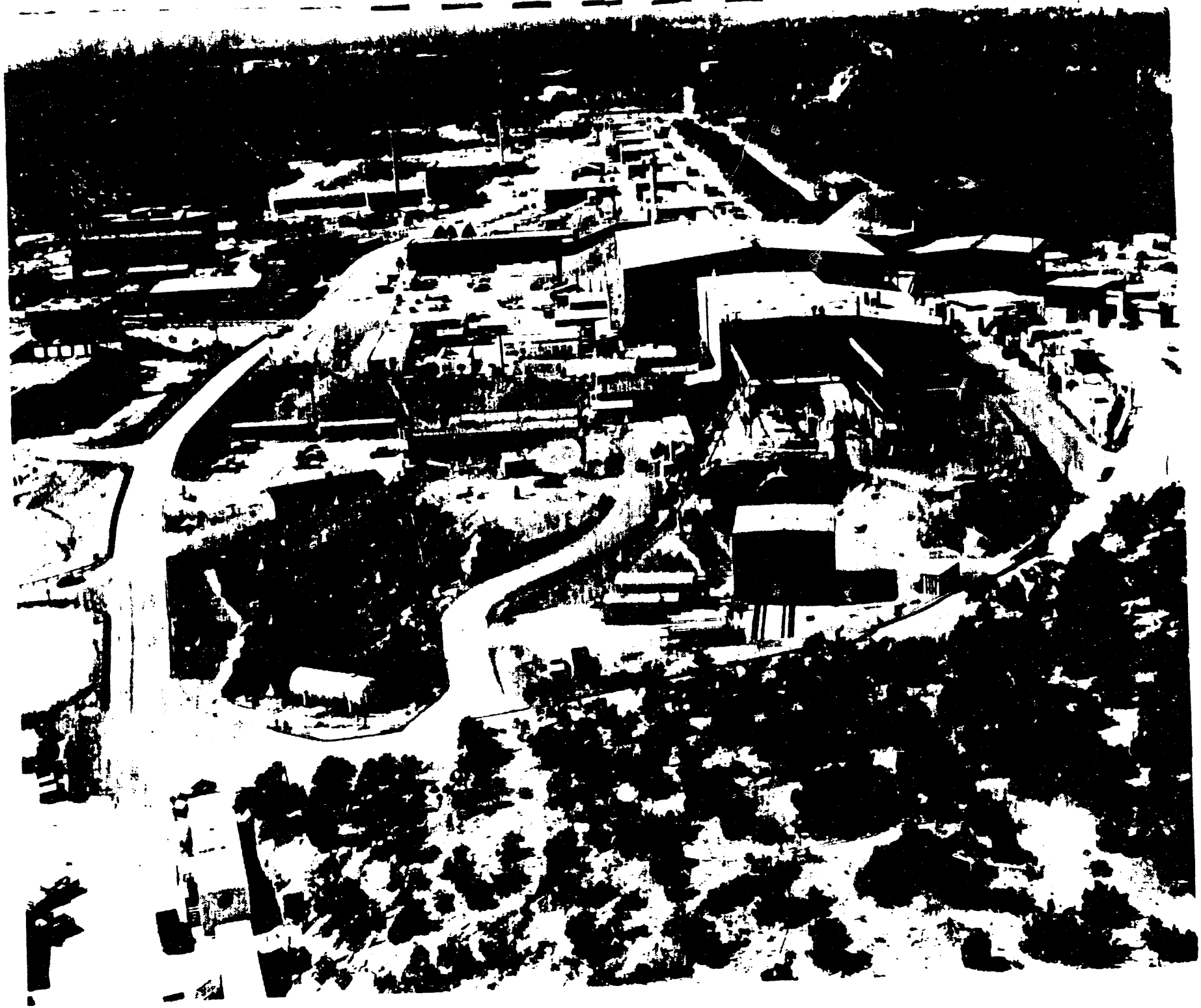
In the following presentation the 250-mA/1.6 GeV linac design worked out for APT is described along with the technology base supporting that concept. The presentation concludes with how this design might be applied to the less demanding requirements of an ATW.

High-Power Linac Concept was Developed for Tritium Production Proposal (APT)

- **APT accelerator specifications**
  - 1.6 GeV protons
  - 250 mA cw
  - 75% availability
- **1989 APT conceptual design developed to meet these requirements**
- **Conceptual design was reviewed by ERAB Panel (9/89 - 2/90)**
- **Transmutation schemes generally call for lower beam currents**
  - Initial defense-waste application (Tc burner) requires 55 mA
  - Energy production without a high-level waste stream requires 30 mA
- **RF linear accelerator is the only practical machine approach**

ATW Facility Would be Similar in Scale to LAMPF

The photograph shows the kilometer-long Los Alamos Meson Physics Facility (LAMPF), looking back along the linear accelerator from the experimental areas. The proton linac is in a tunnel covered by an earth berm. The surface buildings house the RF power sources for the accelerator, as well as magnet power supplies and control stations. The overall scale of LAMPF and its experimental areas provides a rough idea (within a factor of 2) of what an accelerator-driven waste transmuter might look like. One major difference is that the ATW spallation target and transmutation blanket (with supporting systems) would be buried 30 to 40 meters underground, well below the level of the accelerator.



Accelerator-Driven Nuclear Process Concepts Have a 40-Year History

The idea of using a high-power proton (or deuteron) accelerator for nuclear process applications goes back 40 years. Groups at several laboratories worldwide have (at different times) proposed accelerator production of plutonium for nuclear weapons, conversion of fertile material to fissile fuel for commercial reactors, regeneration of reactor fuel rods, tritium production, and transmutation of nuclear waste.

In the late '40s and early '50s the Lawrence Livermore National Laboratory proposed to build a very high power deuteron linac (500 MeV, 320 mA cw, 50 MHz) to produce plutonium from U238. This machine, referred to as the Materials Test Accelerator (MTA), was extremely ambitious for its time. A low-energy 12-MHz prototype accelerated a 225 mA proton beam to 12 MeV for short periods, and operated cw at 50 mA.

Canadian interest in electronuclear fuel breeding has been long standing. In the early '60s, a group at Chalk River Nuclear Laboratories (CRNL) proposed a 65-mA, 1-GeV spallation neutron source for materials research and fuel-breeding studies. In the late '70s and early '80s, there was serious interest in building a demonstration fuel-breeding facility, and a staged high-intensity linac development program (ZEBRA) was initiated at CRNL.

Brookhaven National Laboratory (BNL) developed several concepts during the late '70s for accelerator-driven reactor fuel element regeneration, power production, and nuclear waste transmutation. The latter idea, as in all previous ATW concepts, invoked the use of a *fast* neutron spectrum in the conversion blanket.

The Russians have proposed various concepts for accelerator-driven fuel breeding and transmutation over the past 20 years, with much of the activity concentrated at the Moscow Institute for Theoretical and Experimental Physics (ITEP).

The Accelerator Approach is Not a New Idea

- **Materials Test Accelerator (LLNL, 1949 - 1954, Pu production)**
  - Proposed: 500-MeV, 320-mA, 50-MHz cw deuteron linac; NaK-cooled Be target with U238 multiplier
  - Prototype: 12-MeV, 12-MHz DTL; 50 to 225 mA (protons)
  - Cancelled after uranium discoveries in US southwest

- **Canadian (CRNL) interest in 1960's (ING); early 1980's (ZEBRA)**
  - Fuel breeding (U233 and Pu239 for CANDU reactors)

- **BNL concepts developed in late 1970's**
  - Fuel element regeneration & power production; transmutation

- **Soviet concepts (ITEP)**

Materials Testing Accelerator (MTA)

Nearly 40 years ago, an ambitious development program was begun at the Livermore Research Laboratory (LRL) for electronuclear breeding of nuclear material (plutonium) using a high-power linear accelerator. A prototype drift tube linac (similar in purpose to that in the second accelerating stage of APT) was completed in early 1952. This DTL, known as the Mark 1 linac, was 60 feet in diameter and 87 feet long, and required 9 MW of RF power at a frequency of 12 MHz. A 20-foot opening was used for access to the vacuum chamber.

RF operation of the Mark 1 was achieved in March 1952, followed by proton acceleration to 33.5 MeV in May. Rearrangement of the drift tubes for 22.5 MeV output provided better operating conditions, allowing intermittent proton acceleration of up to 225 mA and extended continuous (cw) proton acceleration at a current of 50 mA. The ion source/injector could provide 500 mA for several hours before the electodes and focusing grids would burn out. The Mark 1 was dismantled in December 1953.

Mark 1 was the prototype demonstrator for a planned $427M plutonium production plant (MTA) that was to be based on 500 mA of deuterons accelerated to 350 MeV, using a 12 MHz drift-tube linac requiring 400 MW of RF power. The MTA project was cancelled in August 1952 after the discovery of plentiful deposits of uranium in the western USA. A revised production accelerator proposal (the C-50 linac concept) was put forward in 1954, but was not funded; this machine was to be half the cost of MTA for a 500-MeV, 320-mA deuteron linac operating at a frequency of 50-MHz.

Twenty years later a more modern linac, LAMPF, was built for medium energy physics research employing techniques and developments that occurred over those two intervening decades. Many additional developments have taken place since LAMPF was built 20 years ago; these developments have been fully utilized in the conceptual design of APT.

High-Power Linac Technology Base

Accelerator-based nuclear waste transmutation and other nuclear process applications have been discussed for decades, but only recently has the accelerator technology base reached the point that such schemes can be considered feasible. Important factors behind the significant technology advances that have occurred in the past few years have been the large investments in high-current accelerator development made by DOD/SDI programs, and the demanding performance requirements made by these programs. The beam quality and reliability specifications (among others) for SDI applications considerably exceed those required for APT or ATW.

Linac technology has now reached the point that designs in the required parameter regime for transmutation and other applications can be projected with high confidence and assurance of operability.

A Solid Technology Base for High-Power Linear Accelerators Has Been Established in the Past Few Years

- Accelerator-based transmutation was considered earlier, but the technology base for a high-power cw linac was not ready
- Large DOD/SDI investments are producing high-current ion linacs with beam quality specs exceeding APT or ATW requirements
- Linac technology has reached the point that designs in the required parameter regime can be projected with high confidence of operability

Linear Accelerator Terminology

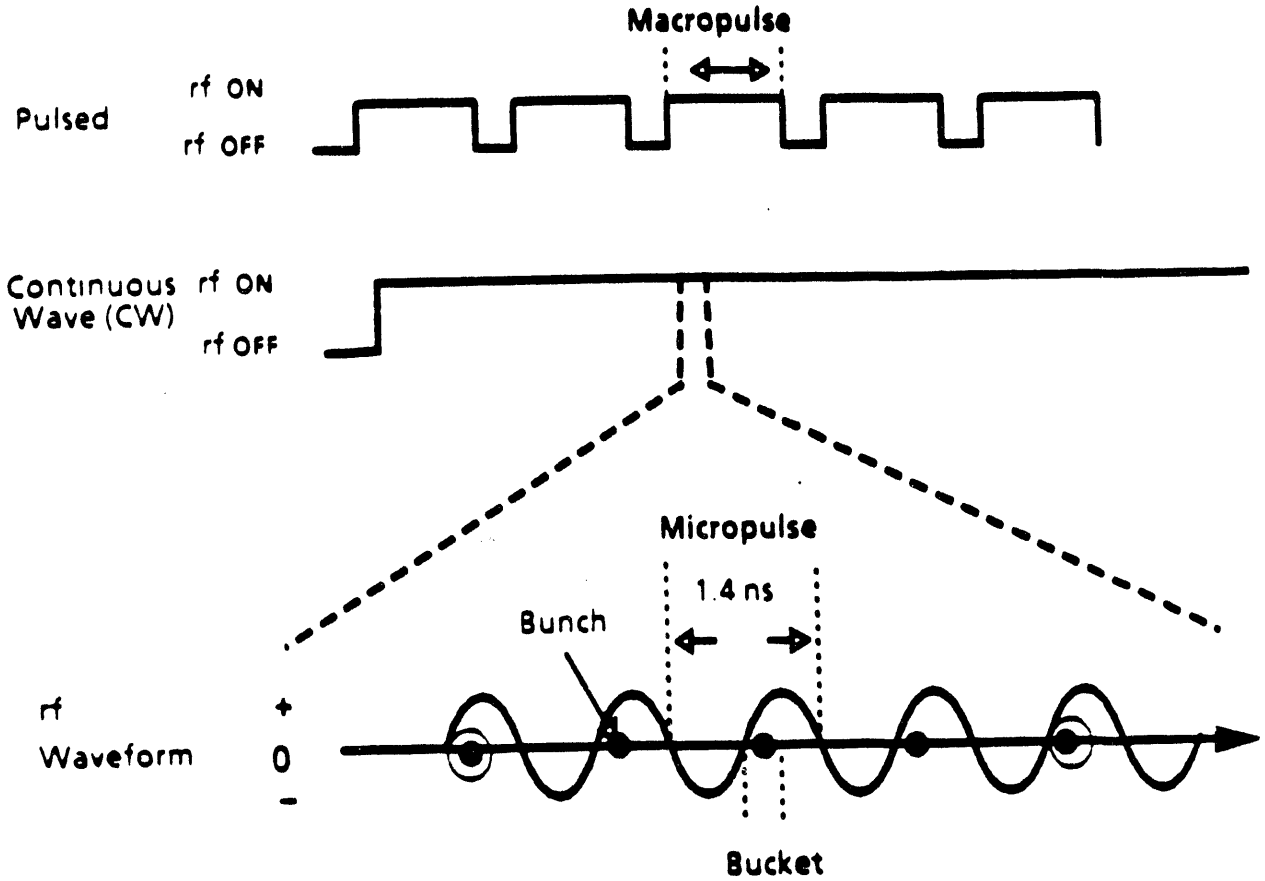
The characteristic current outputs from a pulsed RF linac and one operating continuous wave (cw) are compared in the upper portion of the viewgraph. In a pulsed linac the periods during which the RF cavities are excited (and beam is accelerated) are called macropulses; pulse lengths vary from microseconds to milliseconds, depending on the accelerator application. In a cw RF linac, which is the concept that was proposed for APT, the RF cavities are excited continuously and beam is accelerated as an uninterrupted sequence of micropulses.

RF electromagnetic fields maintained in sequences of resonant cavities provide accelerating and longitudingal focusing forces for the beam particles. Transverse focusing is provided by quadrupole magnets located in field-free regions either inside the cavity structures or between cavity modules. In the center of the viewgraph the RF waveform is shown expanded in time over a short interval for a 700-MHz linac; as indicated in the cartoon the proton beam travels in the accelerator as a steady stream of bunches (micropulses), spaced at intervals of the RF wavelength, 1.4 nanoseconds. Each bunch, represented by the solid circles, remains locked in phase with the RF accelerating field because of stability conditions that exist while the field amplitude is rising, a region known as the RF "bucket".

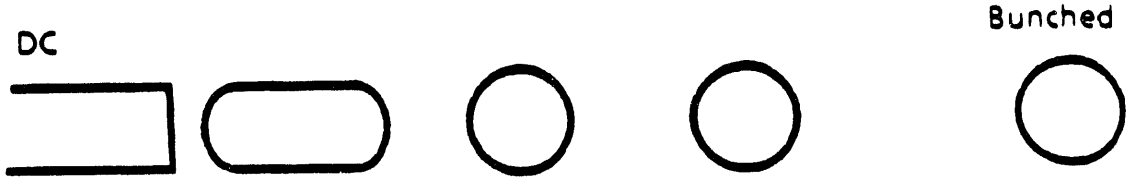
In the APT accelerator design, the frequency relationship between the low-energy (low-beta) portion and the high-energy (high-beta) portion of the machine, coupled with a beam funneling scheme, assure that every 700-MHz rf bucket is filled with protons. This is distinct from LAMPF where only one-quarter of the high-beta accelerator buckets contain beam bunches (indicated as open circles).

The proton beam begins the acceleration process as a steady (DC) stream of low velocity particles extracted from an ion source plasma. In the first accelerating stage, a Radio-Frequency Quadrupole (RFQ) linac, the protons are adiabatically (gradually) captured in RF fields and formed into bunches, which are then accelerated. Following the RFQ the beam bunches are further accelerated by a Drift-Tube Linac (DTL) and then finally by a Coupled-Cavity Linac (CCL). These are resonant structures specifically designed for efficient acceleration of higher and higher velocity particles. Longitudinal focusing inherent in the RF acceleration process maintains the bunch structure as the proton velocity increases.

RF Linear Accelerator – Some Terminology



- Bunch experiences rf fields that provide acceleration and focusing
- ATW linac uses every rf "bucket", unlike LAMPF where only every fourth contains protons
- Beam starts off DC; it is adiabatically captured in rf fields and accelerated in bunches

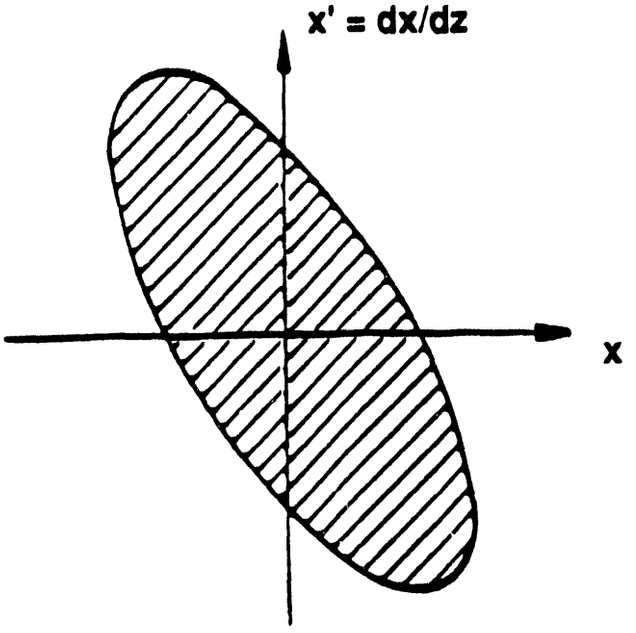


Description of Beams

The upper figure represents the area of one of the three phase plane projections (x - x') of the six-dimensional "hyperellipsoid" phase-space volume bounding the spatial and relative-momentum coordinates of some characteristic fraction of the beam-particle distribution, usually the rms value. A second phase-volume projection is in the other transverse-motion plane (y - y'), and the third is in the longitudinal motion plane represented by (ΔW - ϕ), the relative particle energy and phase compared to the center of the RF bucket. The emittance conveys a measure of the degree of thermodynamic order in the beam (the relative thermal energy of the particles and how close they are to their common axis). It provides a convenient means for comparison of beams, and for representing accelerator performance. Lower emittance implies a higher beam quality and vice versa. The 6-dimensional phase volume of the beam is a conserved quantity and remains constant as long as all particle interactions involve linear fields. In a real beam the emittance increases somewhat as particles are accelerated, especially in the low energy accelerating structures, because the forces are in fact not quite linear. Modern linac design procedures and greatly improved understanding of emittance-control mechanisms now keep this growth fraction to a minimum.

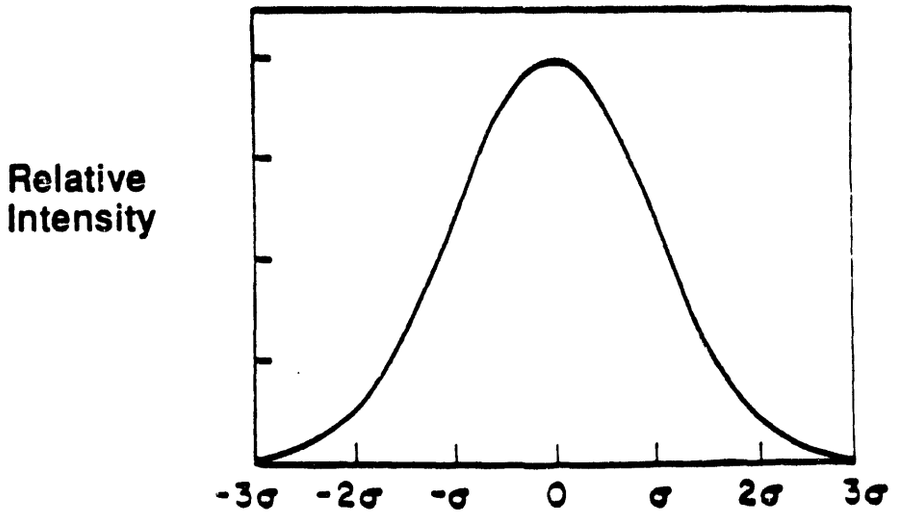
The lower figure shows the cross section through a beam in one of the transverse dimensions, giving the particle density versus distance from the beam axis. A proton linac beam distribution typically has a nearly Gaussian core with a non-Gaussian tail that extends out beyond 3 times the rms value (σ) of the distribution. This tail is known as the "halo". For high-current proton beams (e.g. LAMPF) a design rule (based on measurements of characteristic distributions) is that the accelerator structure or beam pipe aperture dimension must be 5 to 7 times the rms beam size to avoid loss of a significant fraction of the particles populating the outer reaches of the halo.

Describing a Beam – More Terminology



- Emittance is the phase space volume occupied by the beam, described as projected areas in x-x', y-y', ΔW-ϕ planes

- Radial beam intensity distribution is roughly Gaussian with a "halo" persisting out well beyond the $\pm 3\sigma$ points



Accelerator Technology Advances That Enable High Power Linacs

Some of the key recent advances in the technology of high-current linear accelerators are listed. Taken together these improvements in beam-dynamics understanding, high-current beam simulation, system design tools, and device performance and capabilities now make it possible to build and operate machines in the APT and ATW class.

Key Advances in Accelerator Technology That Enable High Power Linacs

- **Radio-Frequency Quadrupole (RFQ)** – dramatic improvement in first stage of acceleration.
- **Beam Funneling** – intensity doubling with no emittance penalty.
- **Beam Dynamics** – analytic understanding of emittance preservation.
  - Higher-frequency accelerating structures
  - Ramped accelerating gradients
  - Strong transverse focusing
- **Beam Simulation** – accurate codes for predicting detailed behavior of high current beams; benchmarked on LAMPF and ATS.
- **RF Sources** – availability of efficient high-power (1-MW) cw klystrons at 350 MHz – 1000 MHz.
- **Beam Optics** – high order optics codes allow beam-shaping using nonlinear magnetic elements (octupoles).
- **Controls and Diagnostics** – microprocessor-based distributed intelligence; fast non-intercepting beam position monitors.

Progress in Linac Design and Performance – From LAMPF to APT

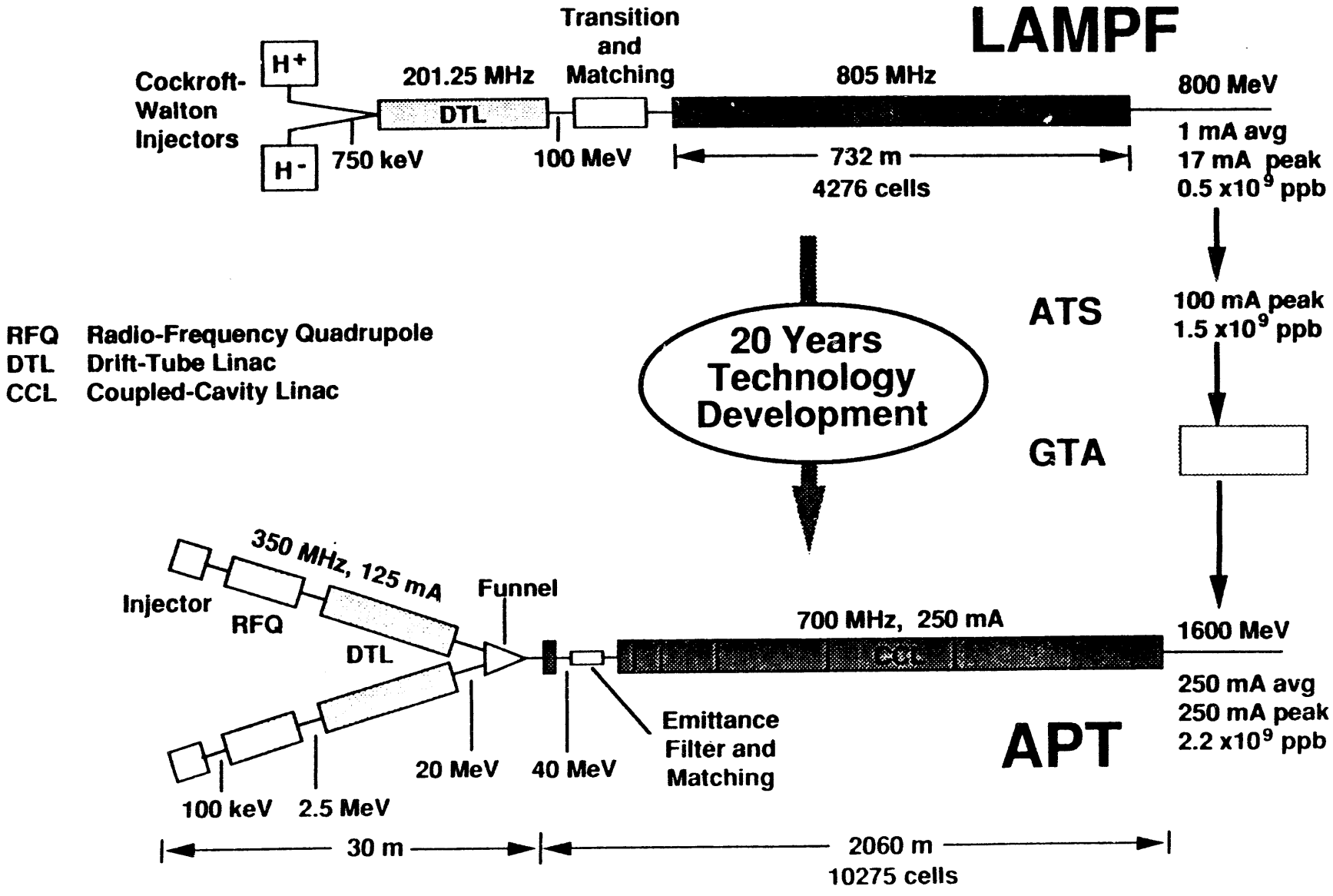
Twenty years of linac technology development has taken place since LAMPF was built. There have been great advances in component development, beam-dynamics understanding, new methods and techniques, and new design tools, much of which has been concentrated in the past ten years – stimulated partly by Strategic Defense Initiative (SDI) requirements for high performance linacs that can operate in a space environment.

The RFQ in APT provides much improved beam performance in the initial acceleration stage, compared with the high-voltage injectors used in older linac designs, delivering a high quality beam to downstream accelerating stages. The APT injector operates at the low potential of 100 kV, which provides more reliable and improved performance with cw beams. For reasons of efficiency, improved performance and modularity, the APT linac makes the change from DTL to CCL at 20 MeV, a much lower transition energy than in LAMPF.

Funneling, recently demonstrated at Los Alamos, is used to double the beam current at 20 MeV without a significant emittance increase, and to fill every RF bucket in the 700-MHz CCL. This permits attainment of 250 mA in the CCL with low emittance and also minimizes the charge per bunch – thereby reducing the potential for beam loss and resultant heating and activation in the accelerator. Although the average current increases by a factor of 250 from LAMPF to APT, the increase in the charge-per-bunch is only a factor of 4.5.

As part of the SDI Neutral Particle Beam (NPB) program, the Accelerator Test Stand (ATS) at Los Alamos was used to demonstrate advances in low-energy high-current linear accelerator technology. Acceleration of protons to 7 MeV was demonstrated on ATS with a charge-per-bunch about 70% of that for APT, and with a much lower emittance. Code verification experiments and operation of ATS provide confidence that the front end of APT should work as designed. The Ground Test Accelerator (GTA) at Los Alamos is providing a ground demonstration of an accelerator that could be used as part of the SDI/NPB architecture. GTA, which is a fully integrated accelerator system test with components similar to APT, has performance and operating objectives significantly more demanding than that for APT. It has a smaller beam emittance, cryogenic operation, fully automated turn-on, higher accelerating gradients in the RFQ, DTL, and CCL, and a very complex output beam transport system.

Progress in Linac Design and Performance



LAMPF Provides a Relevant Basis for Extrapolation

Standard production conditions for LAMPF, which is a pulsed RF linac designed more than 20 years ago, provide an 800-MeV proton beam with 15-mA peak current and 1-mA average current. The macropulses are delived at a rate of 120 pulses per second and are 560 μs in length, giving a 6.7% duty factor. LAMPF has the highest beam power of any accelerator worldwide in this energy range and has operated as a proton production factory (3000 – 4000 hours/year) for more than ten years with demonstrated availability (fraction of scheduled beam time) greater than 85%. Medium-energy physics program funding levels have restricted beam operation during the past few years to about 6 months per year.

LAMPF has accelerating structures that are similar to those needed for APT – an H^+ injector for proton beam initiation, a 200-MHz drift tube linac accelerating the beam to 100 MeV, and an 800-MHz coupled cavity linac accelerating the beam to 800 MeV. LAMPF components, such as beam diagnostics, feed-forward RF amplitude control, ion pumps, quadrupole focusing magnets, RF power sources, and beam transport elements, are also similar to components needed in APT. Machine safety and maintenance arrangements are very relevant to the APT design. Finally, LAMPF has a user community that demands reliable output beam with constant and consistent performance over long periods of time. Many APT components would be straightforward extensions of existing LAMPF hardware. Other components would be replaced or modified to take advantage of recent technology advances. The RFQ, which had not been invented when LAMPF was built , is a replacement for the high-voltage DC injector used in LAMPF; it dramatically improves beam performance in the first accelerating stage of APT.

Beam losses and activation levels are very low for most parts of the LAMPF linac. Total losses are carefully restricted during machine tuning and production operation by a fast beam-loss monitoring system. Unrestricted hands-on maintenance is possible along the entire length of the accelerator. Well developed remote-handling techniques are employed for maintenance and removal of highly activated components in the experimental area (production targets).

LAMPF Provides a Historical Technology Base

Parameters

- 800 MeV proton energy
- Peak current: 15 mA
- Pulse length, rate: 560 μs, 120/s
- Average current: 1 mA
- Output emittance: 0.07 π cm-mrad (n-rms)
- Side-coupled 805-MHz CCL
- Only 1/4 of CCL rf buckets contain protons
- 44 klystrons (1.25 MW peak power)
- H^+ and H^- beams accelerated simultaneously
- 1.91 cm CCL structure aperture (radius)

Operations

- Beam availability: > 85% of scheduled time
- Beam loss at 800 MeV: < 0.2 nA/m (2×10^{-7}/m)

Design Issues for High Power Linacs

The most important design concerns for high-power proton linacs are:

1) Control of beam losses, including the slow beam loss associated with interception of the beam halo by the accelerator structures and point losses caused by off-normal conditions and faults.

2) RF power capital costs and efficiency. The former has a dominant effect of the capital cost of the accelerator, and the latter, expressed as the ratio of RF power delivered to the beam to the total RF power delivered to the accelerator system, has a dominant effect on the operating cost of the machine.

3) Machine availability. For ATW applications the accelerator must have a total availability exceeding 75%. This requirement translates into demanding specifications for component lifetime, speed and efficiency of maintenance, and minimal activation for most of the accelerator.

4) Integrated system operability. An APT-class accelerator will be a complex high-power system involving thousands of components. Although many relevant accelerator parameters (peak currents, emittance values, etc.) have been demonstrated at APT/ATW performance levels, there is as yet no existence proof of integrated cw operation of a high-power accelerator facility in the required power range. There are many operational details that must be addressed, including beam startup, recovery from faults, protection of equipment, safety, etc.

Design Issues for High Power Proton Linacs

- Beam loss control
- RF power cost & efficiency
- Machine availability
- Integrated system operability

APT Accelerator Design Framework

The high-power linear accelerator concept developed to address APT requirements (250 mA cw, 1.6 GeV, 75% availability) was prepared within a conservative framework to produce a convincing reference model. Initial parameter selections for the RFQ, DTL, and CCL accelerating stages were made analytically using a uniformly-filled ellipsoidal bunch model. End-to-end beam simulations (with built-in errors to represent "real-world" conditions) were carried out to confirm and adjust these selections, and to determine beam performance, emittance estimates, etc.

LAMPF operating experience was consulted extensively during the design process, especially to obtain information on beam losses and machine activation, machine availability, component maintenance requirements, and klystron lifetimes. In addition, the LAMPF linac was simulated from end to end with "as-built" data using the same design codes that calculated APT performance, in order to provide a detailed benchmark for making beam loss projections for APT.

Preliminary engineering design was carried out for critical components, such as the radiation-hard electromagnet quadrupoles in the DTL drift tubes, to verify feasibility. Engineering analysis was carried out for certain areas of the accelerator design, such as the thermal response of the CCL cavities, to verify performance within acceptable parameter ranges. Accelerator and tunnel layouts were produced to check configurational reasonableness.

A preliminary fault and error analysis was completed, to assess the threat to accelerator components of various kinds of malfunctions and off-normal conditions. The effects of RF station failures and magnet failures were analyzed, and estimates were made of the beam shutdown time allowances to prevent damage to the accelerator system.

The APT accelerator front end (up to about 40 MeV) was designed to launch a high quality, low halo beam. The CCL, where most of the construction cost and operating cost impact is concentrated, was designed to assure very low beam losses while maintaining a high RF efficiency.

The APT accelerator concept was reviewed in the fall of 1989 by the Energy Research Advisory Board (ERAB) and was found to be technically credible, assuming a suitable development and engineering demonstration program.

APT Accelerator Concept was Developed in Summer 1989 and Reviewed by ERAB Panel

- **Conservative overall design framework**
  - Parameter selection using uniform ellipsoid bunch model
  - Comprehensive beam simulation including built-in errors
  - Assessment of LAMPF operating experience
  - Engineering analysis of critical components
  - Machine configuration layouts
  - Analysis of off-normal and fault conditions
  - Frequency choice to match available high-power RF sources

- **Front end is designed to launch high quality, low halo beam.**

- **CCL is designed for low beam loss, high rf efficiency.**

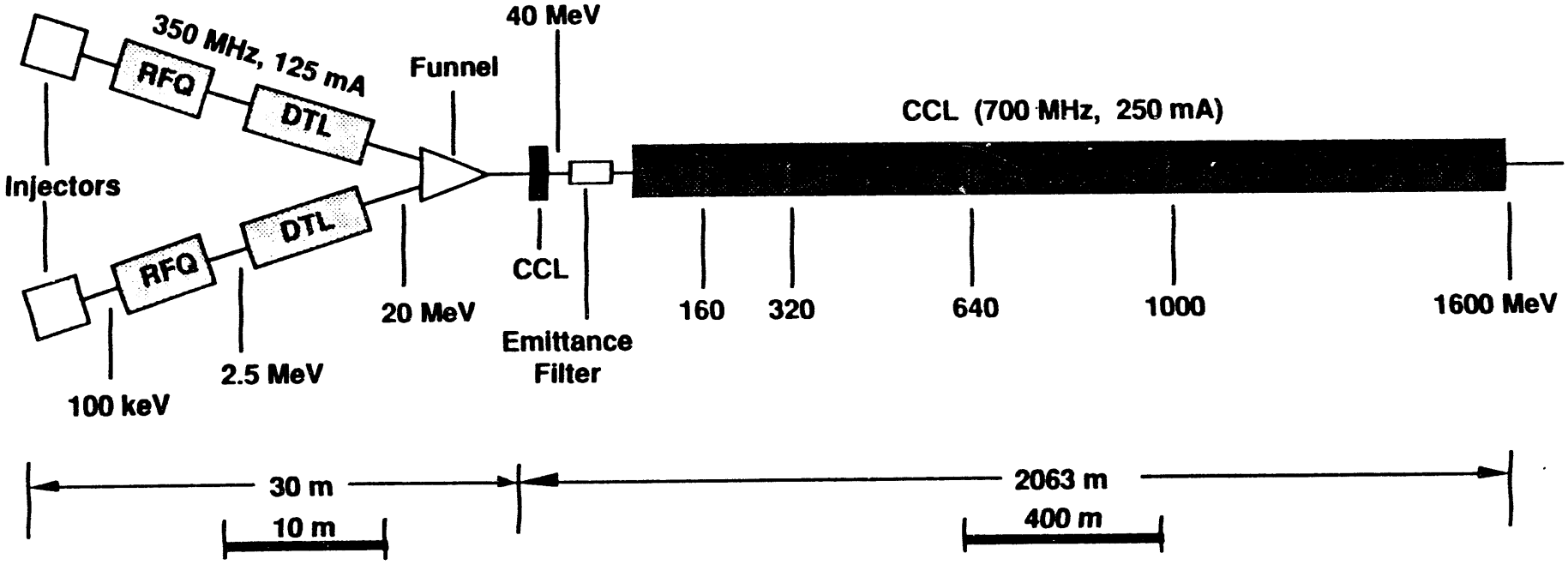
Reference APT Accelerator Concept

The APT linac sketched in the figure is composed of two principal parts, a low-energy beam launcher, and a coupled cavity linac (CCL) that makes up most of the accelerator's 2-km length. The beam launcher is a funneled accelerator system composed of two H^+ injectors, two 350-MHz RFQs, and two 350-MHz DTLs. It combines two 125-mA, 350-MHz beams at an energy of 20 MeV to provide a 250-mA, 700-MHz beam to the CCL. The first segment of the 700-MHz CCL is shown in dark shading immediately to the right of the funnel. The beam launcher is optimized to generate a high-current, low emittance beam that has little halo, taking advantage of the advances in accelerator technology listed earlier, while the CCL is designed to assure very low beam loss and provide relatively high RF efficiency.

The use of radiation-hard electromagnet (EM) quadrupoles in the DTL drift tubes instead of permanent magnet quadrupoles is mandated by the significant radiation damage threat during turn-on and tuning. This decision imposes an upper limit to the DTL frequency at about 400 MHz because of elecromechanical constraints in the EM quad design, and leads to a natural frequency choice of 350 MHz, where a commercially manufactured 1-MW cw klystron is available. The DTL is of $2\beta\lambda$ type to accommodate focusing and acceleration requirements. The CCL is designed in a modular fashion, and is divided into seven sections in which each accelerating cavity has the same number of coupled accelerating cells. The number of cells per module increases from 2 in the lowest-energy CCL section to 10 in the highest-energy section. Following each accelerating module is a diagnostic station and a focusing quadrupole (in a FODO sequence). The density of quadrupoles along the CCL is about four times greater than in LAMPF, one of the factors producing a small beam size in the APT linac.

The table lists a number of important power parameters for the entire linac, including the RF power to the cavity walls, the power into the beam, the ac-to-beam power conversion efficiency, and the total ac power requirement. It also lists the average accelerating gradient in the CCL, the number of particles in each CCL bunch, the ratio of aperture to rms beam size in the CCL, and the transverse input and output emittance values derived from an end-to-end beam simulation in which matching errors were deliberately intoduced to simulate "real world" conditions. The aperture/rms-beam size ratio is much larger than in LAMPF, where this ratio at the end of the linac is about 6.3.

APT Accelerator Concept (1.6 GeV, 250 mA)



RF Power (beam)	400 MW	Input Emittance	0.020 π cm-mrad
RF Power (cavities)	118 MW	Output Emittance	0.068 π cm-mrad
1.25-MW (cw) Klystrons	482	Aperture/Beam-Size	15 – 20
AC to Beam Efficiency	0.48	Accel Gradient (CCL)	1.0 MV/m
AC Power Input	910 MW	Protons/Bunch (CCL)	2.2×10^9

Beam Launcher Design

The low energy portion of the APT accelerator, up to 20 MeV, is referred to as the beam launcher. It is a funneled system composed of two100-keV H^+ injectors, two 350-MHz RFQs accelerating 125-mA beams to 2.5 MeV, and two 350-MHz DTLs accelerating these beams to 20 MeV. Funneling takes place at 20 MeV. Following a matching section, the beam is injected into the first stage of the 700-MHz CCL.

The APT beam launcher design takes advantage of several important advances in the technology of high-current low-beta linacs. It uses RFQs for bunching and initial acceleration to provide high current with good beam quality in the critical first accelerating stage, funneling for doubling the current into the CCL without a corresponding increase in emittance, a high accelerating-structure frequency in the RFQs and DTLs (350 MHz) to minimize transverse emittance growth in these structures, and a ramped accelerating field in the DTLs (1.1 to 3.1 MV/m) to control longitudinal emittance growth. Beam simulations show that with these design features the beam launcher delivers a high current (250 mA) beam to the CCL with low emittance and very little halo.

Although permanent-magnet quadrupoles have proven advantageous for strong focusing in high-frequency linacs intended for short service times, as in the Neutral Particle Beam SDI program, their performance degrades after accumulated neutron fluences as low as 10^{15} n/cm^2-s. Because of the significant neutron damage threat over long operating times in APT, their use was precluded in the DTL design. Radiation-hard electromagnets are specified instead, and the constraints associated with fitting such EM quads into drift tubes leads to a design choice of 350 MHz for the DTL operating frequency. A proven 1-MW cw klystron is available commercially at this frequency and is a another factor governing the frequency selection.

Beam Launcher Design Features

- Beam funneling
- RFQs for initial bunching and acceleration
- High accelerating structure frequency (350 MHz)
- Ramped accelerating field in DTL
- No PM quadrupoles in DTL; radiation damage threat
- Proven 1-MW cw klystrons available at 350 MHz
- Single frequency transition (at 20 MeV)
- Matching at transitions between structures

CCL Design Features

The principal objectives governing design of the APT CCL are to minimize beam loss in the accelerator, and to maintain a high efficiency for conversion of electric power to beam power. The former impacts the maintainability of the accelerator, and the latter dominates both the capital cost and operating cost of the faciltiy.

To achieve very low beam losses, the acceptance in both transverse and longitudinal phase spaces is made as large as possible compared with the phase space occupied by the beam bunches. With low beam emittance, strong transverse focusing (high quadrupole density), and unusually large apertures in the accelerating structures, it was possible to achieve a ratio of structure aperture to rms beam size in the range 15 to 20, much higher than in LAMPF, which has a ratio of about 6.3. A large ratio of "bucket" to bunch dimensions is also achieved in the longitudinal phase plane.

Additional features assuring low beam loss are the elimination of major transitions in either the transverse or longitudinal acceptances, careful matching between different CCL segments, a possible emittance filter (beam halo scraping system) after the initial CCL section (at about 40 MeV), and a high density of beam diagnostics to permit precise control of the beam bunches within the transverse and longitudinal acceptance boundaries. Fast acting (20 to 30 microseconds) beam abort systems would be used to shut off the beam within times short enough to avoid thermal damage to the accelerator in the event of off-normal conditions.

High RF efficiency is achieved by using a low average accelerating gradient (1 MV/m) to minimize RF losses in the accelerating cavities. This value is close to optimum for minimizing projected facility lifetime costs. A new RF tube is needed at 700 MHz for powering the CCL. Accelerator capital and operating costs could be greatly reduced through a development program to produce a low-cost high-efficiency RF source for this application.

CCL Design Features

Beam-Loss Control

- **No major acceptance transitions**
- **Beam phase space occupies small fraction of CCL acceptance**
  - Strong transverse focusing (short tanks)
  - Ratio of structure bore to rms beam size is very large
  - Ratio of RF bucket length to bunch length is large
- **High density of beam diagnostics; tight control**
- **Emittance filter system at 40 MeV**
- **Fast shutdown in fault situations (< 30 μs)**

RF Efficiency

- **Low accelerating gradient (1 MV/m)**
- **New high-efficiency 1-MW cw 700-MHz rf source**

Beam Distributions from Simulations

Shown are the results of an end-to-end beam simulation for the APT linac design at the output energy of 1.6 GeV. The calculation includes deliberately introduced mismatches to simulate "real world" tuning conditions. Each dot represents a macro-particle that was accelerated from the injector to the end of the CCL. Approximately 7500 macro-particles were used in the simulations.

The two upper figures are emittance diagrams for the orthogonal transverse phase planes, x - x' and y - y'.

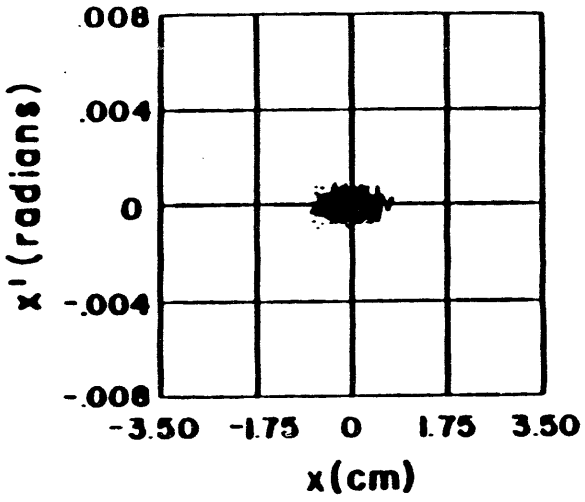
The lower left figure shows the real transverse beam distribution (in x - y space) as well as the structure boundary (transverse aperture), revealing that the design acceptance is very much larger than the transverse phase space occupied by the beam.

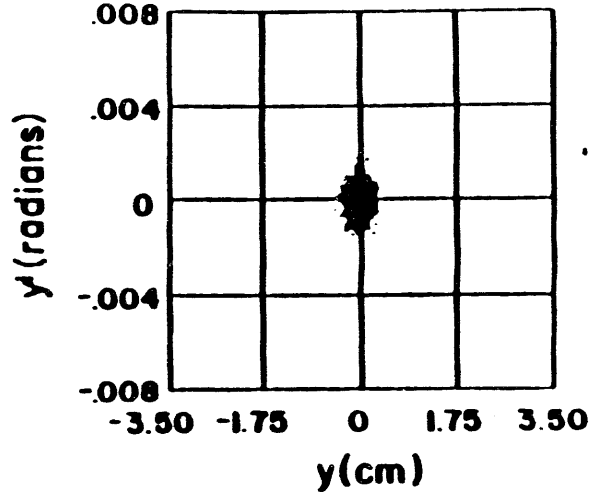
The lower right figure shows the beam distribution in longitudinal phase space, and also the available RF bucket stability area, indicating that the longitudinal acceptance of the linac is much larger than the longitudinal emittance of the beam bunches.

Because the ratios of acceptance to emittance are so large, the beam performance is excellent and beam loss is very small.

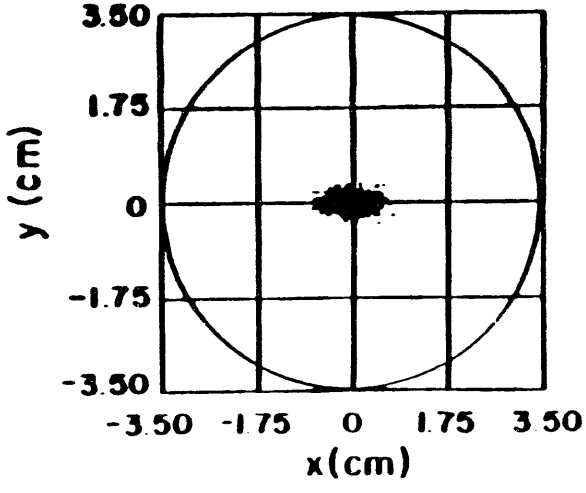
Output Beam Distributions from Linac Simulation

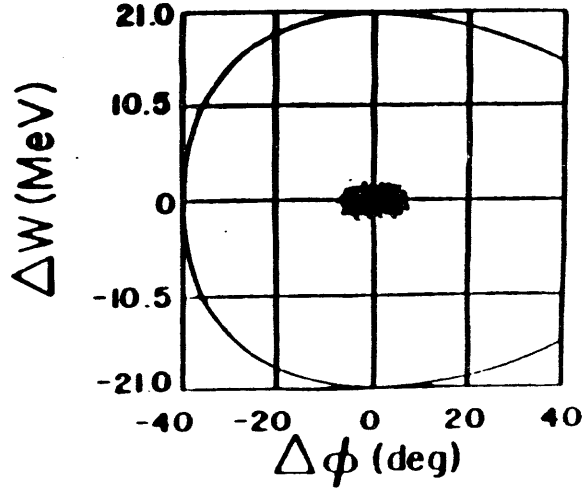
Results confirm that beam dimensions are much smaller than transverse and longitudinal acceptances.





Calculation was run to approximate realistic alignment errors and optical mismatches.



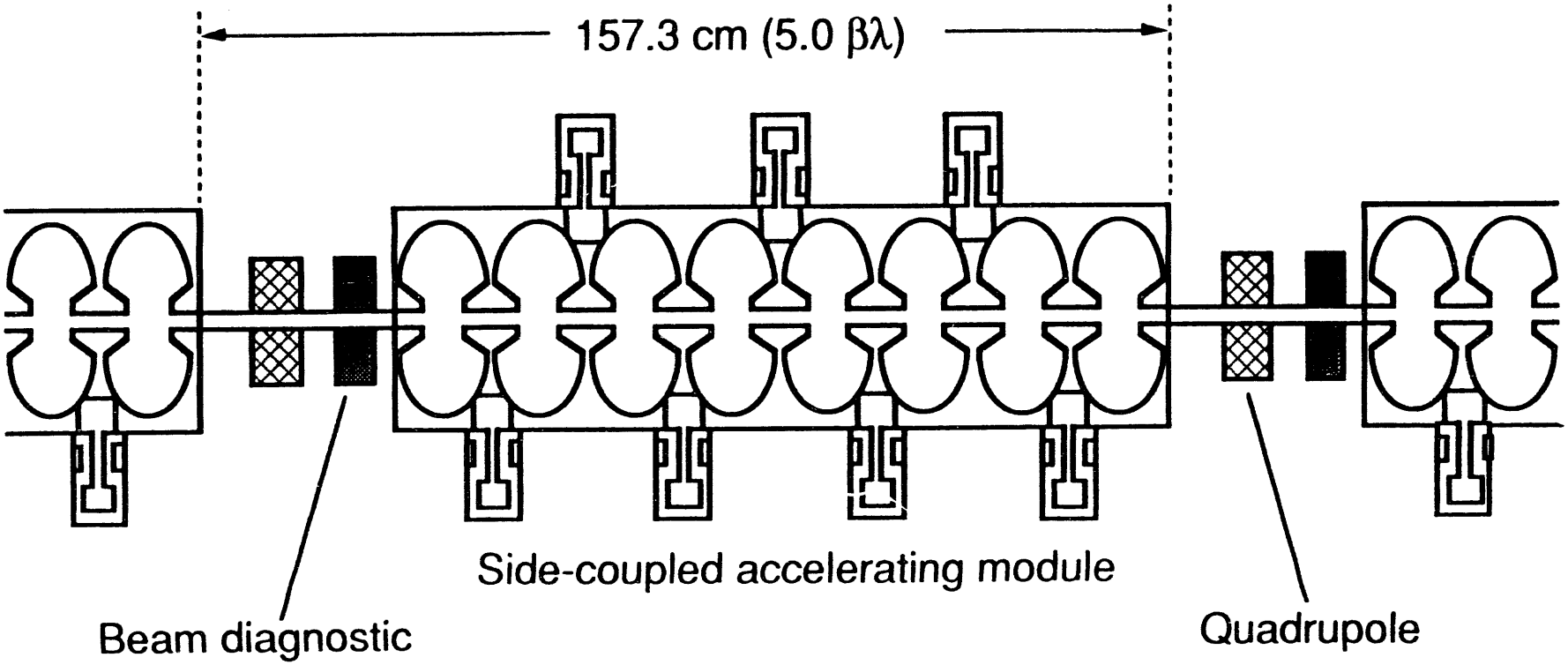


Typical CCL Accelerating Module

A typical side-coupled-cavity accelerating module in section 5 of the CCL is shown. This type of lattice unit is used for accelerating the proton beam between 320 MeV and 640 MeV (where the relative velocity β ranges from 0.66 to 0.80), and has eight accelerating cells, spaced at intervals of $\beta\lambda/2$. As shown, each lattice unit includes a quadrupole and a beam diagnostic station. There are 266 similar units in this section of the CCL, with each one (on the average) requiring 380 kW of RF power input when beam is on. The units are bridged together in pairs, with each pair supplied by a single 1-MW 700-MHz klystron. Connecting the modules with bridge couplers minimizes the number of individual RF connections and feeds, and eases control aspects because the RF fields are locked for longer segments of the llinac.

The mean lattice-unit length is 157 cm, and the cavity shunt impedance is 24 MΩ/m. The structure-average accelerating gradient is 1.25 MV/m, and RF power dissipated in the cavity walls of each unit is an average of 80 kW. The structure thermal response for cw operation has been calculated and is shown to be well within acceptable bounds for a water-cooled system.

CCL Eight-Cell Lattice Unit (Section 5)

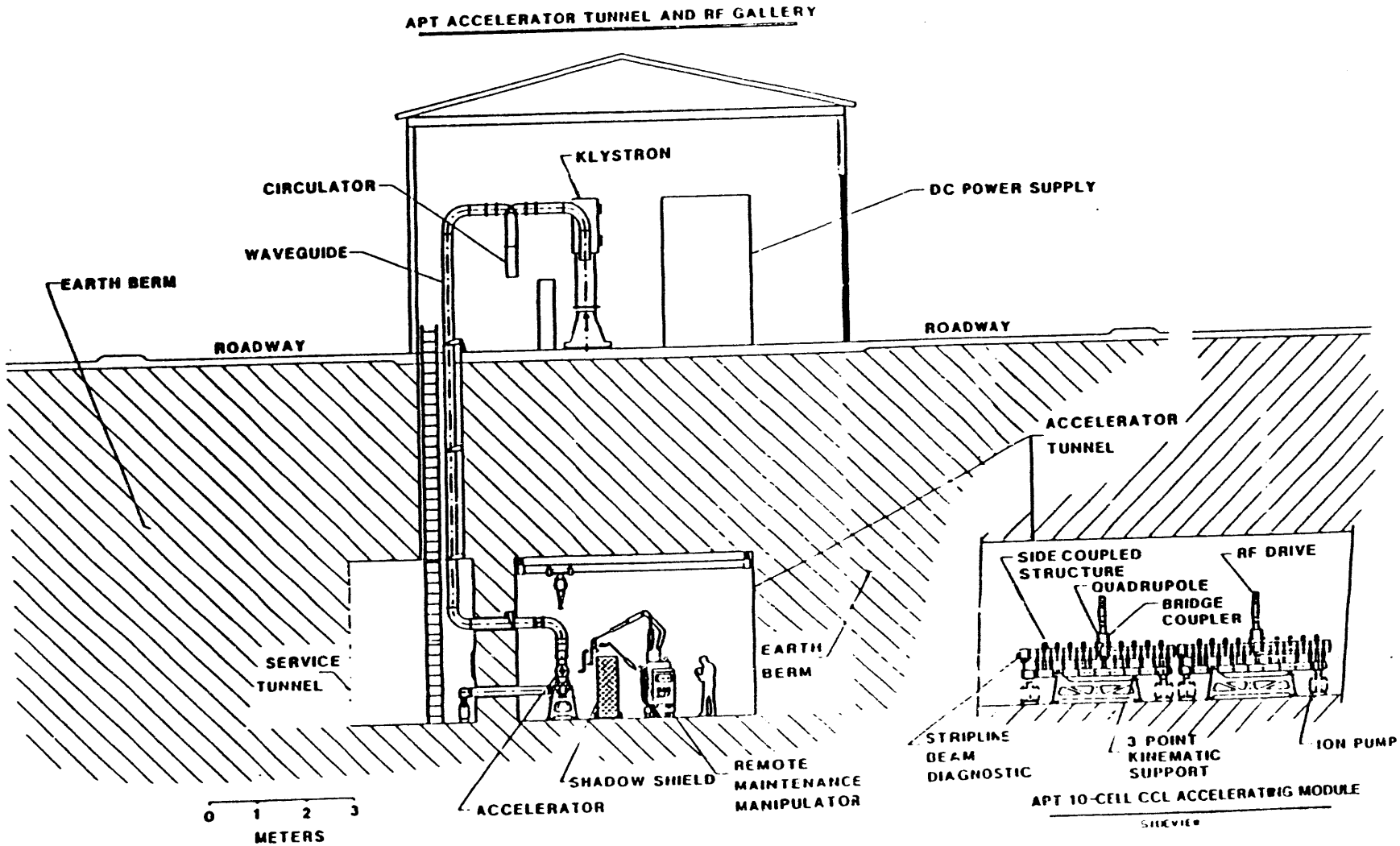


Machine and Tunnel Configuration

The accelerator and tunnel configuration for APT would be very similar to that for LAMPF. The figure shows a section through the coupled-cavity linac, indicating that the accelerator is in a tunnel under about 15-20 meters of earth overlay for shielding. Above the earth shield is a building extending the full length of the linac that houses the RF sources (klystrons), circulators, RF amplifier controls, power supplies for the klystrons, focusing magnets, and ion pumps, and the control system interface for all the foregoing as well as for the beam diagnostics. Shielding will be sufficient that this area is a low radiation zone during operation, and is accessible to the accelerator staff when beam is on. RF power is delivered to the accelerating cavities by WR1150 waveguide run through the earth overlay in such a way as to avoid neutron streaming from the linac tunnel to the klystron gallery. A service tunnel situated adjacent to the accelerator tunnel houses the relatively high-maintenance vacuum pumps, the cooling interface equipment, and certain diagnostic instrumentation that must be close to the machine. Shielding between the service tunnel and the accelerator reduces radiation to low enough levels to prevent damage to the electronics located there and to ensure that activation will be low enough to permit unlimited service access when beam is off. The machine tunnel is equipped with a radio-or-manually controlled bridge crane, and shadow shielding for hands-on maintenance in locally "hot" sections of the linac, if required. The modular nature of the linac design allows relatively straightforward removal of quadrupoles, diagnostic stations, rf windows, and part or all of a given accelerating structure. Connections are designed specifically for compatibility with remote maintenance equipment.

To the right of the machine tunnel cross section is shown a side view of four 8-cell cavity modules (section 5 of the CCL) placed on their kinematic mounts. Between each pair of cavity modules is a quadrupole magnet and a stripline-type beam diagnostic. At this location in the CCL the cavity modules are bridged together in pairs, with each pair supplied with RF power by a single 1-MW klystron.

Accelerator/Tunnel Configuration



RF System Efficiency

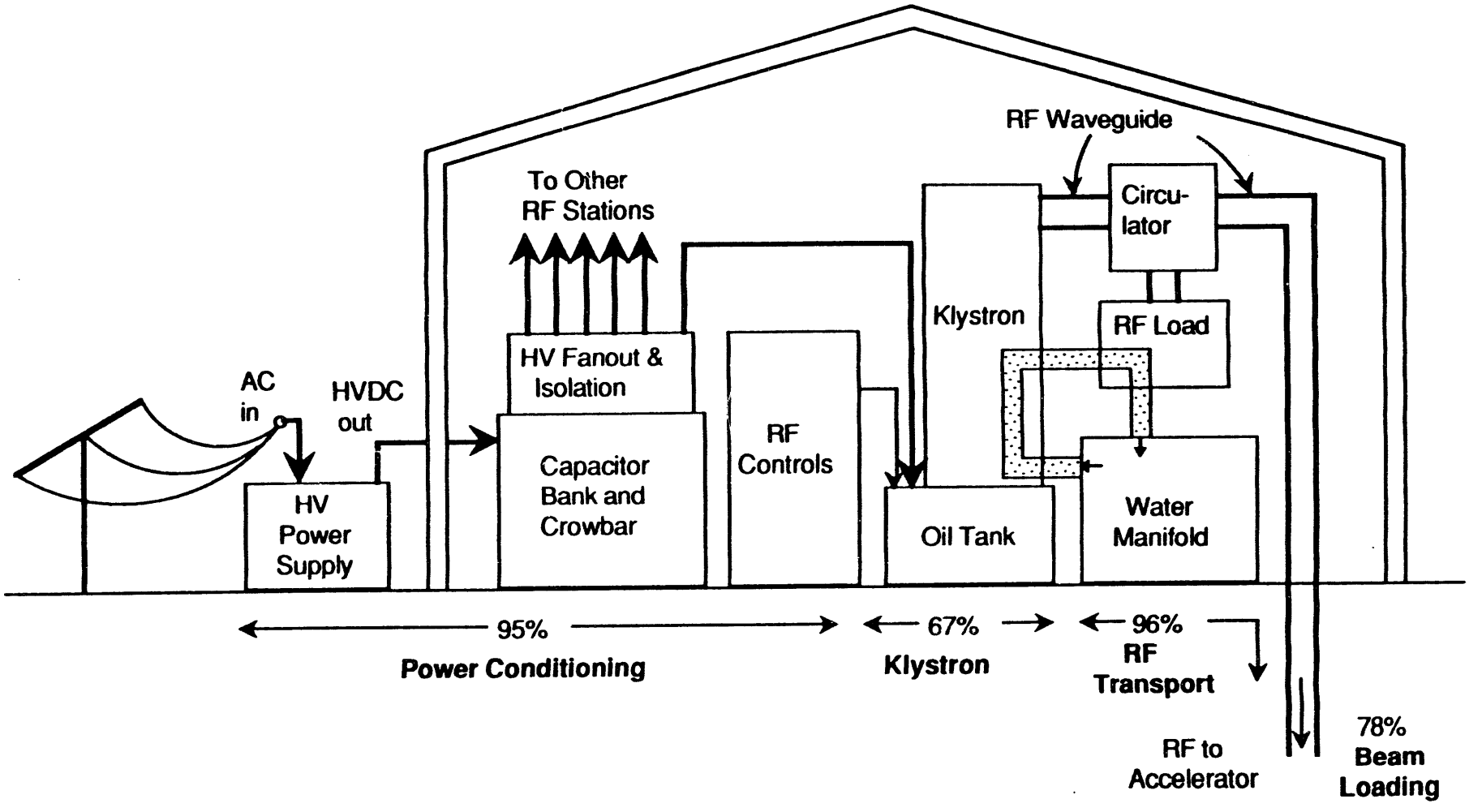
The saturated (dc-to-RF) efficiency of a modern cw 1-MW klystron is 70%. In order to allow margin for control, the design output level should be 95% of saturation, which reduces the effective klystron efficiency to 67%. Improvements in RF generator efficiency are possible, and other kinds of generators and techniques which are still in the developmental stage have predicted dc-to-RF efficiencies greater than 80%.

The RF transport efficiency is taken as 96%, implying a total loss of 0.2 dB. Most of this loss would occur in the circulator used to protect the klyston from reflected power during transients and faults. The losses might be reduced with the use of a "magic tee" as the RF tube isolation device. Other considerations for the tube isolation hardware, such as system complexity and flexibility, lead to the tentative choice of circulators for the APT baseline RF system design.

Losses in the power conditioning portion of the RF system occur primarily in the high-voltage power supply and in the fanout and isolation section. The use of reactive isolation instead of resistive isolation can minimize the losses; an overall efficiency of 95% for this portion of the system is feasible.

The final efficiency term comes from the beam loading factor. Higher beam loading leads to less wasted power in the accelerator. The 78% beam loading in APT is already very high for a room-temperature machine, but there remains the possibility of raising this factor somewhat in an overall design optimization.

RF System Overview



Electrical Efficiency = $\frac{\text{Beam Power}}{\text{AC Power}}$ = 48% **Klystron Power = 1.25 MW cw (saturated)** **Klystron Efficiency = 70% (saturated)**	**Number of Klystrons** **700 MHz 449** **350 MHz 10**

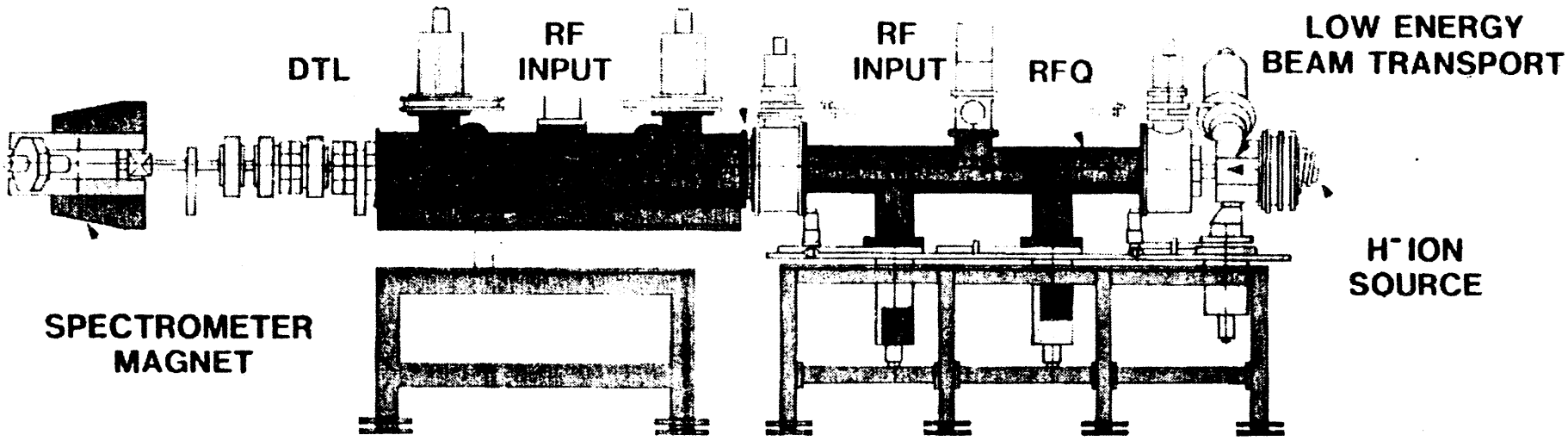
Chalk River High-Power CW RFQ

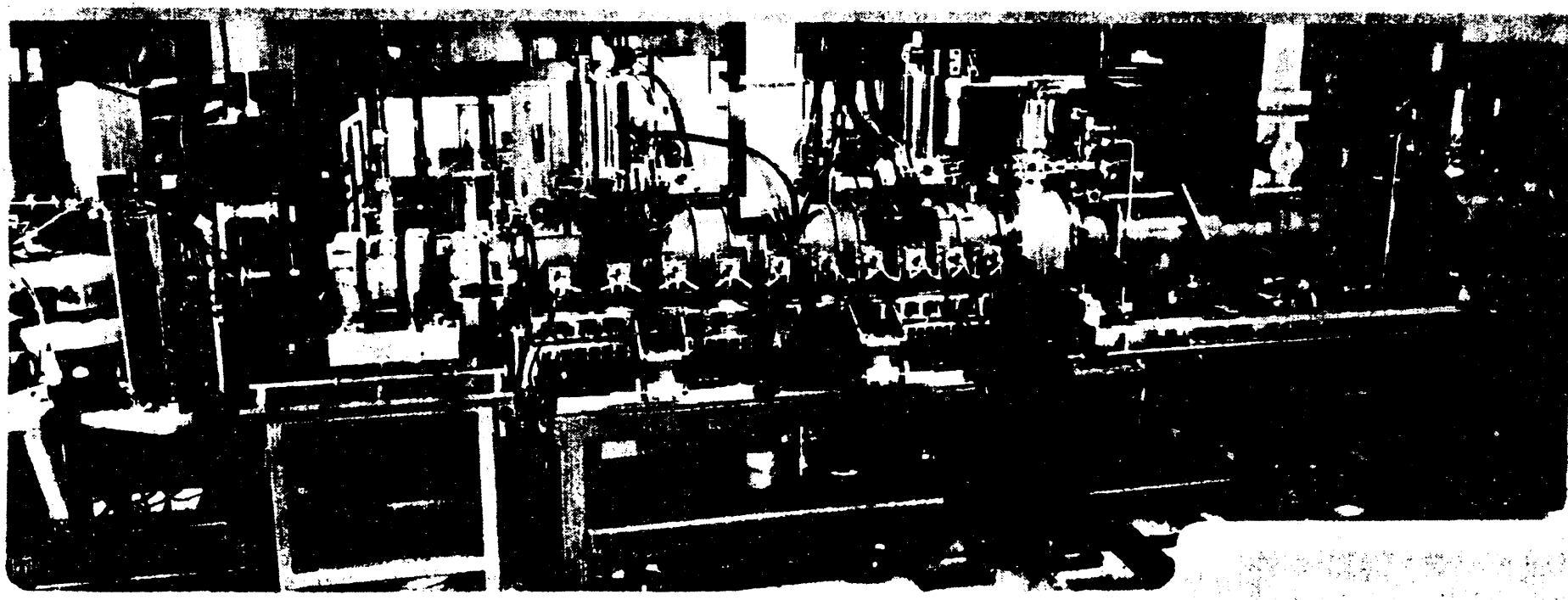
The photo depicts the 269-MHz, 600 keV proton RFQ presently undergoing tests at Chalk River Nuclear Laboratories. This RFQ has recently produced a 70-mA cw beam of protons within an emittance similar to that needed for APT. The output current is close to the theoretical design current limit for this device.

Accelerator Test Stand

The composite picture shows the Accelerator Test Stand hardware and a layout diagram. Acceleration of pulsed (low duty factor) 100-mA proton beams up to an energy of 7 MeV has been demonstrated at this test facility with a 425-MHz RFQ and ramped-gradient 425-MHz DTL. Detailed beam performance measurements made on ATS have provided overall benchmarking of high-current design and beam simulation techniques. It has operated as an integrated low-energy high-current accelerating system validating design procedures for injectors, and RFQ/DTL combinations in the same parameter range as used in the APT concept design.

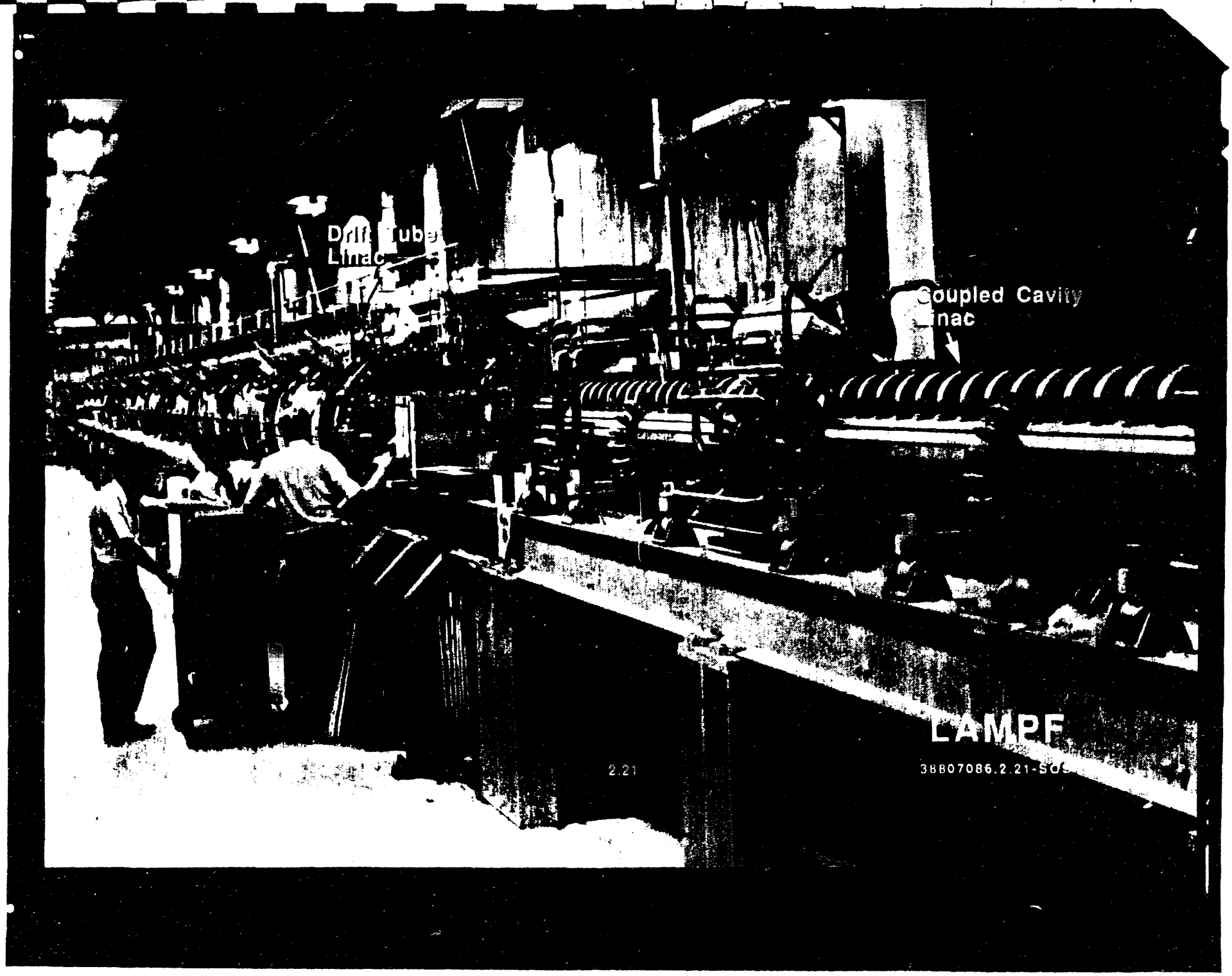
NEUTRAL PARTICLE BEAM
Accelerator Test Stand





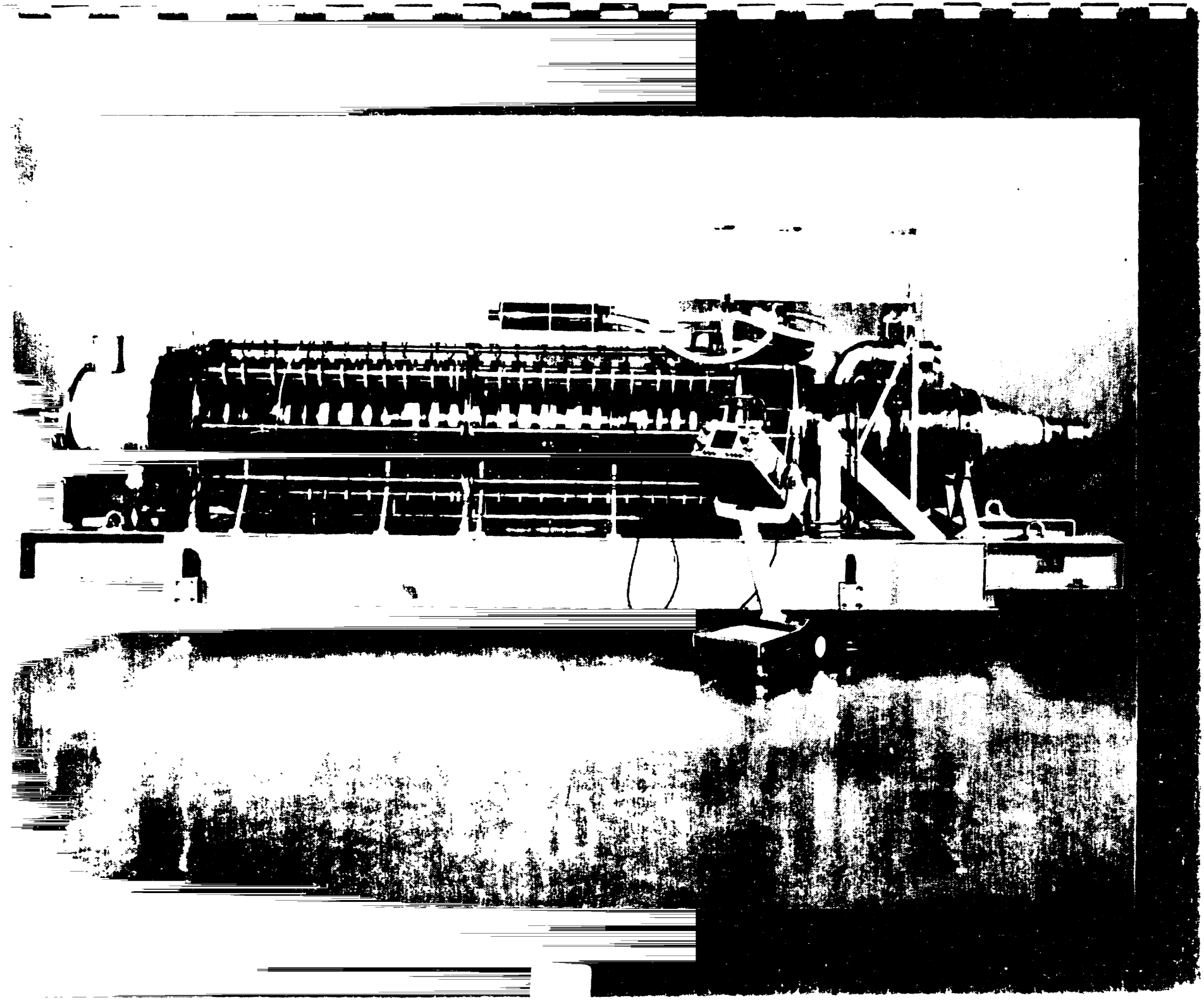
Chalk River CW Drift-Tube Linac

The photo depicts a 269-MHz drift-tube linac that has operated cw at Chalk River Nuclear Laboratories, accelerating a 3-mA beam of protons to 3 MeV (from 750 keV). This device was part of the ZEBRA project, a high-current cw proton linac R&D program aimed at implementation of an electronuclear fuel breeding facility in Canada.


Drift Tube
Linac
Coupled Cavity
Linac
LAMPF
2.21
38807086.2.21-SO

352-MHz cw High-Power Klystron

The photograph shows a 1.1-MW 352-MHz cw klyston, an RF tube developed by two manufacturers to power the Large Electron Positron (LEP) collider at CERN. These klystrons have DC-to-RF efficiencies of 67 to 70% and projected lifetimes of 20,000 to 50,000 hours. More than 20 units are in operation, but lifetime experience is limited so far due to their short service history. The klystrons are candidate power sources for the APT 350-MHz RFQs and DTLs. They are manufactured by Thompson CSF (France) and Valvo-Phillips (Germany), and sell for about $500K per unit.



Key Technical Issues Were Addressed in the APT Design Study, But We Don't Have All the Answers for High-Power CW Operation

A list of major technical issues confronting the APT accelerator design is given. The following material shows how some of these concerns were addressed – by reference to the existing technology base, to operational experience at LAMPF and other accelerator facilities worldwide, and to performance estimates based on beam simulations and other criteria.

Even though the information and projections developed in the APT study are convincing, a high-power cw engineering demonstration of the accelerator front end would still be very important to address systems integration and control questions.

Accelerator Technical Issues

- **Beam loss**
  - Degrades accelerator component performance
  - Produces activation levels that complicate maintenance
- **Power conversion efficiency**
  - Development of a high-efficiency rf tube will significantly reduce capital and operating costs.
- **Accelerator availability**
  - Component lifetime
  - Ease and speed of maintenance
- **Integrated cw operation**
  - Individual components have functioned near ATW specifications
  - An integrated high power cw system has not operated at ATW specifications

Assessment of Beam-Loss Threat in APT CCL

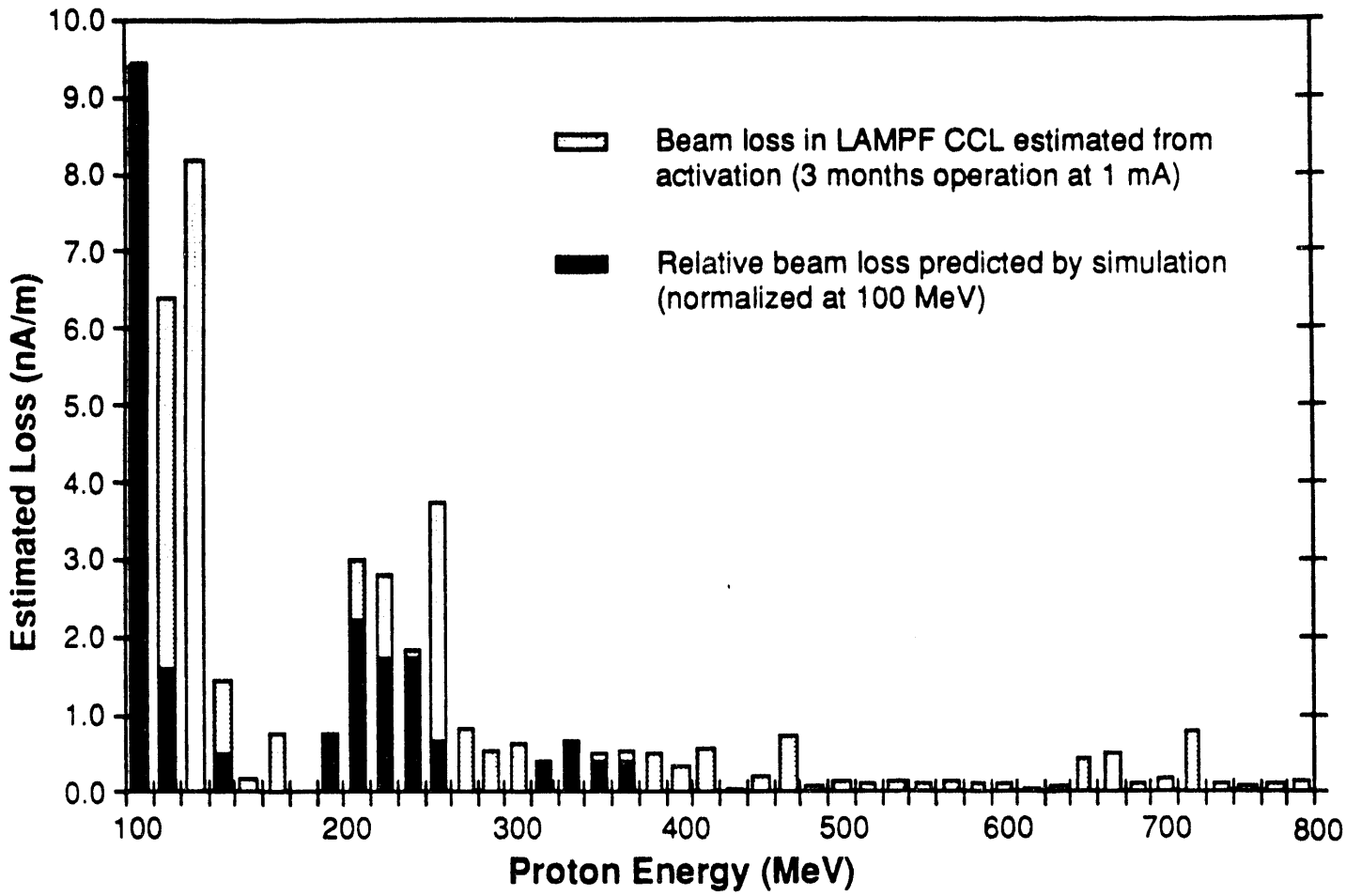
The issue of beam loss in the APT coupled-cavity linac was addressed by combining extrapolations from LAMPF beam loss data and halo simulations using a calibrated LAMPF calculational model. The figure shows a comparison of estimated beam loss in the LAMPF CCL from activation data (lightly-shaded histograms) with the relative loss predicted by simulations (black histograms).

The beam loss estimate is based on activation measurements taken along the CCL after shutdown from prolonged (three months) operation at an average current of 1 mA, and is computed using the energy-dependent thick target yield of neutrons from proton interactions with copper. It is seen that over most of the CCL length, fractional beam losses are $< 2 \times 10^{-7}$/m, except for two high-loss regions. The high beam loss just downstream from 100 MeV point can be explained by the factor-of-4 jump in accelerating-structure frequency occuring at that location, which results in a corresponding reduction of the longitudinal acceptance. The high loss zone just beyond 200 MeV is explained by the x 2 increase in quadrupole spacing that occurs there, which results in a corresponding reduction in transverse acceptance. The APT design avoids such drastic acceptance transitions.

A complete end-to-end simulation of LAMPF using "as-operated" parameter values, was carried out. This simulation correctiy predicted the measured beam emittance values and the known beam loss locations and somewhat overstated the loss magnitudes. A derived beam-halo model produced a simulated relative loss pattern indicated by the black histograms, showing rough agreement with the measured loss pattern. Using this model for the APT CCL it is possible to project that fractional losses in that machine would be two orders of magnitude lower than in LAMPF, providing an acceptable activation level for hands-on maintenance. This result is due mainly to the very large ratio of aperture to rms beam size in the APT design (15 to 20 in APT versus 6.3 in LAMPF), but also benefits from the minimal beam halo achieved by careful attention to matching and the absence of sharp acceptance transitions in the CCL.

The conclusion of these studies is that hands-on maintenance can probably be achieved for most of the APT accelerator, except for a few hot spots. Well-developed remote maintenance techniques can be used to service these locations.

LAMPF Operations + Simulation Results Provide Assessment of Beam Loss Threat



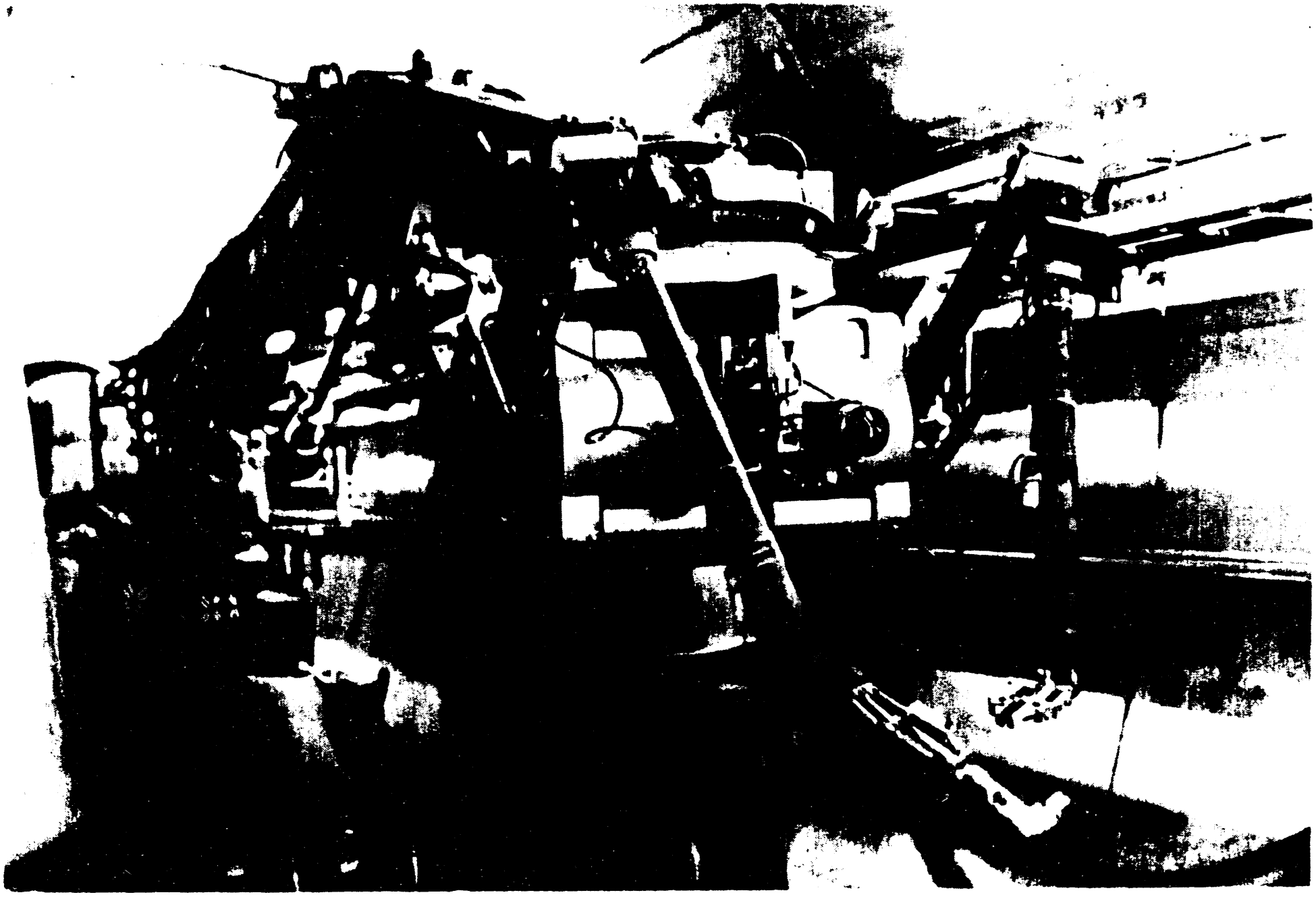
- Fractional beam loss is $< 2 \times 10^{-7}$ /m for most of CCL.
- High loss locations are downstream from longitudinal and transverse acceptance transitions. ATW design avoids such transitions.
- LAMPF simulation predicts loss location; overstates loss magnitudes.
- ATW aperture-to-beam-size ratio is much larger than in LAMPF (20 vs 6.3).
- ATW halo simulation shows much lower fractional losses than LAMPF.

Conclusion

ATW can achieve conditions needed for "hands-on" maintenance.

Remote Maintenance Systems

Master-slave remote maintenance machines have been developed specifically for servicing highly activated accelerator and experimental area components at several nuclear research facilities around the world, and have reached a high level of sophistication. Most of the design and operating experience has been concentrated at LAMPF, where portable manipulators are used to maintain the very radioactive production targets in the experimental area as well as the main beam stop, and at CERN, where a self-propelled machine is used to perform remote maintenance in the SPS (Super Proton Synchrotron) beam channels. The CERN master-slave-servomanipulator (MSSM) system, called MANTIS, is pictured here. The system utilizes servo arms with force reflection, i.e., "feel" that, along with television eyes and microphones, provides the remote operator with a sense of presence. All conceivable operations such as grinding, cutting, welding, soldering, seal replacement, and component replacement can be achieved with this equipment. The operator is connected electrically via cables to the MSSM equipment. The MANTIS is self-mobile and can "find" its target. It should be noted, however, that although most manual functions can be performed with this equipment, its remote nature requires that a factor of five to ten more time must be allowed for a given job compared with hands-on operations.



CW Klystron Lifetime Experience

Klystron lifetime is determined by many factors, but the cathode lifetime is by far the dominant one. Although high power is generally detrimental to cathode reliability, the klystron vendors believe that a 50,000 to 100,000 hour life is achievable in a 1 to 1.5 MW klystron. In addition, extensive instrumentation on the klystron can detect most wearout failures before they happen, allowing replacement of the klystron during regular maintenance periods without loss of running time.

Current experience with high power cw klystrons is limited at this point. The PEP ring at SLAC has twelve 0.5-MW cw klystrons at 353 MHz. Three of the original units (installed in 1977) are still operating after 60,000 hours of operation. One design fault and one materials fault have been corrected, and the second generation of these klystrons is expected to last much longer. The TRISTAN ring at KEK in Japan now has over 20 cw klystrons with output power capabilities between 0.8 and 1.2 MW at 508 MHz. This facility is still relatively new, but one klystron has accumulated over 12,000 hours. The PETRA ring at DESY in Germany has had a number of 500-MHz cw klystrons with output powers close to 1 MW in operation since 1978. The information available on these tubes, though sketchy, supports the general expectation of 50,000 hour lifetimes.

LAMPF Klystron Lifetime Experience

Parameters: 805 MHz, 1.25 MW (peak power)
12% duty (1 ms pulses, 120 pps)

	VARIAN 862A	LITTON 5120A
Number of klystrons	70	26
Average operating hours		
• High-voltage on	31 K	9.2 K
• Filament on	39 K	11.8 K
Long-life performance	**20 tubes with > 75 K hours**	**5 tubes with > 50 K hours**

Accelerator Availability

This table is a compilation of operational parameters from a large number of accelerators used in nuclear research around the world. These machines all function as particle factories for a large user community, and beam availability is an important measure of their performance. The right-hand column lists the claimed beam availability at these facilities as a fraction of the scheduled beam time. While it does not take into account the planned beam-off time, which is usually governed by operating budget constraints, this availability data does give a measure of the reliability of modern particle accelerators. LAMPF is the accelerator closest to APT parameters and operating regime, and has had an overall availabilty > 85% for many years.

Accelerator Availability Data

Accelerator	First Beam	Facility	Peak Current (mA)	Pulse Length (μs)	Duty Cycle (%)	Ann. Hours	% Avail.
50-MeV H^+ linac	1978	CERN	140	110	0.01	7000	99
20-MeV H^+ linac	1969	Saclay	20	500	0.1	5000	98
200-MeV H^- linac	1970	BNL	25	500	0.25	5000	95
200-MeV H^- linac	1970	FNAL	35	60	0.1	8000	97
100-MeV H^+ linac	1967	Serpukhov	100	100	0.01	5000	99
800-MeV H^+ linac	1972	LAMPF	17	500	6.0	4500	85
25-GeV e^- linac	1966	SLAC	50	2.7	0.03	5300	90
40-MeV H^- linac	1974	KEK	10	80	0.16	4000	95
70-MeV H^- linac	1983	Rutherford	12	500	2.5	5000	
400-GeV H^+ synchrotron	1976	CERN				5500	85
26-GeV H^+ synchrotron	1959	CERN				6300	96
12-GeV H^+ synchrotron	1976	KEK				4200	95
7.5-GeV e^- synchrotron	1964	DESY				7500	95
17-MeV e^- linac (RTM)	1986	NIST	0.5	CW	100		
0.6-MeV H^+ RFQ	1988	CRNL	50	CW	100		
2.0-MeV H_2^+ RFQ	1982	FMIT	50	CW	100		

Most data from Catalogue of High-Energy Accelerators, Tsukuba, Japan, July, 1989.
Availability data from 1980 Catalogue.

The ERAB Report on APT Established the Credibility of a High-Power Accelerator

This quote from the Energy Research Advisory Board Report on Accelerator Production of Tritium indicates the summary evaluation of the accelerator design concept.

ERAB Report on APT (2/90) Established the Credibility of a High-Power Accelerator

"The continuous-wave RF linac approach for APT is technically sound. While an integrated accelerator system has not been built and operated at APT conditions, the accelerator feasibility and engineering development issues could be resolved with an adequate research, component and systems development, and engineering demonstration program."

Accelerator for Near-Term Application (Technetium Burner)

The proton current requirement for a transmuter that could burn the technetium in the accumulated defense wastes within 30 years is about 55 mA, average. In order to meet an aggressive implementation schedule and minimize technical risks, the best approach to such a machine might be along the lines indicated in the figure. In cw operation, the peak current carried by the CCL is only 55 mA, equivalent to the same charge per bunch as in LAMPF. Thus, the beam dynamics for such a linac would be within a well understood regime, and beam losses and other performance factors could be predicted with very high confidence. Technical uncertainties would be minimal. While cw operation will not yield the highest electrical efficiency for this case, with design optimization RF efficiencies approaching 60% may be realizable.

This ATW accelerator design approach would permit an aggressive implementation schedule. It should be possible to proceed directly to facility design and construction in a fashion similar to new large accelerator-based research facilities, with a parallel early-start component development program.

Accelerator for Tc - Burner Application

Requirements: 55 mA average current, 1.6 GeV

Use same CW design approach as APT

- Funneled system
- 27.5 mA in each RFQ, DTL
- 55 mA cw in CCL; same charge/bunch as LAMPF

RF efficiency not as high as APT, but could reach 0.58

- CCL RF power

Beam	88 MW
Structure	63 MW
Total	151 MW

- AC power for CCL = 252 MW

Low technical risk approach

Could proceed directly to design and implementation, with parallel component R&D (rf tube, injector)

High-Efficiency Accelerator for Advanced Applications (Energy Production)

For energy production applications, the accelerator design would focus on minimizing capital and operating costs and maximizing electrical efficiency. With this design emphasis a pulsed RF linac is the appropriate solution, with the peak beam current as high as practical. The figure shows one possible set of parameters for such a system, using the same 250-mA peak proton current as used in the APT design. RF efficiencies could be 80% or higher; for a 30-mA linac, this would lead to a total ac power requirement for the CCL of about 100 MW. We have assumed a macropulse rate of 120 per second, with a pulse length of 1 ms, but other combinations are possible.

A high-peak-current pulsed linac involves somewhat more technical risk than a cw approach to attaining the same average current, because of uncertainties regarding beam losses and other factors arising from high space-charge levels. An integrated high-current front end demonstration may be needed before commitment to final design and construction of the facility.

The ultimate in high-efficiency and low-beam-loss accelerating systems for accelerator-driven energy production applications may be a superconducting RF linac. While there is currently no experience with high-current ion beam acceleration by superconducting RF cavities, and cavity designs have not been developed for much of the required particle velocity range (β = 0.2 to 0.9), superconducting RF technology continues to mature and in the long term could provide a favorable design option.

High-Efficiency Accelerator for Energy-Production Application

Requirements: 30 mA average current, 1.6 GeV

Use pulsed RF linac approach; same peak current as APT

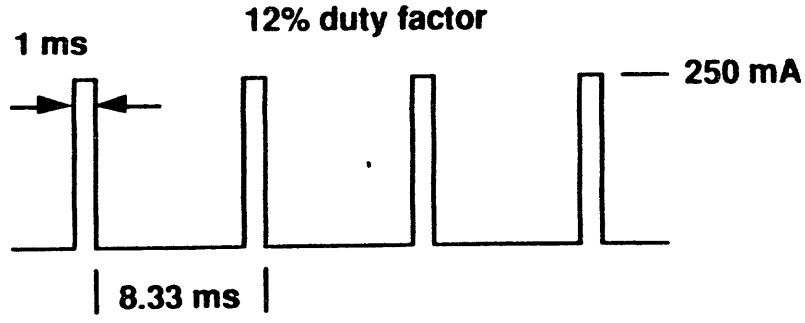
- Funneled system
- 125 mA (peak) in each RFQ, DTL
- 250 mA (peak) in CCL; 4.5 x charge/bunch in LAMPF
- 12% duty factor; 120 pps, 1-ms pulses

RF efficiency can be > 0.80

- CCL RF power

Beam	48 MW
Structure	12 MW
Total	60 MW

- AC power for CCL = 100 MW



Higher technical risk. May need integrated high-current front end demonstration before commitment to final design and construction.

Superconducting RF linac may provide ultimate in high-efficiency low-beam-loss accelerating system for far term applications.

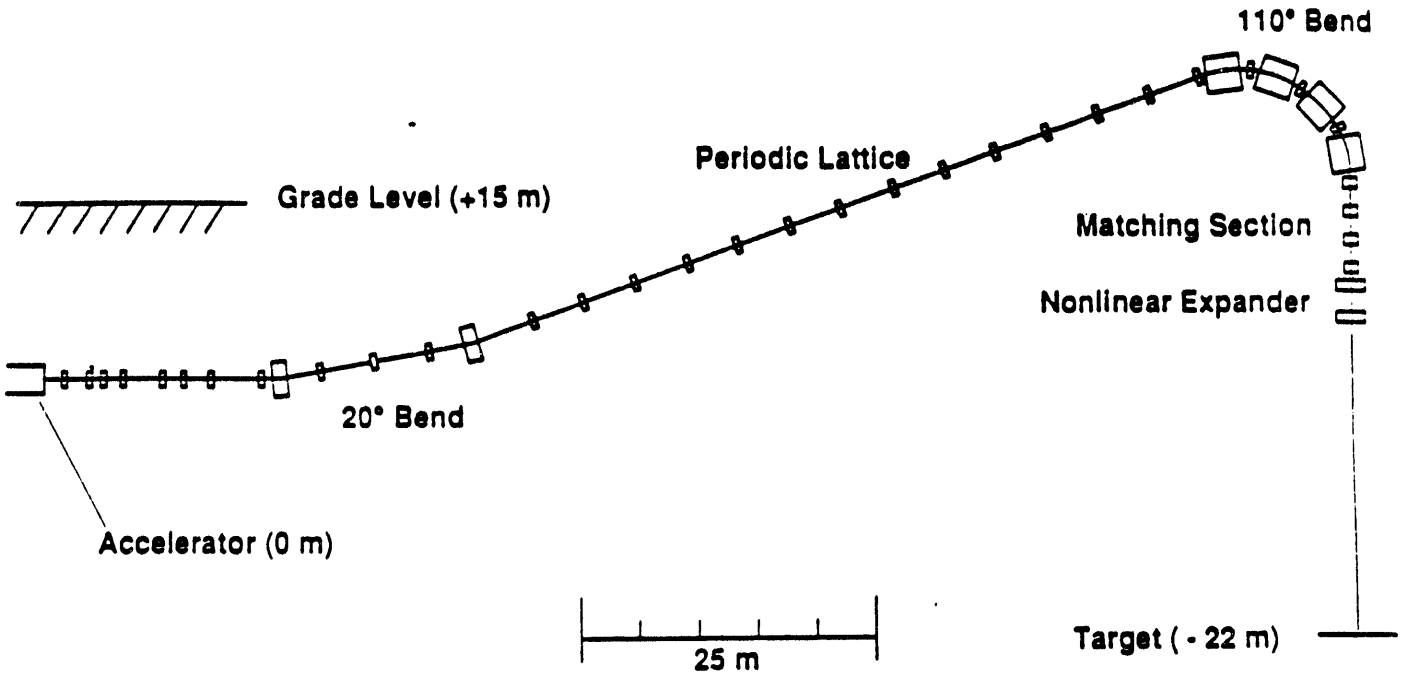
High-Energy Beam Transport to ATW Target

The upper part of the figure shows a schematic elevation view of the high-energy beam transport (HEBT) system that conveys the 1.6-GeV proton beam from the end of the linac to the ATW target. Because the beam is required to be vertically incident on the target a series of bends in the vertical plane is necessary. The design shows a possible configuration in which the beamline first bends upward, in order to reduce the depth underground at which the ATW target/blanket facility is placed. The beamline consists of a periodic transport lattice of quadrupoles, 20° and 110° achromatic bends, a matching section, and an expansion system that employs nonlinear magnetic elements (octupoles).

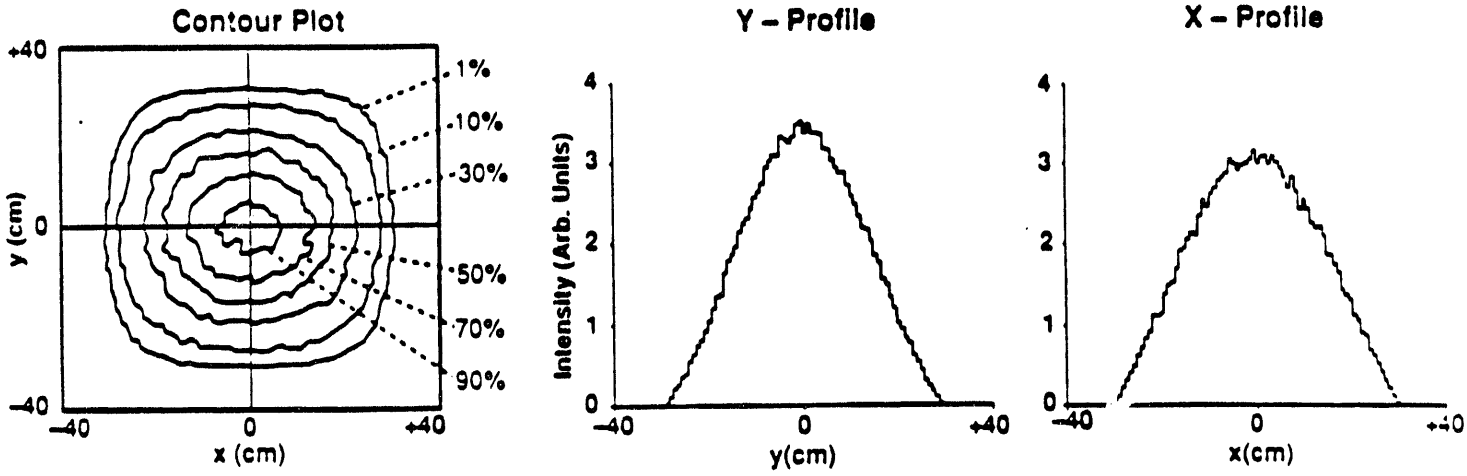
The function of the non-linear expander is to produce a large-area beam cross section at the target , and alter the beam intensity distribution from Gaussian to parabolic, folding in the tails beyond the 3σ points. Removal of the beam tails reduces direct proton damage to the walls of the target containment vessel.

The lower part of the figure shows the calculated x and y beam profiles produced by the nonlinear expander at the ATW target, along with an x-y contour plot of the distribution.

Elevation View of Beam Transport from Accelerator to Target



Intensity Distribution at Target from Nonlinear Optical Expansion



Research and Development Needs for an ATW Accelerator

The table outlines near term and far term development plans for a high-power ATW accelerator matched to the technetium-burning application.

Accelerator Research & Development Needs

Near Term (2 years)

- Design tradeoff studies; parameter selection
- High-efficiency, low-cost, 1 to 5 MW RF source at 700 MHz
- High-availability, high-current, low-emittance proton injector
- Permanent-magnet radiation damage measurements
- Low-cost CCL structure fabrication methods

Longer Term (4 years)

- Continue high-efficiency RF tube development
- Integrated cw high-power front end demonstration

Proceed with Technetium burner implementation within three to four years

Summary

- Proton linac technology has reached the point that cw machine designs in the 50 to 250 mA range at 1 to 2 GeV can be projected with a high degree of confidence.
- While there are no significant physics issues, a machine of this class nevertheless represents a major technical challenge.
- With a suitable component development program, and an integrated front end cw demonstration, the implementation of a high power linac for ATW should be straightforward.
- The average beam current requirement for a Tc burner early application is much less than that for the full-up APT design and may permit a more aggressive schedule.

INTENSE THERMAL NEUTRON SOURCE

ATW Neutron Production

Paul Lisowski
Physics Division

ATW Neutron Production

- Why High Flux is Important
- Spallation Physics
- Separated Target-Blanket Concept
- Spallation Source Operating Experience
- Summary

BURN UP OF RADIOACTIVE NUCLEI IN A NEUTRON FLUX

In a neutron flux the absorption cross section enhances the transmutation rate beyond that of normal radioactive decay.

For decay alone, the decrease in population of an isotopic species is equal to the number of nuclei multiplied by the decay constant:

$$dN/dt = - N \cdot \lambda.$$

In a neutron flux, the rate of decrease is changed by the product of the flux, the number of nuclei present, and the absorption cross section:

$$dN/dt = - N \cdot \lambda - N \cdot \Phi \cdot \sigma_a.$$

The solution of this equation is exponential decay with the decay constant re-defined to include the flux and absorption cross section.

$$N(t) = No.\ e^{-\lambda' t}$$

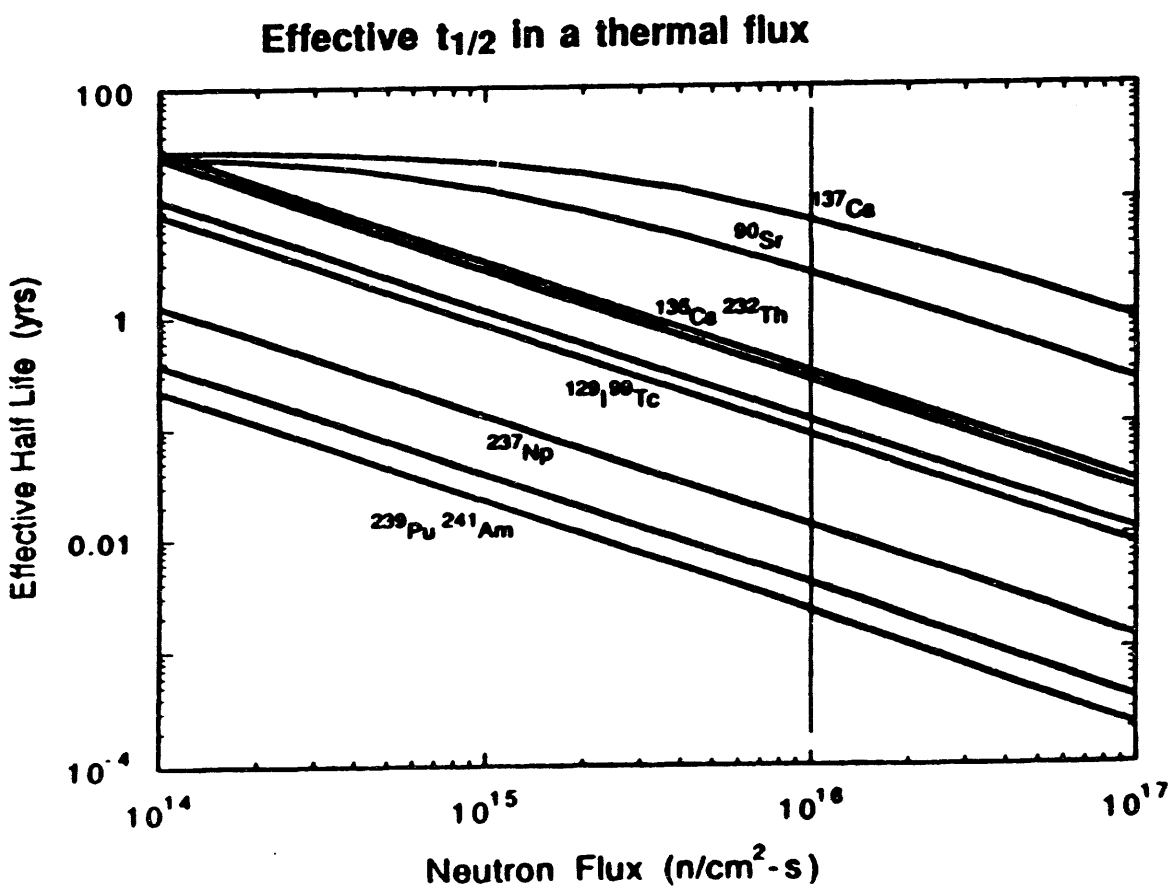
$$\text{with} \quad \lambda' = \lambda + \Phi \cdot \sigma_a$$

For fission products and threshold fissioning actinides, this effect is largest at thermal energies where the capture cross sections are highest. As examples, ^{99}Tc and ^{129}I, with capture cross sections of about 20 b, have their half lives reduced from 213,000 years and 16,000,000 years to effective half-lives of less than one year.

Burn Up Radioactive Nuclei in a Neutron Flux

In a neutron flux: $dN/dt = - N \cdot \lambda - N \cdot \Phi \cdot \sigma a$

Solution: $N(t) = No \cdot e^{-\lambda' t}$ **with:** $\lambda' = \lambda + \Phi \cdot \sigma a$



Largest effect occurs where σ_a is greatest.

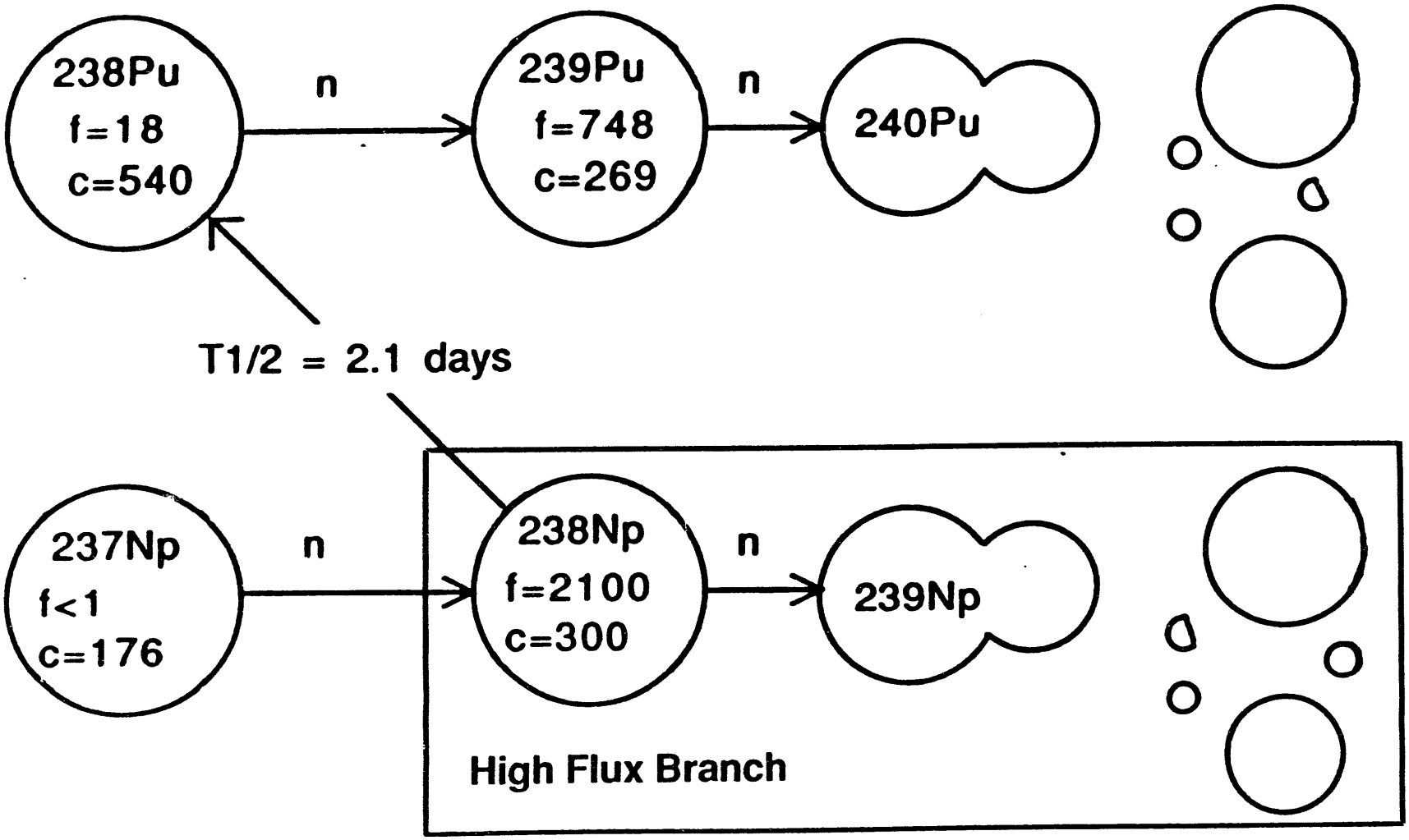
HIGH FLUX FISSION OF HIGHER ACTINIDES

Because many of the actinide waste nuclei are threshold fissioners, conventional wisdom is that a fast neutron flux is required to effectively fission materials such as ^{237}Np and ^{241}Am. That is true at neutron fluxes lower than can be obtained with a spallation neutron source.

At fluxes of 10^{14} n/cm^2-s, roughly those available in a PWR, threshold fission materials act as a poison for the following reason: the fission cross section for ^{237}Np is so small and the capture cross section so high at thermal energies that an interaction with a neutron will lead to formation of ^{238}Np with high probability. At 10^{14} n/cm^2-s, the likelihood that another neutron will interact with ^{238}Np with a 2.1 day halflife before it decays to ^{238}Pu is small. Once the decay to ^{238}Pu occurs, two additional neutron interactions are required before fission occurs. In this scenario, it takes three neutrons to destroy one ^{237}Np nucleus, but only about 2.7 are released. In this instance, ^{237}Np acts as a net neutron absorber or poison.

As the thermal neutron flux increases, an entirely different process can take place. Following initial capture, the ^{238}Np nucleus has a much higher probability of interacting with a neutron before it can decay to ^{238}Pu. Because the thermal fission cross section is 2100 b, most of the time fission occurs with the release of 2.7 neutrons at a cost of only 2, for a net gain of 0.7 neutrons. Although it is necessary to follow the possible capture and fission branches further to complete the proof, the result is that at high thermal fluxes ^{237}Np acts as a fuel instead of a poison.

High Flux Fission of Higher Actinides

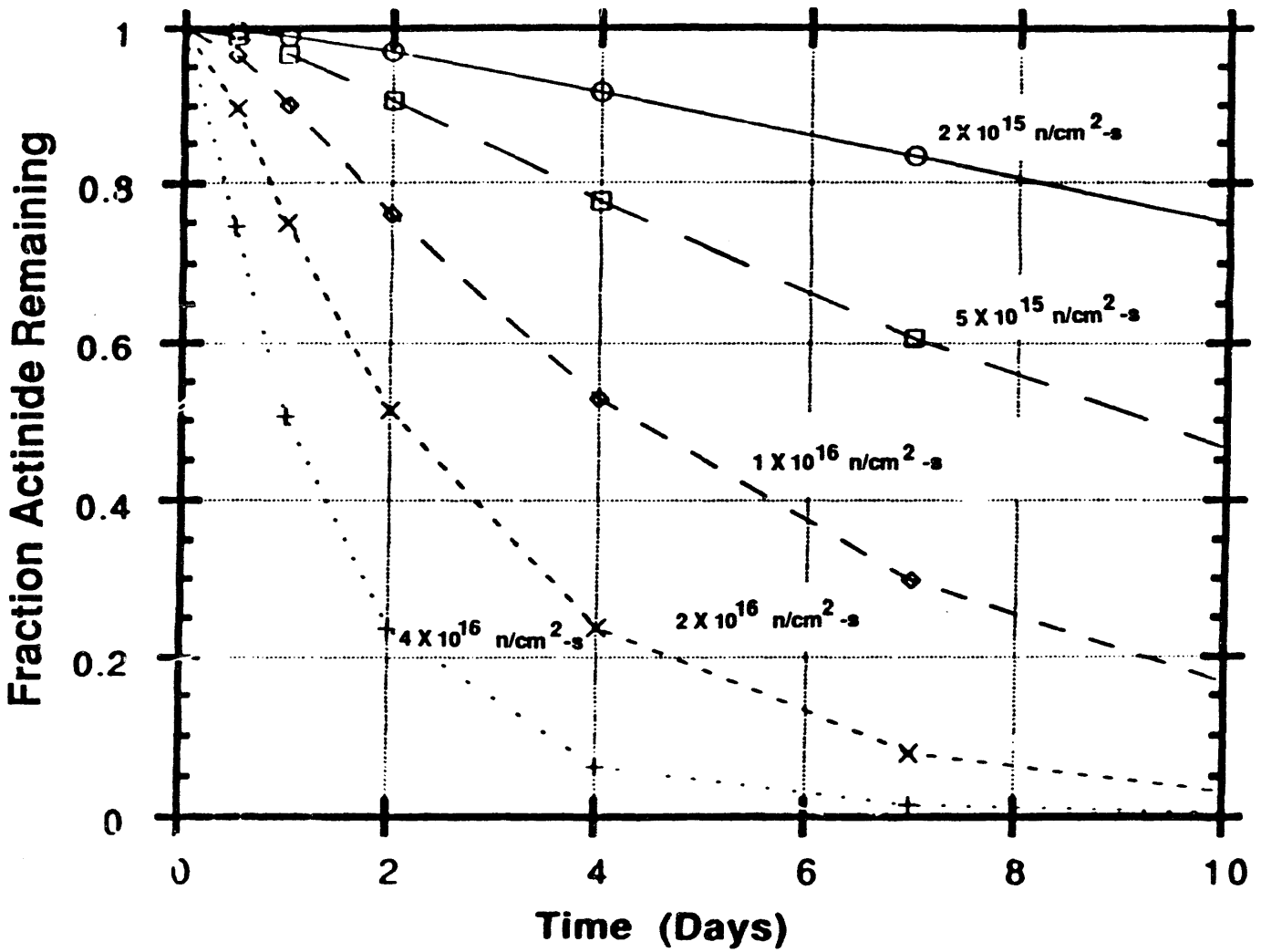


BURN-UP OF ^{237}Np BY FISSION OF ^{238}NP

As an example of the thermal fission of ^{237}Np, we have calculated the inventory of actinides and fission products during a ten-day irradiation at five high, constant flux levels. The CINDER-2 code, designed for light-water reactor burn-up calculations, was used after modification to include fission contributions of all of the actinides produced. This calculation follows fission and capture chains through ^{246}Cm and indicates that for levels between 2 and 5 x 10^{15} n/cm^2-s, a net neutron gain occurs. The result is that ^{237}Np is very rapidly consumed as a fuel.

It was this set of results that convinced us that a conventional fuel rod structure would be impractical for threshold actinide burning because of the frequent change-out required. A flowing system, similar to that used in the Molten Salt Breeder Reactor which was developed at Oak Ridge, began to emerge as the most practical solution. Furthermore, fission heating calculations showed that a very dilute concentration of ^{237}Np would be required. At about that time we solved the appropriate set of time-dependent , infinite-medium coupled differential equations for actinide populations and found that equilibrium concentrations of ^{237}Np at 10^{-4} normal density were approximately correct.

Burn-Up of ^{237} Np by Fission of ^{238}Np



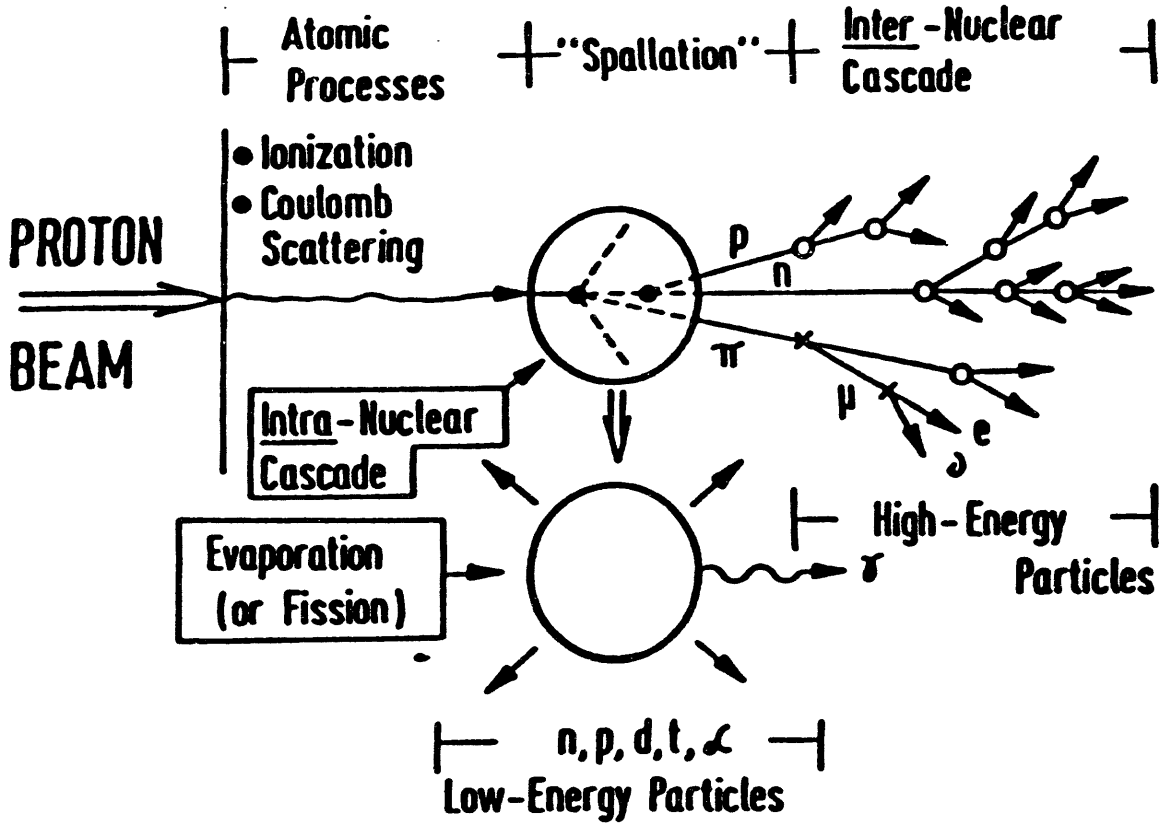
BEAM-TARGET INTERACTION PHENOMENOLOGY

When an energetic proton enters a target it interacts first through atomic processes and begins to lose energy through ionization. Multiple Coulomb scattering broadens the beam as it penetrates the target. Although all possible nuclear reactions such as capture, scattering, and fission can occur at an energy of 1600 MeV, the most likely is spallation. In spallation the proton penetrates a nucleus and interacts with the constituents. The result is that additional energetic protons, neutrons and pions are ejected and continue to penetrate the target material. Terminology for the interactions occurring within the nucleus is that an intra-nuclear cascade occurs. Further interaction of the energetic secondaries results in a 'shower' of particles with decreasing energy in the form of an inter-nuclear cascade. After particle emission the excitation energy of the original nucleus within which the intra-nuclear cascade occured is distributed among the remaining nucleons and dissipated by evaporating low energy particles. These particles are primarily protons and neutrons, and with lower probability deuterons, tritons, and alpha particles. Depending on the nucleus under consideration, competition between evaporation and fission results in fission 10-20% of the time.

If the heavy metal target is surrounded by hydrogenous material as a moderator, neutrons escaping the target into the moderator continue to lose energy through scattering until their energies are comparable to that of gas molecules at room temperature. It is those 'thermal' neutrons that the ATW concept uses to transmute radioactive elements.

Over the past ten years, a major effort has been the development of a code system to calculate energetic proton and neutron interactions. The system was based on a version of the Oak Ridge National Laboratory Monte Carlo code HETC, or High Energy Transport Code. This package, the Los Alamos High Energy Transport Code System (LAHET), utilizes the basic intranuclear cascade model of HETC, with significantly enhanced physics and a coupling to the continuous energy neutron photon Monte Carlo code MCNP. For parametric studies, the complete LAHET - MCNP computation sequence is used to generate source spectra which are used as input to one and two dimensional discrete-ordinate transport codes for reactor and criticality computations.

Beam-Target Interaction Phenomenology



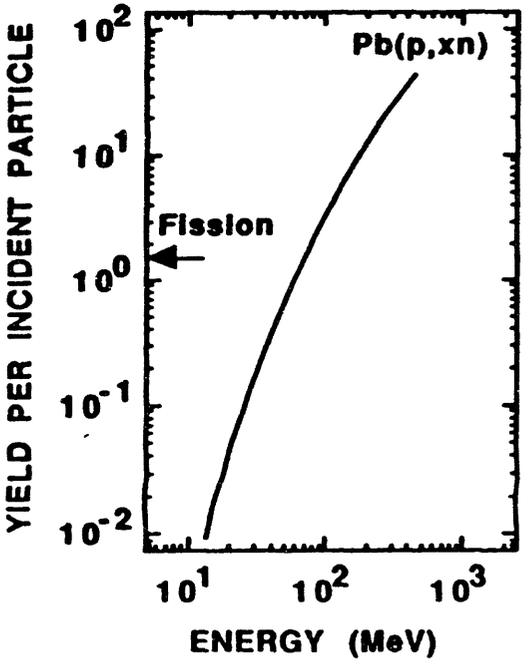
SPALLATION HAS ADVANTAGES FOR HIGH FLUX NEUTRON PRODUCTION

Spallation is one of the most efficient neutron sources available. At energies above 500 MeV, the neutron yield increases nearly in proportion to the bombarding energy. The calculations shown here were redrawn from a paper by Lone et al. (NIM A236(1987)135-142) which compared several different options for producing neutrons. Although the calculations shown here were made for smaller diameter targets than we are considering for ATW, the main features are the same. We get about 50 neutrons per proton, and except for a small leakage, all of those neutrons are available to produce transmutation. In contrast, in a reactor about 2.6 neutrons are produced per fission. One of those must go to sustain the reaction, leaving about 1.6 neutrons.

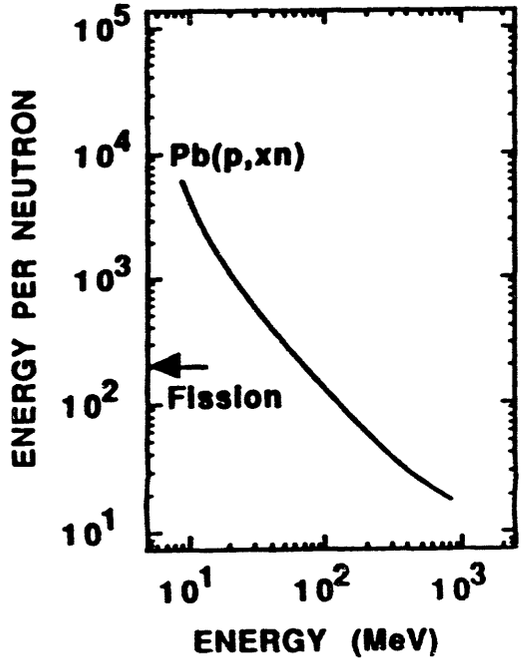
One important limitation on the flux attainable inside a reactor core is the practical need to remove about 200 MeV of that per fission. For spallation system, the energy deposited in the target is less, with only about 32 MeV per neutron produced. On average, about 70% of the proton beam energy is deposited in the heavy metal target.

Spallation has Advantages for High Flux Neutron Production

- High neutron yield per incident particle. We get about 50 n/p at 1600 MeV. from spallation. Fission provides 1.6 n/f.



- Low heat per neutron produced. About 32 MeV/n for spallation and ~200 MeV/n



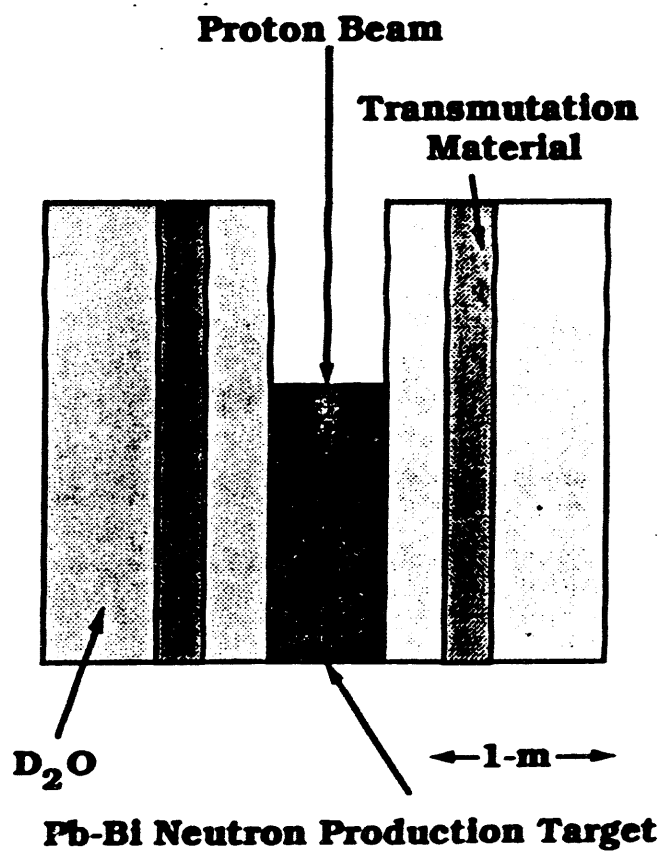
SEPARATED TARGET-MODERATOR CONFIGURATION

In order to produce a thermal neutron spectrum, moderation by one of the hydrogen isotopes is very efficient. By surrounding a central heavy metal spallation source with a deuterated or 'heavy' water moderator, most of the neutrons from the target will be moderated. Our initial choice of a central, flowing Pb-Bi eutectic target was based on considerable research done at Chalk River Canada for a spallation source target, and at Juelich for the German Spallation Neutron Source (SNQ). In our system modest flow rates of the Pb-Bi can achieve adequate heat dissipation with surface Pb-Bi temperatures low enough that vapor pressure poses no vacuum problem. Hence, no entrance window between the accelerator and the target is required. In addition, we believe that the build-up of spallation poisons in the target material will be small enough (~1-2% of the total mass after 30 years of operation) that the target neutronics will not be seriously degraded.

By using a moderation/transmutation region separate from the production target and by taking advantage of the thermal properties of the Pb-Bi, we can completely avoid a pressurized system. The choice of D_2O with its large moderation length and low absorption cross section allows us to have a large volume high flux region in which to transmute radioisotopes or to breed fissionable material. In addition, the neutron flux profile is highest near the Pb-Bi target so that there are optimized locations for different fission products rather than a single compromise location. One advantage of this feature is that substantial amounts of low cross section elements such as ^{90}Sr can be placed close to the target with little effect on the spectrum leaking past.

Separated Target-Moderator Configuration

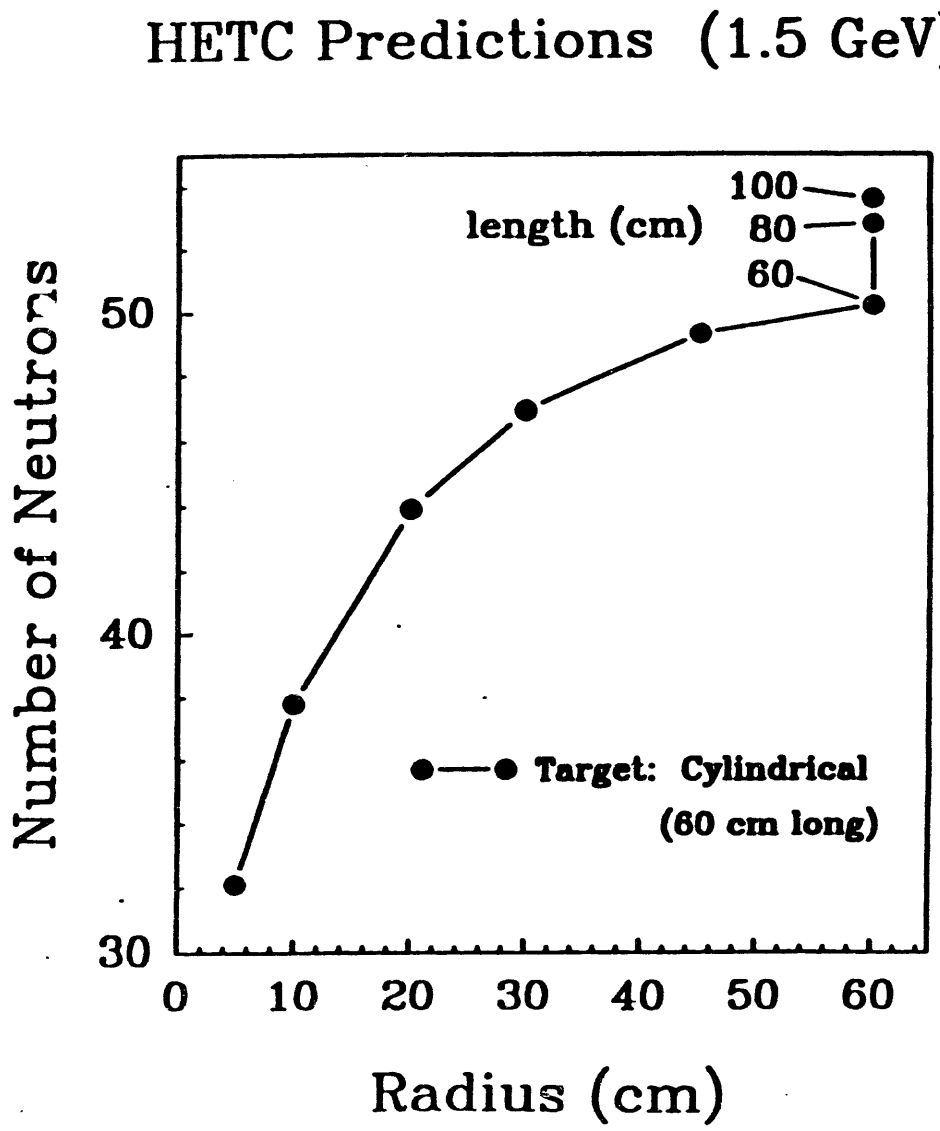
- **Flowing Pb-Bi neutron production target**
  - Based on SNQ design and work at Chalk River
  - Can achieve large heat dissipation
  - Windowless operation
  - Recirculating Pb-Bi target lasts facility lifetime
- **Separate moderation/transmutation region**
  - D_2O provides low-loss, efficient moderation
  - Working volume isolated from n-production region
  - Optimized flux profile for burning



CALCULATIONS FOR LARGE TARGET SYSTEMS ARE HIGHLY GEOMETRY DEPENDENT

An important indicator of how well an accelerator driven system can perform is the total neutron production. This graph was taken from a study performed by T. Ward at Brookhaven National Laboratory which investigated the neutron leakage as a function of target radius and length, and integrates over all neutron energies and angles. The range of a 1500 MeV proton beam in Pb is approximately 100 cm - the yield curve shows a slow increase up to that length. As the inter-nuclear shower progresses, protons and reaction products fan out through the Pb target. Neutron yield increases with increased radius because more of those particles are being contained. Since the target diameter determines the leakage out the top of the assembly, the need to capture most of the neutron production in the moderator will require a trade-off against gains from increased radius. As the target thickness and length are increased, the neutron leakage will be more dependent on transport phenomena and self shielding. Our calculations indicate that about 10% of the yield is generated from multiplication in the Pb-Bi from (n,xn) reactions with high-energy neutrons. In addition, the neutron production along the length of the target is very non-linear, with the most intense region near the top of the target.

Calculations for Large Target Systems are Highly Geometry Dependent



LAHET CALCULATIONAL RESULTS

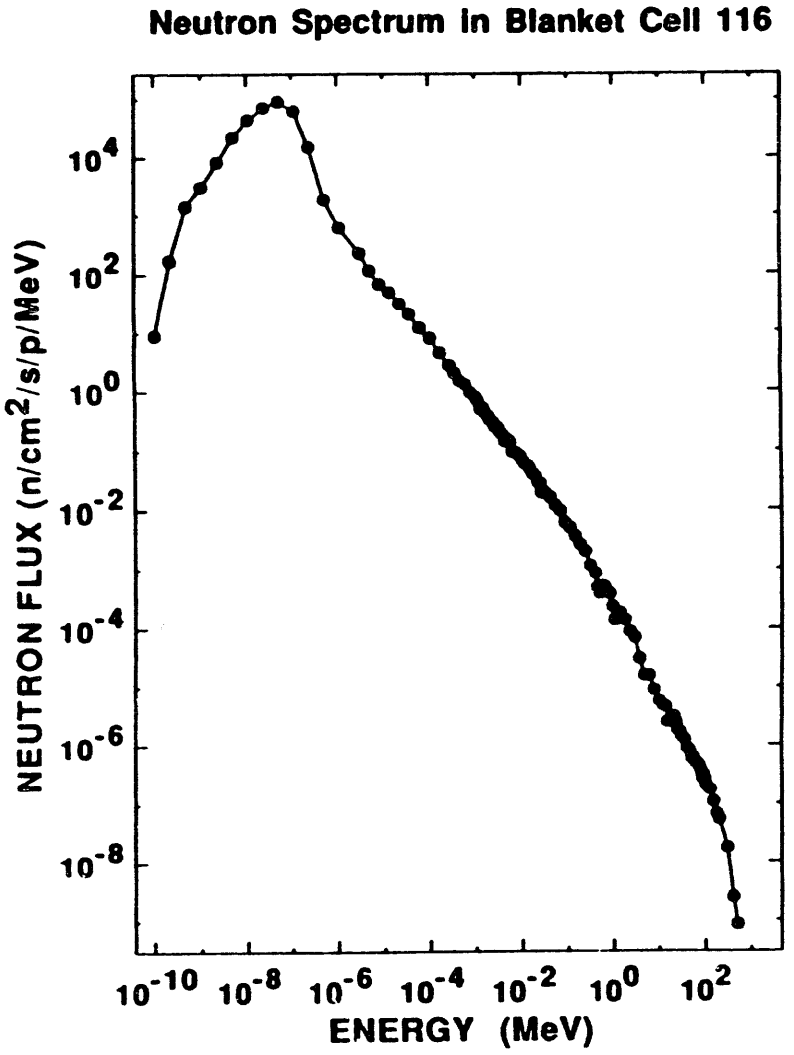
The LAHET code system allows full three-dimensional geometry representations of the target-blanket. The geometry is defined by mathematically describing the target-blanket make-up in terms of connected imaginary 'cells'. It is possible to produce very detailed information about most of the proton and neutron interactions. Examples include information about neutron and proton leakage, neutron loss and gain, neutron fluxes, heat generated, gas production, and residual nucleus production.

For the design that we chose for the defense waste Tc burner concept, the target was taken to be 50-cm in diameter and 125-cm long. Among other things, we confirm that the neutron spectrum becomes highly thermalized within the blanket, that parasitic leakage and capture are acceptable, and that the neutron flux in the wall containing the Pb-Bi target are large, but tractable.

Code predictions such as these are able to predict total neutron leakage to about 25% accuracy based on comparison with measurements. In fact, recent changes in the code benchmarked by Los Alamos neutron spectral measurements may have improved the situation considerably.

LAHET Calculational Results

- Neutron production 55 n/p
- Neutron spectrum in blanket is highly thermalized.
- Parasitic capture and leakage acceptable.
- Neutron flux levels at wall are tractable.



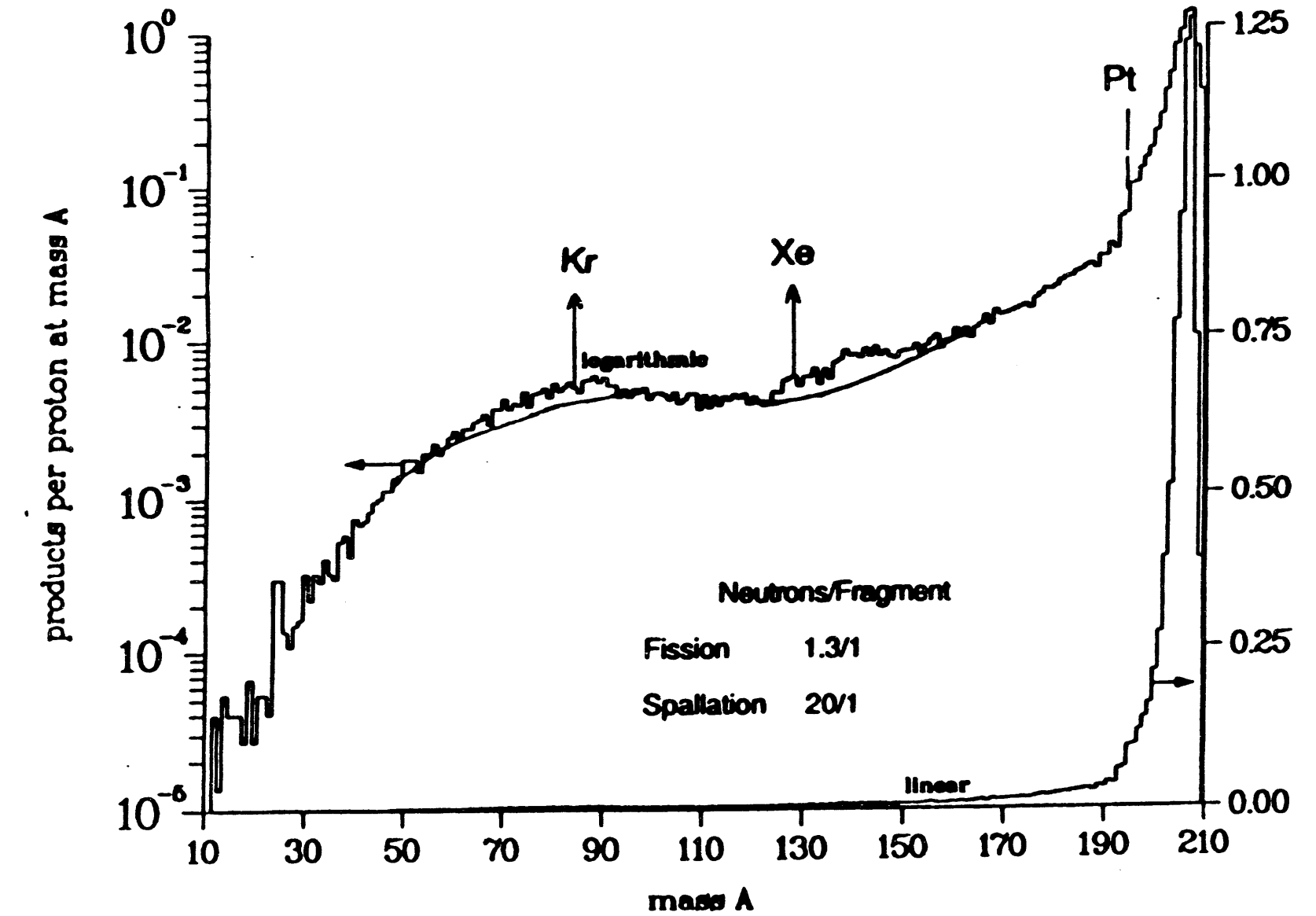
DISTRIBUTION OF SPALLATION AND FISSION PRODUCTS PRODUCED BY 1.6 GeV PROTONS ON Pb-Bi TARGET

This graph shows a plot of products produced at each mass as a function of the mass. This plot was generated from a LAHET calculation for 1600 MeV protons incident on our Pb-Bi target.

The plot is complicated by the fact that there are both linear and logarithmic axes. Looking at the linear data on the right side of the graph we see that most of the spallation products are produced within a few mass units of the target. Those materials in the Pb-Bi are nearly as effective as a neutron producer as the original target itself. If we take the material heavier than Pt as roughly equivalent in neutron yield to Pb-Bi, then the effective number of neutrons per fragment for spallation is about 20/1, considerably lower than for fission, where the number is about 1.3/1. Furthermore, light nucleus production is considerably lower than in fission. There is fission induced by high-energy protons and neutrons in the Pb-Bi. Evidence for that can be seen in the logarithmetic representation in the two small bulges in the yield curve near Kr and Xe.

The fact that stable or short-lived noble gas is produced in the spallation process indicates a potential mechanism for removing contaminants. The Pb-Bi is operated at temperatures between about 200 and 400 degrees in a liquid state. Because the noble gas can freely evolve and it can be captured in holding tanks until all short-lived radioactive species decay. After a long period of operation, we believe that spallation product production and spallation product burn-up will reach a 'quasi-equilibrium' with the lightest spallation products capturing up to Kr and being released, heavier ones capturing to Xe and being released, and others continuing to gain mass and moving towards Pt, which may a reasonable material to consider for chemical removal.

Distribution of Spallation and Fission Products Produced by 1.6 GeV Protons on Pb-Bi Target



NEUTRON DAMAGE IN THE TARGET WALL IS COMPARABLE TO THAT IN PROPOSED OR OPERATING FACILITIES

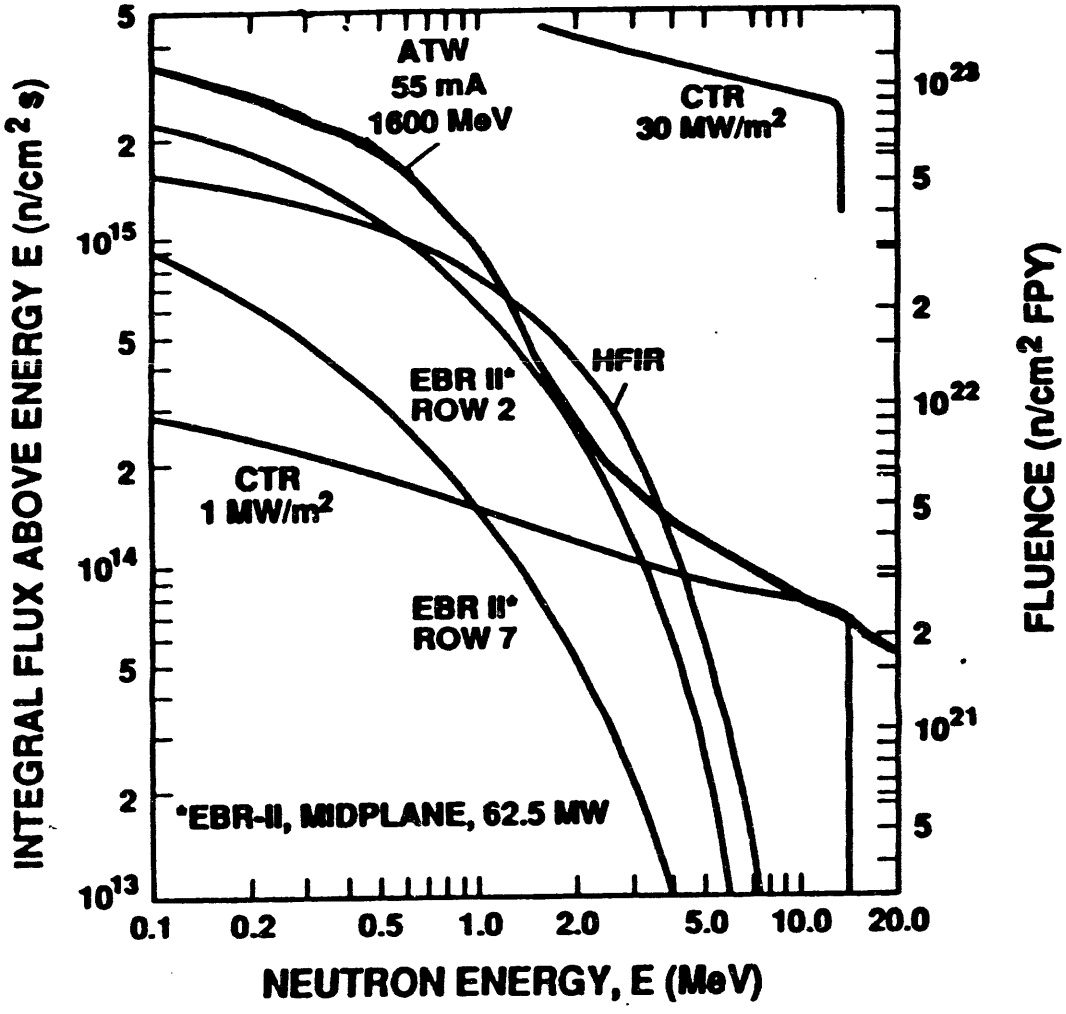
Information about neutron damage in the Pb-Bi container will be one factor in determining what the useful operating lifetime for the ATW system will be. The preliminary results for the container wall shown demonstrate that a 10^{16} n/cm^2-s flux system is comparable to proposed or operating facilities.

The plot shows the integral flux above an energy as a function of neutron energy compared with that at several operating or proposed facilities. This data is for our worst case - the cell that is immediately below the surface of the Pb-Bi target. The right-hand axis shows the neutron fluence in one year of operation.

In the ATW wall, flux levels up to energies of about 5 MeV are comparable with those in operating reactors. Above that energy, the flux shows a plateau extending out to 100 MeV; it is in that region that few damage studies have been performed. Improved designs may mitigate this plateau considerably. First of all, the mechanical loading of the wall in this region is quite low because it is near the top. Therefore, structural degradation is not as important as it would be in a high-pressure system. In addition the damage is quite well localized and may be avoided by a clever mechanical design in which the most heavily irradiated section is actually moved to a lower irradiation region by means of a sliding wall section. A neutronic solution would be preferable.

Comparison of these data with the result for a two times larger diameter target shows that both the 'knee' and the intensity of the integrated flux are reduced and are comparable to the data for HFIR. The reason for this is that the Pb-Bi is itself an efficient moderator; increasing the radial dimension softens the neutron spectrum considerably. Further optimization may reveal that a larger target diameter should be employed near the entrance.

Neutron Damage in the Target Wall is Comparable To That In Proposed/Operating Facilities



WHAT IS THE EXPERIENCE BASE FOR SPALLATION SOURCE OPERATION?

There have been spallation neutron sources in operation world-wide for over 25 years. Much of the information presented here has been obtained from that experience base. Although there are some uncertainties, most of the important trade-offs in optimization of neutron flux have been considered. In addition, modern developments in code design and nuclear data allow a computational treatment of many of the important ES&H issues relating to residual radioactivity, target activation, decay heat, and shielding.

As the next table will show, although there is reasonable expectation that we can operate targets in the range of 40 MW, there are no currently operating sources with beam powers above 1 MW.

Spallation Neutron Source Research Yields Considerable Information for Spallation Source Design and Operation

- **Because of the design of spallation sources for research, many of the trade-offs in optimization of low-energy flux have been considered. The cost and performance minimum indicates that the best energy will be near 1.6 GeV.**

- **Calculational tools have been developed to treat important design issues related to areas such as target activation, decay heat, residual radioactivity, and shielding.**

- ***There are no existing spallation sources with beam power near that required for ATW. Currently operating spallation sources have beam power <1 MW. ATW will likely operate at > 40 MW.***

SPALLATION SOURCE FACILITIES

This table was taken from a presentation by A. Armstrong of SAIC to the Energy Research Advisory Board during their review of the Accelerator Production of Tritium (APT) concept. It compares the operating parameters of spallation sources world-wide.

Los Alamos has been a pioneer in the development of spallation sources, with the second longest operating sources. The Los Alamos Neutron Scattering Center (LANSCE) which operates in the thermal and epithermal energy range holds the record for the highest beam power.

Spallation Neutron Source Facilities

Facility	TRIUMF	LANSCE/WNR	KENS	IPNS	ISIS	SIN	SNQ
Location	Vancouver Canada	Los Alamos, USA	Tsukuba Japan	Argonne USA	Rutherford U.K.	Villingen Switzerland	Germany
Accelerator Type	cyclotron	LINAC	synchrotron	synchrotron	synchrotron	cyclotron	LINAC
Proton Beam Energy (MeV)	500	800	500	500	800	590	1100
Average Beam Current (μA)	140	100	10	14	212	1000	5000
Target Material	Pb	W	W/U238	enriched U	U-238	Pb-Bi	Pb (baseline)
Coolant	None	H_2O	H_2O	H_2O	D_2O	Pb-Bi convection	H_2O
Status	operational 1974	initial operation, 1977; upgrade 1986	operational 1980	operational 1981	operational 1984	start up expected 1990	study

RESEARCH NEEDS

We need additional optimization studies of the target-blanket. Experimental verification of these stdies will impact the design operating regime for the accelerator. In addition to neutron yield, differential information to verify that the neutron spectrum in the wall is as calculated is needed to resolve material lifetime questions. Initial measurements and benchmarks can be conducted at 800 MeV at the Weapons Neutron Research (WNR) Facility using the LAMPF beam. If needed, the equipment developed for those measurements could be moved to some facility with a 1600 MeV proton beam for further study.

Whether or not the Pb-Bi target reaches an equilibrium condition which allows indefinite use must be resolved calculationally. At present all of the tools are available to do that, but the mechanics of their use is difficult. Specifically, the LAHET code package needs to be linked to the CINDER-2 burn-up code to provide a time-dependent material inventory for the spallation and Monte Carlo transport codes. The issue of compatiability of the materials produced in the Pb-Bi target is closely linked with the elemental abundance predicted by LAHET.

We need to verify that LAHET calculates accurate spallation yields for our target materials, especially for long-lived species. That has been studied before, but the interest was mainly in short-lived nuclei.

Many of the cross sections involved in our calculations are poorly known over some energy regions, or have never been measured. For the scoping calculations now underway, and at the accuracies needed now, we believe that the data base is reasonable. We have identified a few candidates which will need better nuclear data as we proceed with detailed design. A study that will determine the ATW nuclear data needs and decide how to resolve them will be important. Fortunately, experimental measurements for many of the critical, short-lived nuclei are now possible using the LANSCE and WNR facilities.

Research Needs

- Efficient neutron production and knowledge of spectrum is critical - benchmark experiments/code verification needed.
- Target radionuclide production needs better information - important issue for understanding waste stream.
- Code improvements - better physics treatments and links between code packages.
- Nuclear data will be needed for important isotopes in the near future.
- Radiation damage and materials compatability issues need better understanding.

SUMMARY

The possibility of producing extraordinarily high levels of neutron flux over a substantial volume has opened an entirely new suite of possibilities for dealing with high-level waste and energy production.

It is the use of a spallation source that makes it possible to produce the very high thermal neutron fluxes. By coupling that source to an efficient heavy-water moderator, a very flexible arrangement for isotope transmutation can be designed.

The recirculating Pb-Bi target avoids the issue of material damage in the target itself, can easily handle the proposed beam power, and offers the potential of having no high-level radioactive waste stream during facility operation.

Because material burn-up in the blanket is so rapid, it is possible to operate with very dilute loadings and flowing systems.

Although we have already successfully dealt with many problems, others still need resolution. Issues such as radiation damage are more difficult and require further study. At this time we have found no insurmountable obstacles.

Summary

- Spallation neutron source makes possible very high thermal flux over a large volume.
- Efficient transmutation wi[illegible]ilute loadings.
- Threshold fission materials become fuels not poisons.
- Recirculating Pb-Bi target - considerable past research

  no window for damage

  handles beam heat loading

  potential for no high-level radioactive waste discharge.
- No 'show stoppers'.

INTENSE THERMAL NEUTRON SOURCE

ATW Target/Blanket Design:
Application to Hanford Defense Waste

Michael Cappiello
Nuclear Technology and Engineering
Division

Contents

- Conceptual Design of the Target/Blanket Region
- Materials
- Neutronics
- Target Thermal-Hydraulics
- Heat Transport
- Research Needs
- Summary

Proposed Target/Blanket Layout

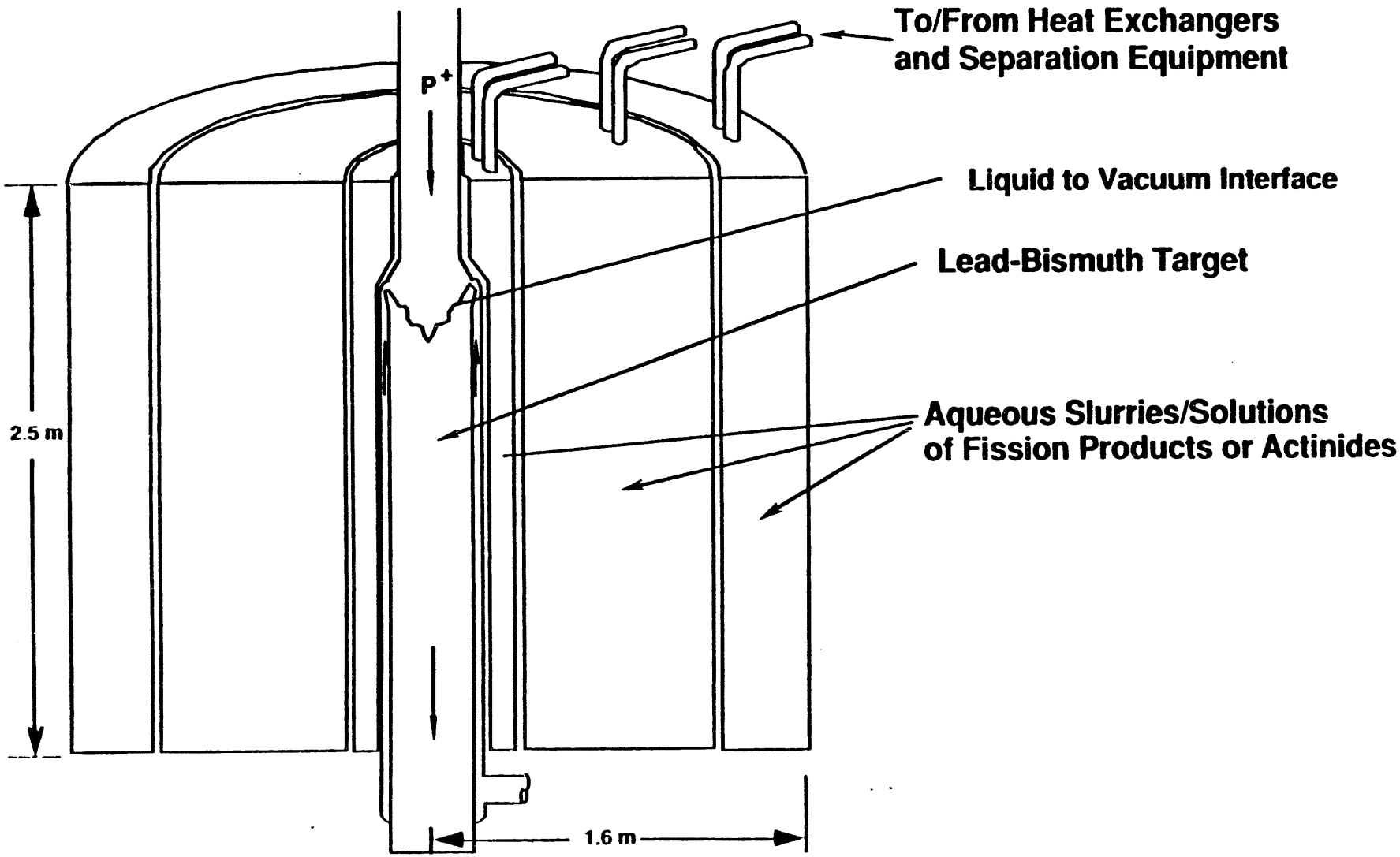
The target/blanket region is made of a central spallation target surrounded by a blanket region which acts as a neutron moderator and mass transport medium. The current spallation target material is a flowing liquid lead-bismuth eutectic, and the blanket is made of heavy water (D_2O) aqueous slurries or solutions. The heat transport loop for the lead-bismuth consists of a heat exchanger, pump, drain/charging tank, gas separator and pressurizer. Lead-bismuth is a toxic material. Adequate measures will be taken in the design of the system to ensure protection of the public/environment during normal and off-normal events.

In this design, it is proposed to transfer the heat in the target to a sodium-potassium eutectic (NaK) heat transport loop. The heat is then dumped to the atmosphere through a forced air heat exchanger similar to that used in the Fast Flux Test Facility (FFTF). The NaK heat transport fluid offers the advantage of a liquid metal coolant with a low melting point (19 C). It is particularly suitable in this case because of the low temperature heat that needs to be transported. The high heat transfer coefficients associated with liquid metal also help reduce the size of the heat exchanger surface area, and therefore minimize the volume of target material.

The target is surrounded by heavy water (D_2O) aqueous slurries or solutions of fission products and actinides. The D_2O moderates the neutrons, and acts as a mass transport medium for the fission products and actinides. Heavy water was chosen over light water due to the low neutron absorption and therefore high moderating ratio.

A modular design is proposed to allow change out of components with relative ease. It will most likely be necessary to replace the container wall of the lead-bismuth due to neutron damage on a yearly basis. Use of advanced alloys or materials may reduce the replacement frequency.

Proposed Layout



Proposed Target Material

A flowing liquid lead-bismuth eutectic alloy is currently proposed as the spallation target material. This material has been chosen as a spallation target material in projects in Europe and Canada. Because of the intense proton beam, a significant amount of heat is produced in the target. The flowing target allows the heat to be transported through convection and conduction of the target material itself. An alternative concept to a flowing liquid target would be solid target material with a fairly complex liquid metal cooling system. The flowing liquid target avoids these problems and offers a simple, effective method for providing a good spallation source, and a heat transport mechanism.

In order to provide a good spallation source of neutrons, the target material must have a high atomic number. Lead is a good candidate, but melts at a relatively high temperature (327 C). A lead-bismuth eutectic (55.5 % Bi) melts at about 125 C, and allows operation at a lower temperature. Lead-Bismuth is compatible with low-Cr, low-Ni stainless steel alloys. Also, the vapor pressure of the material is relatively low making the task of maintaining an interface with a hard vacuum much easier. It is noted that the target material is not yet fixed. A study needs to be performed to determine the advantages/disadvantages of other materials with respect to neutron generation, spallation products, engineering design and safety.

Lead-Bismuth has been proposed for the German Neutron Source (SNQ), and the SIN facility in Switzerland. A number of experiments with lead-bismuth have been performed recently at Kernforschungszentrum Karlsruhe (KfK) to investigate a vertical nozzle design for the SNQ. Also, in the late 1960s at Chalk River Nuclear Laboratories in Canada, a lead-bismuth eutectic was proposed for the Intense Neutron Generator project. A test loop was operated to determine the suitability of various alloys, and protective coatings.

Proposed Target Material

Proposed Target Material

- Lead-Bismuth Eutectic (55 % Bismuth)
- 125 C melting point
- High atomic number
- Good heat transfer properties
- Low vapor pressure (1e-5 mm Hg at 450 C)
- Compatible with low-Cr, low-Ni stainless steel alloys or steels with Mo coatings

Experience Base

- Scaled experiments for the German Neutron Source (SNQ). Vertical nozzle design studied.
- Flow tests performed at Chalk River
- Many corrosion tests cited in the literature
- Vendors available for pumps, valves, heat-exchangers etc.

Proposed Blanket Materials

For the blanket region, it is proposed to use heavy water (D_2O) aqueous slurries or solutions of fission products and actinides. The D_2O moderates the neutrons, and acts as a mass transport medium for the fission products and actinides. Heavy water was chosen over light water due to the low neutron absorption and therefore high moderating ratio. Dilute oxide slurries are proposed for the transport of technetium and the actinides. A dilute salt solution is proposed for iodine.

Solutions are used to a great extent in the chemical industry and are well understood. The dilute salt solution proposed here is compatible with the proposed container material (zirconium alloys) and will stay in solution at the temperatures proposed (less than 80 C).

The use of aqueous slurries offers many advantages in the transport of the fission products and actinides, especially in the dilute systems proposed here. Experiments with oxide slurries were carried out in the 1950's at ORNL for the development of homogeneous fluid reactors. Tests were performed with thorium dioxide slurries in concentrations up to 1500 gm/liter to study erosion/corrosion of materials, sedimentation, effects of particle size and shape and particle chemistry. In the slurries proposed here, the concentrations are much smaller (less than 100 gm/liter), therefore making erosion and corrosion less of a problem. The zirconium alloys were found to have good resistance to erosion/corrosion and are recommended. Zirconium also offers a distinct neutronics advantage due to its low neutron absorption. All slurries will settle to some extent. The larger the particle, the higher the settling rate. Solid particles less than 40 microns can be typically transported in laminar flow without settling. For the actinides, a particle size greater than 10 - 20 microns is suggested to keep most of the fission products from being ejected from the particle. In the design of the heat transport system for the slurries, care must be taken to avoid standing eddies or dead spots where particle accumulation may occur.

Proposed Blanket Material

Proposed Blanket Material

- D_2O Slurries or solutions
  - Tc, Np, Am: oxide slurries
  - Iodine: salt solution
- D_2O a good neutron moderator with low absorption
- Compatible with Zirconium alloys
- Low temperature and pressure

Slurry Experience

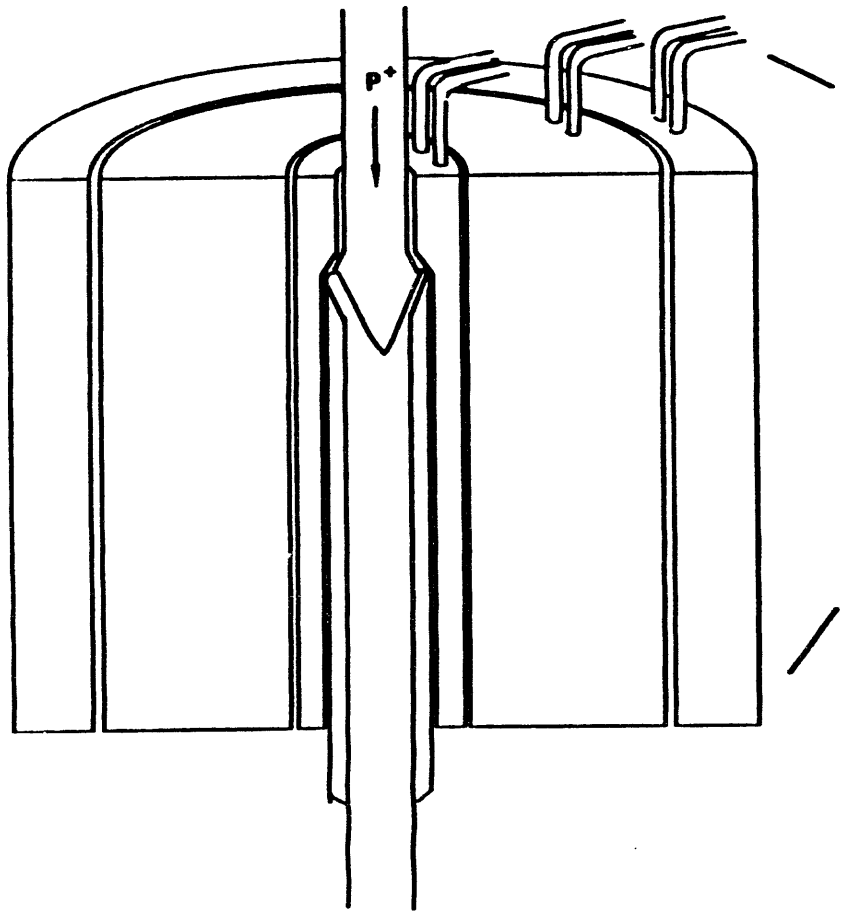
- Many tests performed at ORNL in the development of the Homogeneous Breeder Reactor
- Slurries of Thorium Oxide tested up to 1500 gm/l, and 300 C
- Techniques developed to control particle size and erosion problems

Neutronics Methods

Neutronics calculations are continuing in order to establish an optimum design for the transmutation of fission products and actinides. The overall goal is to make efficient use of the neutrons, and therefore reduce the size of the accelerator driver. Although not shown, the analysis performed here supports a more global system-wide analysis that takes into account the separation facility and accelerator.

Several calculational tools are used in the analysis. The LAHET code provides the neutron production from the high energy proton spallation and evaporation reactions. The MCNP code calculates in detail the neutron interactions in the target/blanket using the Monte-Carlo method. The ONEDANT and TWODANT codes are used for neutron transport calculations using discrete ordinates. The CINDER code is used to estimate equilibrium concentrations by performing burn-up calculations. And the TRANSX code is used to generate cross sections for the transport calculations.

Neutronics Methods



Tools

- LAHET
- ONEDANT
- TWODANT
- MCNP
- CINDER
- TRANSX

Sensitivity Studies

- Size of Blanket
- Geometric Location
- Material Densities
- Structural Materials
- Cross sections

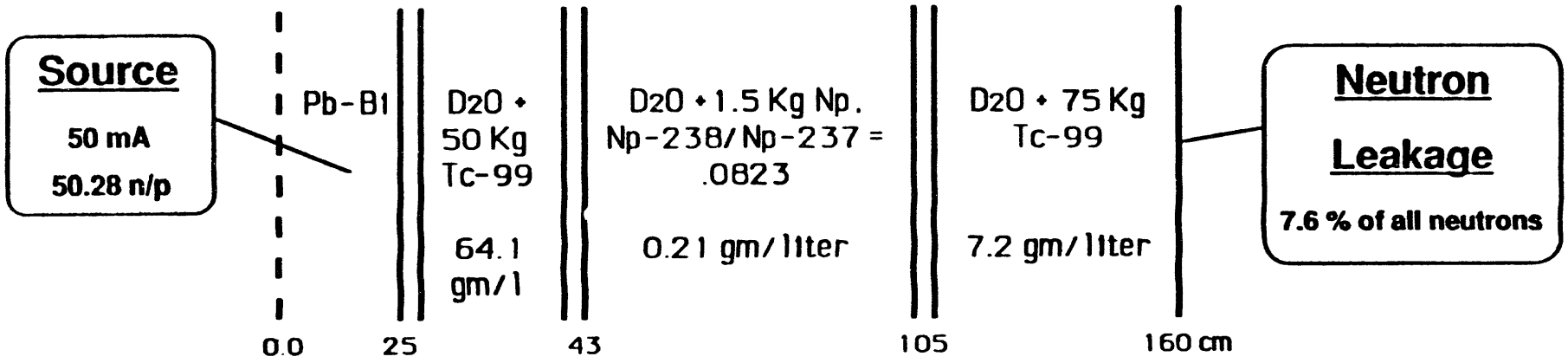
Optimize the Transmutation Rates for a Particular Application

Neutronics Results (1-D)

Several calculations have been performed to optimize the design using ONEDANT, TWODANT, and MCNP. Some results of sensitivity studies performed with ONEDANT are shown here. For these calculations, a total of 69 energy groups were used. The equilibrium concentration of Np-238 was found by iteration. For this particular geometry we have exceeded our goal transmutation rates for actinides. However, if one takes into account the axial leakage (about 9% based on previous 2-D calculations), then these transmutation rates are very close to our goal.

It is obvious from these first calculations that we can increase the efficiency of the design with a few simple changes. For instance, additional Tc-99 in the outer region will help reduce the leakage. Minimizing or changing the structural materials will reduce the parasitic capture. Also, increasing the amount of Np-237 will give us extra neutron multiplication through the capture and then fission of Np-238. Sensitivity studies will continue to optimize the design, and understand the influence of the various cross sections.

Neutronics Results (1-D)



Neutron Absorptions

Pb-Bi	5.10 %
Tc-99	61.4 %
Np-237	2.79 %
Np-238	3.12 % (89 % fission)
SS	12.0 %
Zr	11.9 %
D_2O	3.69 %

Neutron Production (per source neutron)

0.166 fission

0.028 n,2n

k-eff = 0.24

Transmutation Rates

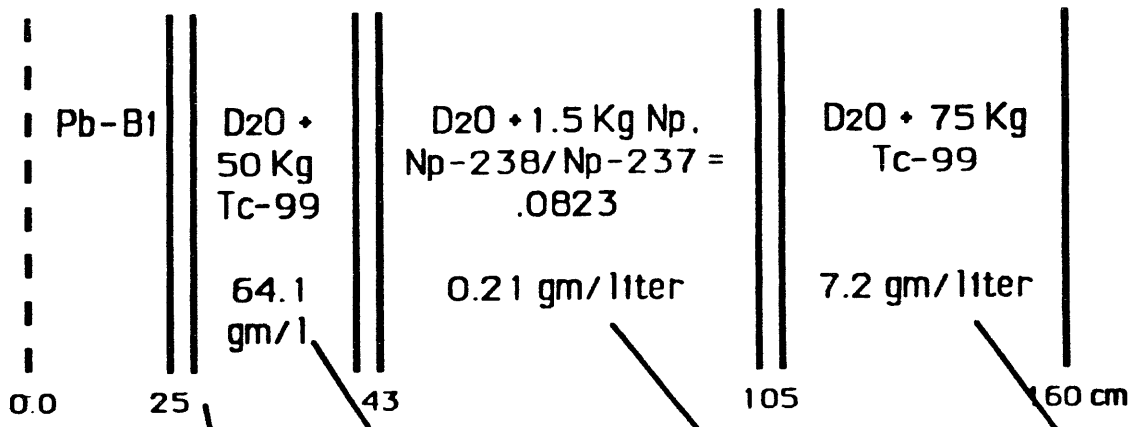
Tc-99 = 52.7 Kg/yr

Np = 12.1 Kg/yr

Calculated Neutron Flux

The calculated total neutron flux is shown at various locations radially outward from the source. The values shown are for the horizontal plane that represents the peak values.

Calculated Neutron Flux



Note:

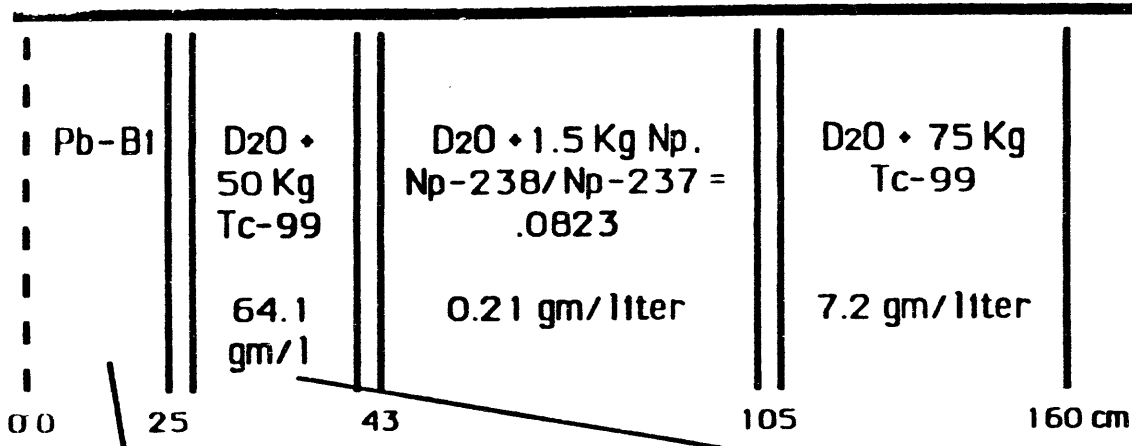
Flux levels assume a 50mA accelerator

Total Peak Flux (n/cm2-s)	8.27e15	5.74e15	2.73e15	0.49e15

Calculated Neutron Spectra

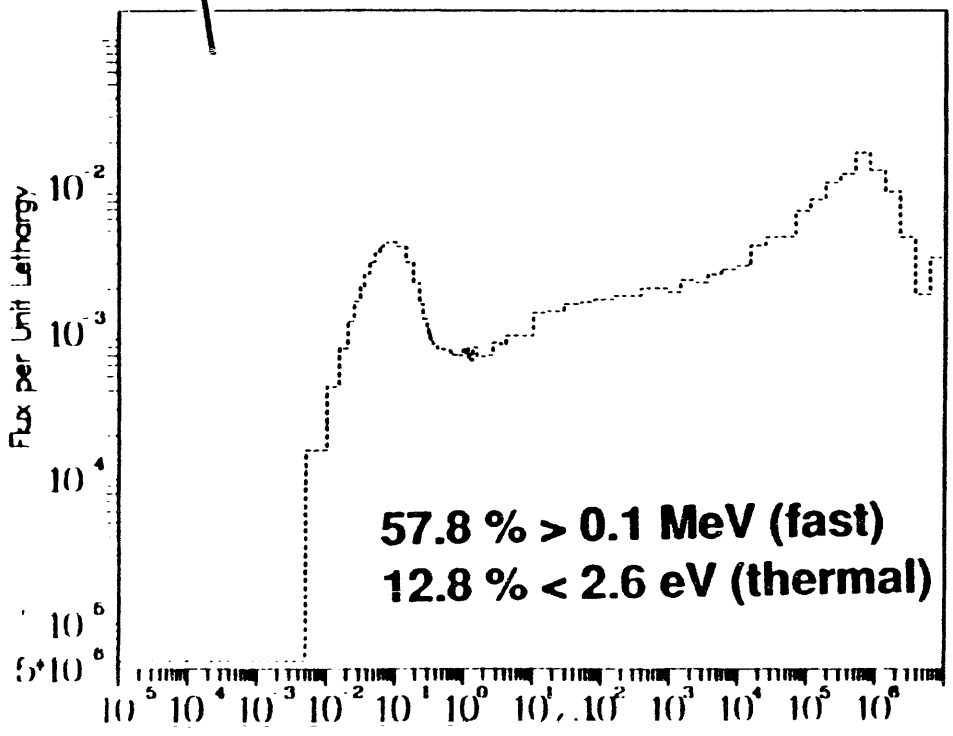
The neutron spectra for the central target region and the inner-most technetium slurry region are shown. The fluxes are on a per incident neutron basis, and are presented in terms of flux per unit lethargy. This quantity is the calculated group scalar flux times the average energy of the energy group, divided by the group energy width. The results show that in the target region, the spectrum is fairly hard (57.8 % above 0.1 MeV). But as one moves into the blanket region, significant moderation has occurred, and the flux is mostly thermal (45 % below 2.6 eV).

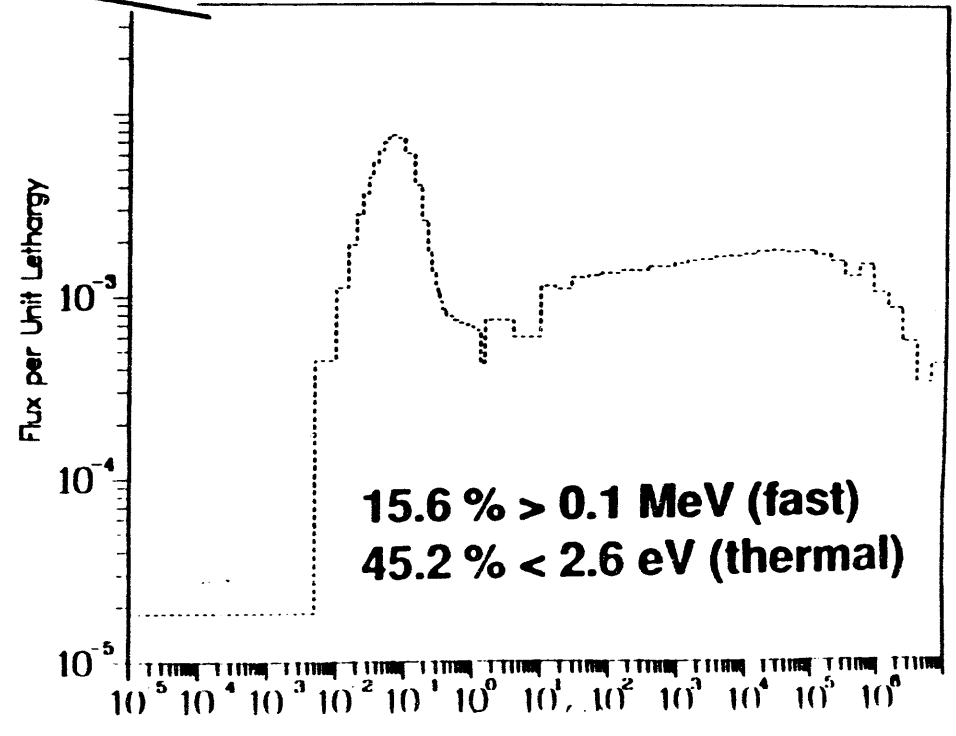
Calculated Neutron Spectra



Note:

Fluxes are on a per incident neutron basis. Flux per unit lethargy is $\Phi g^* \hat{E}/\Delta E$.





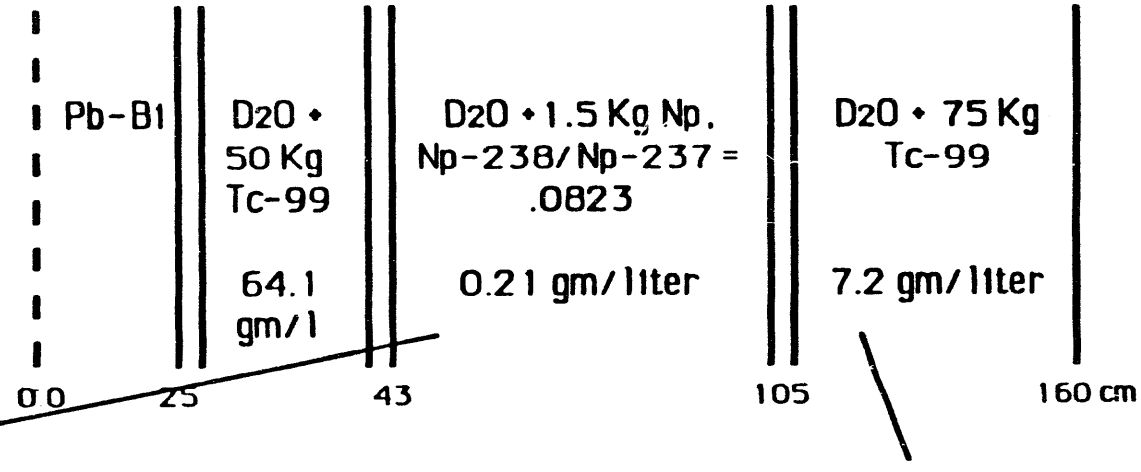
Calculated Neutron Spectra

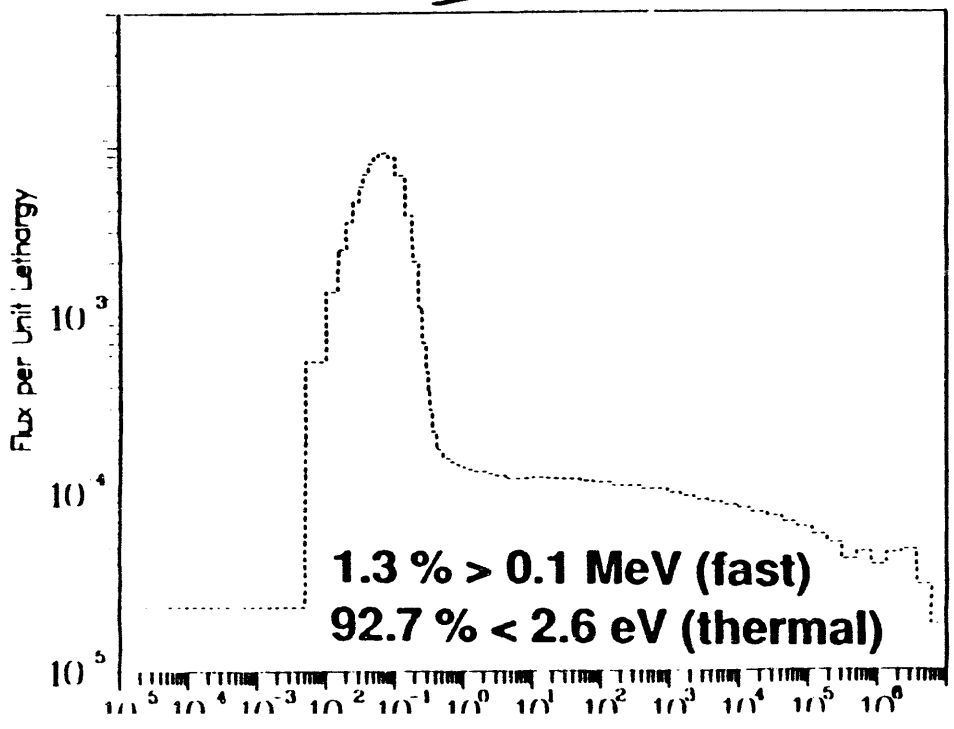
The neutron spectra for the actinide region and the outer-most technetium slurry region are shown. Here, the spectra is very thermal with the fraction of neutrons below 2.6 eV at 93 and 99 %, respectively.

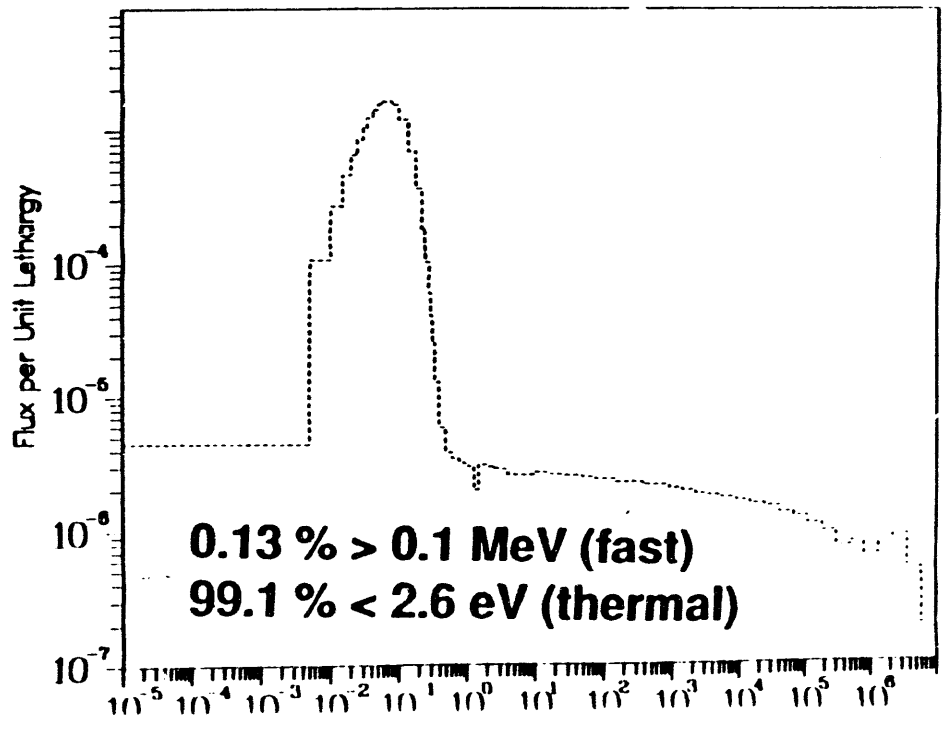
Calculated Neutron Spectra

Note:

Fluxes are on a per incident neutron basis. Flux per unit lethargy is $\Phi g^* \hat{E}/\Delta E$.







Lead-Bismuth Heat Transport

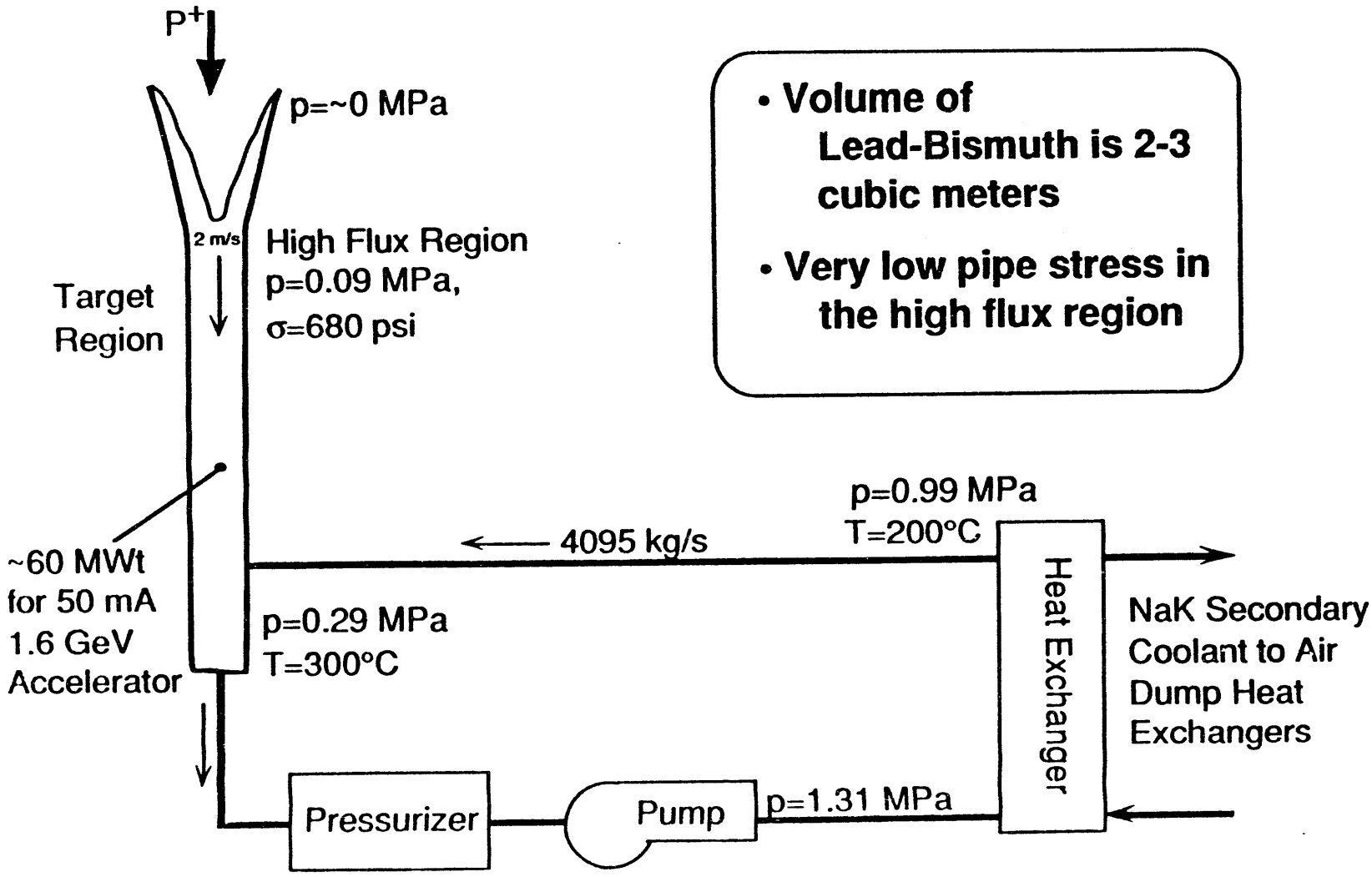
The proposed target is vertical with the liquid lead-bismuth flowing out of a ring-type nozzle into a cone shaped open channel. Here the fluid interacts with the proton beam and is in direct contact with a vacuum. The fluid coalesces in a pipe that is 50 cm in diameter, and flows downward at about 2 m/s. The flow exits the bottom of the target region, and is pumped through a heat exchanger, and then back to the target. One possible design is to pump the fluid up an annulus surrounding the center pipe, and then direct the flow back down through the center channel through the nozzle. Another possible method is to pump the fluid into an upper holding tank which then feeds the nozzle from the top using gravity. The latter technique may be beneficial from a safety standpoint because flow could be maintained for some time (depending on the volume of the holding tank) in the event of a pump shutdown. For either design, the volume of the fluid is about 2-3 cubic meters.

Approximately 75% of the power of the proton beam is deposited in the target. For a 50 mA, 1.6 GeV beam, this amounts to 60 Mw of heat. For a flow rate of about 4095 Kg/s, the temperature rise in the target is 100 C. The detailed power density and temperature distribution in the target are shown in a separate slide.

It is proposed to transfer the heat to a NaK coolant and then to the atmosphere through forced air heat exchangers. Dump heat exchangers of this type are used in the Fast Flux Test Facility (FFTF).

In the high flux region, the static head of fluid is very low (0.09 MPa or 13.2 psi). The calculated hoop stress in the pipe wall is only 680 psi assuming a wall thickness of 0.5 cm. This is very beneficial from a materials standpoint because a high strength alloy is not required.

Lead-Bismuth Heat Transport



- **Volume of Lead-Bismuth is 2-3 cubic meters**
- **Very low pipe stress in the high flux region**

Target Nozzle Fluid Dynamics

The fluid dynamics of the nozzle region are being investigated to determine a reasonable design and establish a basis for future flow tests. We have used a simplified model of the fluid governing equations (Navier-Stokes) by employing boundary layer approximations. The resulting equations are the conservation of mass and momentum in the axial direction. By introducing the appropriate scaling, the equations are dimensionless. A marching numerical solution method with adaptive error control is used to solve the resulting system of equations for local flow velocity and cross-sectional area until the point of coalescence is found.

The geometry is described in terms of non-dimensional variables. R is the cone radius, d is the film thickness, a is the angle of inclination for the cone, and H is the height at which the film coalesces into a single stream. The variable which is used to scale the system is the initial cone radius, R_o. The dimensionless film thickness is $D_o = d_o/R_o$ ($D = d/R$), and the dimensionless velocity is $U = u/u_o$. The effect of wall drag is represented through the skin friction coefficient, C_f which is a function of Reynolds number. The relative magnitude of the gravitational force is given by the Froude number,

$$Fr = \sqrt{\frac{U^2}{g R_o}},$$

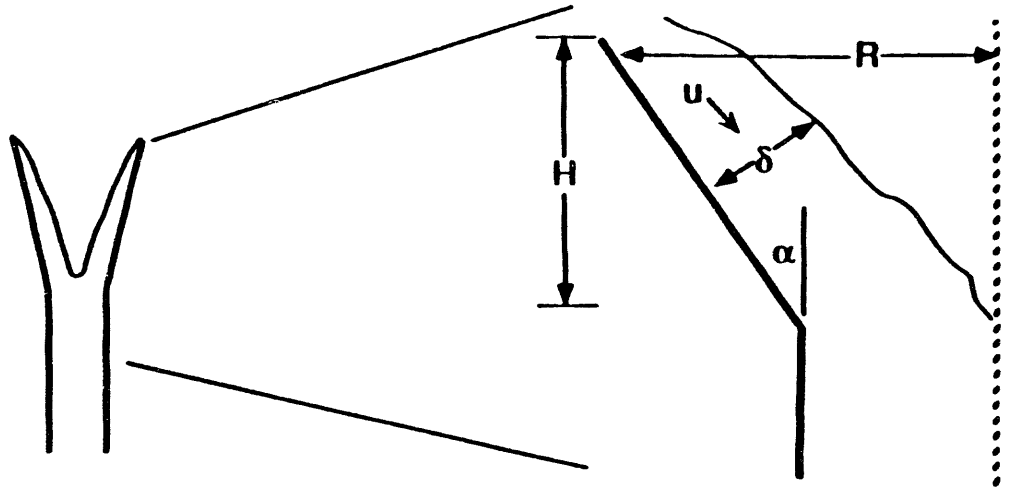
where g is the gravitational constant. For a coordinate system oriented with the sides of the cone, where h is the direction parallel to the side, the governing mass and momentum equations are

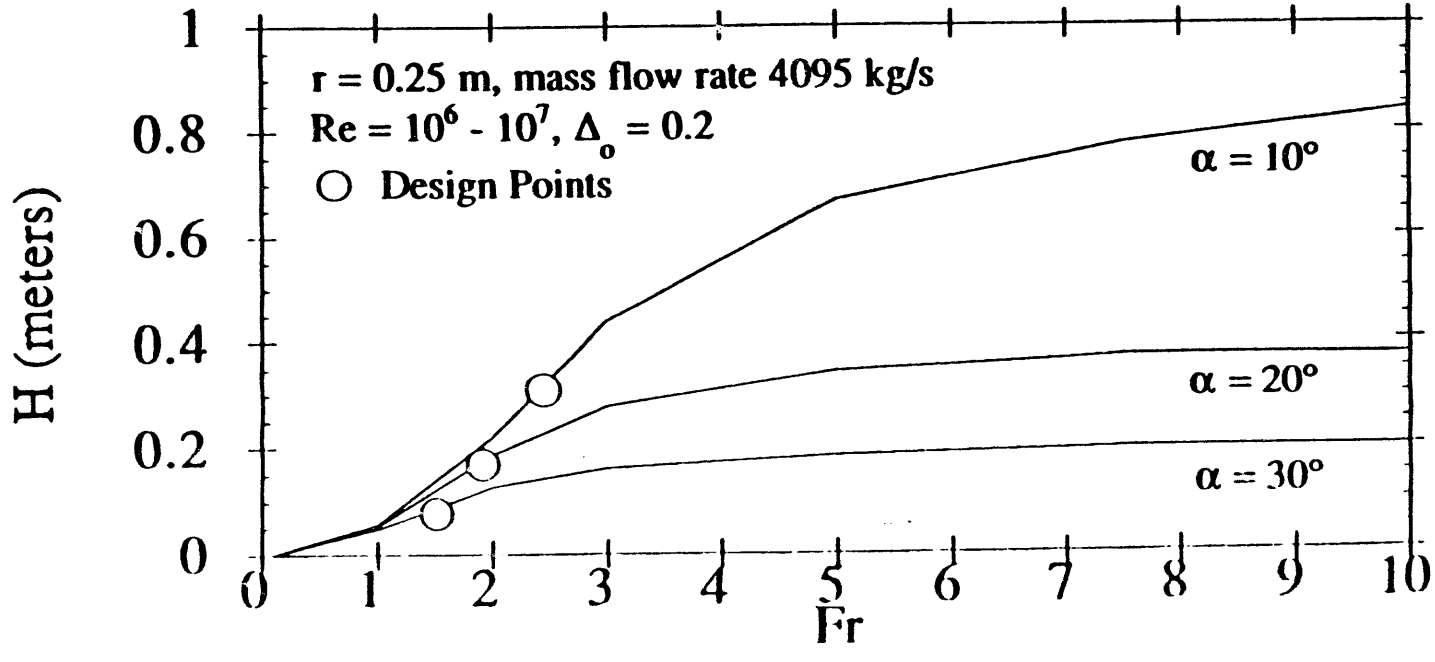
$$\frac{\partial U}{\partial \eta} = 0,$$

$$U\frac{\partial U}{\partial \eta} = \frac{C_f}{2\Delta}U^2 + \frac{\cos\alpha}{Fr^2},$$

The solution for the coalescence point is then found in terms of the flow conditions, height or radius of the coalescence point etc.

Nozzle Fluid Dynamics





Target Power Density and Temperature

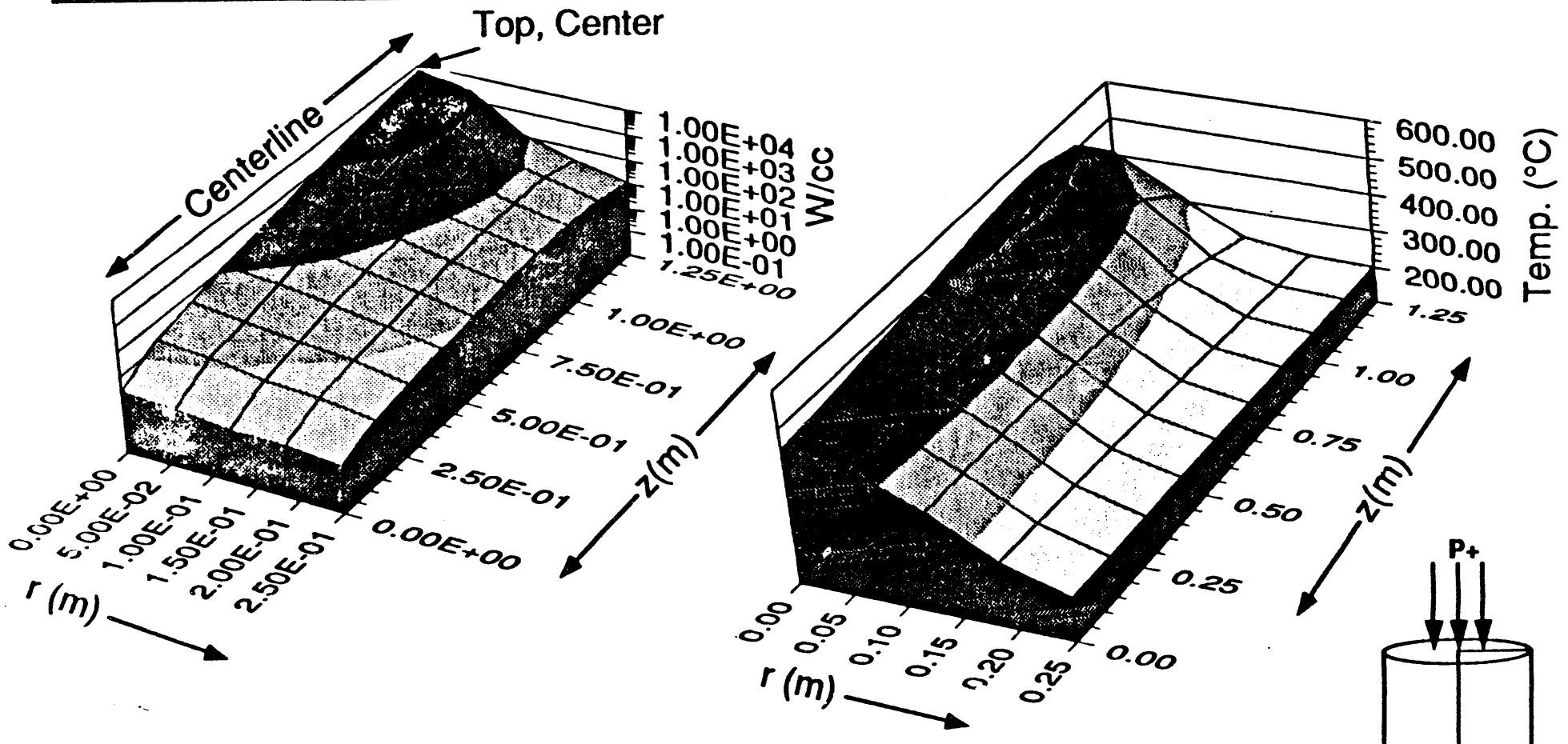
The power deposition is determined using the LAHET code for the protons and high energy (> 20 MeV) neutrons. The MCNP code is used for the lower energy neutrons (< 20 MeV). The summation of the two provides the total power deposition as a function of radial and axial distance. The target is assumed to be cylindrical, with a flat top surface. The noding used was 5 radial and 10 axial computational cells. The beam radius is assumed to be 25 cm which corresponds to the first two radial rings. The remaining 3 radial rings model the remaining lead-bismuth target out to a radius of 25 cm. Also, in the determination of the power density, the calculated values were scaled up 125 % to be conservative. Therefore, the numbers shown are for a 50 mA proton beam, multiplied by 1.25.

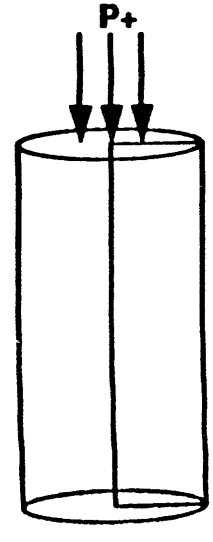
As expected the peak power density occurs in the center of the target. Also, most of the heat is deposited in the top 25 cm of the target. With the uncertainty factor of 1.25 applied, the peak power density in the top center calculational cell is about 6410 w/cc.

The temperature distribution was determined by solving the two-dimensional energy equation. The calculation includes the effect of axial convection and radial conduction. Also, the turbulent dissipation was estimated from Prandtl mixing length theory and used to calculate an effective radial conductivity. Because the Peclet number is sufficiently large, axial conduction was neglected. As stated above, in order to make a conservative estimate of the temperatures, the calculated power density was multiplied by 1.25.

As expected, the peak temperatures occur along the centerline, and due to convection, the peak occurs about 25 cm from the surface. The peak temperatures at the vacuum interface are sufficiently low to prevent boiling (409 C). Also, the vapor pressure at this temperature is sufficiently low (1.0e-05 mm Hg) so as not to contaminate the vacuum.

Target Power Density and Temperature





Maximum Surface Temperature 409°C
Maximum Power Density 6400 W/cc

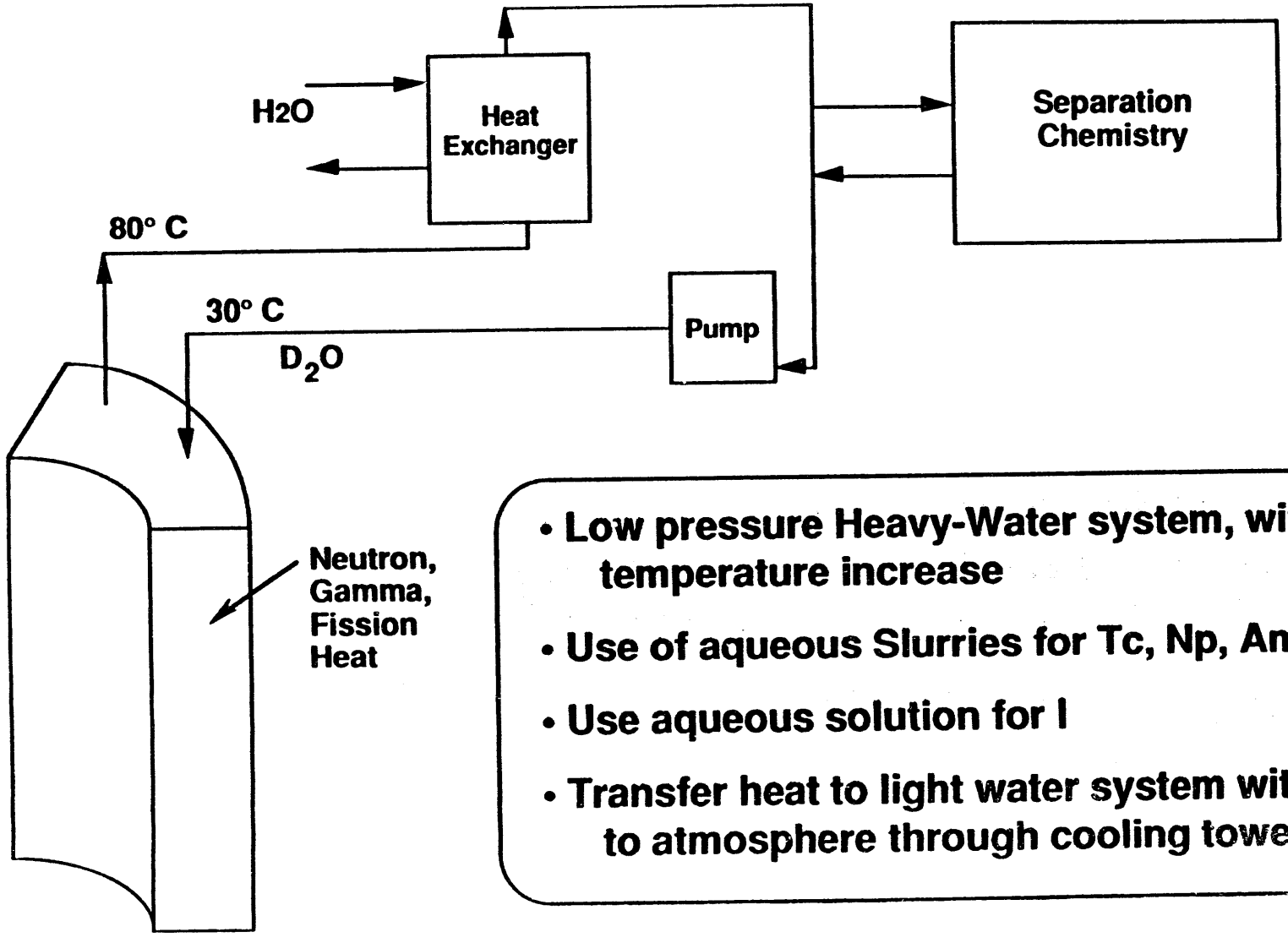
Fission Product/Actinide Heat Transport

The fission products and actinides are transported either as oxide slurries, or aqueous salt solutions, depending on the chemical separation process. For the fission product technetium, an oxide slurry is proposed since this offers a simple separation process for the transmutation product, ruthenium. For the actinides, oxide slurries are also proposed. And for the fission product iodine, an aqueous salt is proposed.

The heat deposition in the fission product aqueous slurry or solution is caused by neutron and gamma heating. Several MW of heat will be deposited in the solution, depending on the distance from the neutron source. It is proposed to transfer this heat to a light water system through a heat exchanger with eventual dumping to the atmosphere through a cooling tower. The heat exchanger can be placed immediately downstream of the blanket therefore cooling the slurry before entering the separation chemistry area. The aqueous slurry/solution is kept well below the boiling point at all times. The heat exchanger will be placed at an elevation higher than the blanket so that natural circulation will occur during a pump failure event. After accelerator shutdown, very little decay heat is produced thereby not requiring extensive decay heat removal systems as are required in nuclear reactor facilities.

In the actinide slurry, heat is deposited from neutron and gamma heating, as well as fission. The amount of heat deposition will depend on the actinide loading. As in the case of the fission products, heat is transported to a light water heat exchanger which in turn dumps the heat to the atmosphere through a cooling tower. Because the slurry is very dilute (less than 20 gm/liter), the amount of fissile material is small, and prevents the possibility of an accidental criticality. Also, for the same reason, the amount of decay heat is small, and can be transported easily through natural circulation in the event of a loss of pump event.

Fission Product/Actinide Heat Transport



- Low pressure Heavy-Water system, with 50 C temperature increase
- Use of aqueous Slurries for Tc, Np, Am
- Use aqueous solution for I
- Transfer heat to light water system with dump to atmosphere through cooling tower

Research Needs

Several areas of research and development need to be completed before a target/blanket system can be built. In the target area, experiments need to be performed to verify the nozzle design, determine material compatibility, and determine purification requirements. Also, we must investigate methods to minimize the volume and waste stream and determine the amount of decay heat. The choice of the target material is not fixed, perhaps a more suitable material is available. The material used to contain the target must also be resistant to the intense neutron flux. Alloys are available from the Liquid Metal Reactor research that exhibit very little damage, and may be suitable for use. These alloys need to be investigated.

In the areas of slurries and solutions, we need to verify our design with scaled experiments to ensure that no dead spots or eddies exist in the system that would accumulate material. Also we should verify the material compatibility and ensure that there are no erosion/corrosion problems. In this region, we need to reduce the parasitic absorption. The material we choose must have a small neutron absorption cross section, especially in the thermal energy region.

In the area of neutronics, research is needed to verify the nuclear cross sections because of their importance to the overall process. Also, if possible we need to verify the neutron production rate of the spallation neutron source.

Research Needs

Pb-Bi Target

- Fluids experiments to verify nozzle design
- Purification requirements
- Spallation products
- Decay heat

Neutronics

- Cross section validation
- Neutron generation

Slurries/Solutions

- Fluids experiments to eliminate eddies and "dead spots"
- Erosion/corrosion and material compatibility

Materials

- Compatibility with Pb-Bi, lifetime in intense neutron flux
- Low swelling rate alloys
- Protective coatings
- Reduce parasitic absorption
- Activation and decay heat

Summary

Self Explanatory

Summary

- Work has begun on the conceptual design of the target/blanket region of an accelerator-driven transmutation system with application to Hanford.
- The results of one-dimensional neutron transport calculations are encouraging. Goal transmutation rates of 50 Kg/yr Tc-99 and 10 Kg/yr Np are possible with a 50-60 mA current accelerator.
- It appears feasible to use aqueous slurries or solutions for the continuous flow of actinides and fission products in the blanket region.
- The heat transport of the spallation target will require about 2-3 cubic meters of lead-bismuth. The calculated fluid dynamics of the nozzle region and the interface with the vacuum show sufficient mixing and convection to prevent vaporization.
- In the near term, work will continue on the design of the target/blanket region to optimize the transmutation rates and therefore reduce the size of the accelerator. An investigation will be continued (in collaboration with the physicists and material scientists) in the areas of neutron generation and spallation products in the target in an effort to reduce the target volume and long-lived waste.
- If research funds become available, we will complete optimization and system studies, perform experiments to verify the target fluid dynamics, perform experiments if necessary to verify the slurry fluid dynamics and material compatibilities, identify safety issues and requirements, and begin work on a conceptual design and cost estimate for a particular application.

CHEMISTRY AND MATERIALS

Aqueous Chemical Processing for the Tc and Tc/Np Transmutation Blankets

Stephen Yarbro
Nuclear Materials Technology Division

What can this portion of the ATW program do for the nuclear complex?

Tc^{99} and I^{129} are more likely to reach the biosphere than the actinides. Many models have been developed for predicting how the radionuclides will behave in a repository over long time periods. The general conclusion is that the actinides will be sorbed by the soil. Therefore, over a long time period, e.g., a million years their hazard will be lessened because of radioactive decay and dispersion. However, some of the long-lived fission products are not sorbed and could potentially reach the environment over a few thousand year period. Hence, they could present a significant safety hazard.
Ref. M. Benedict, T.H. Pigford and H.W. Levi, *Nuclear Chemical Engineering*, McGraw-Hill Book Co., New York, NY, pp.620-624

Aqueous processes are well-known. This allows a reasonable extrapolation of their capability to process the material defined in the ATW program. Additionally, facilities and expertise are available within the complex to allow a solid development program to be successfully completed for a reasonable cost and time investment.

What can this portion of the ATW program do for the nuclear complex?

- Transmute Tc and Tc/Np to alleviate the long-term storage problem
- Use efficient aqueous chemical processing to process the blanket with a minimum of secondary waste produced
- Capitalize on the experience and facilities within the nuclear complex, such as Oak Ridge, Los Alamos and Hanford

What types of chemical processing does the program require?

Methods for removing tank sludges will need to be developed and tested. A potential option is to use a variation of the "jet pump" method that was developed at Hanford in the early '80's for removing sludge waste from the single-shelled tanks for volume reduction. The jet pump is mechanically simple and was successfully used to move thousands of gallons of high-level waste which was then pumped to evaporators. The concentrated sludge was then pumped back to single-shelled tanks for long-term storage. The system of transfer lines, expertise and equipment could potentially be used to remove the waste and transport to the chemical processing plant.

Waste processing will need to be very efficient for the program to be successful. However, this waste processing problem is similiar to problems encountered at plutonium processing sites such as the Los Alamos Plutonium Facility. An agressive program is underway to develop methods for scrubbing liquid wastes to remove all traces of radioactivity before discharge to the Los Alamos Low-Level Waste Handling Facility. The expertise developed at Los Alamos for actinides coupled with the expertise at Oak Ridge for removing fission products, such as, strontium and cesium, will provide a powerful combination for attacking this problem.

What types of chemical processing does the program require?

- Recover and prepare the stored defense wastes
- Process the Tc and the Tc/Np blanket
- Process low-level liquid wastes

Flowsheet for Hanford tank waste recovery

This represents the initial draft of a potential flowsheet for addressing the processing of civilian reactor fuel. However, exciting recent developments in separation science may revolutionize the processing scheme. New extractants based on molecular recognition technology will enhance actinide/lanthanide separations. Further improvement in the TRUEX-based processing will allow a complete recovery of the actinides/lanthanides from the initial tank waste. Also, improvements in the technetium processing flowsheet are being pursued. Overall, the assessment is that much of the technology for processing the tank waste is either demonstrated or within one to two years of demonstration.

Ref. J.A. Rawlins, "CURE: Clean Use of Reactor Energy", Westinghouse Hanford Co., WHC-EP-0268, May 1990

Hanford tank waste recovery flowsheet

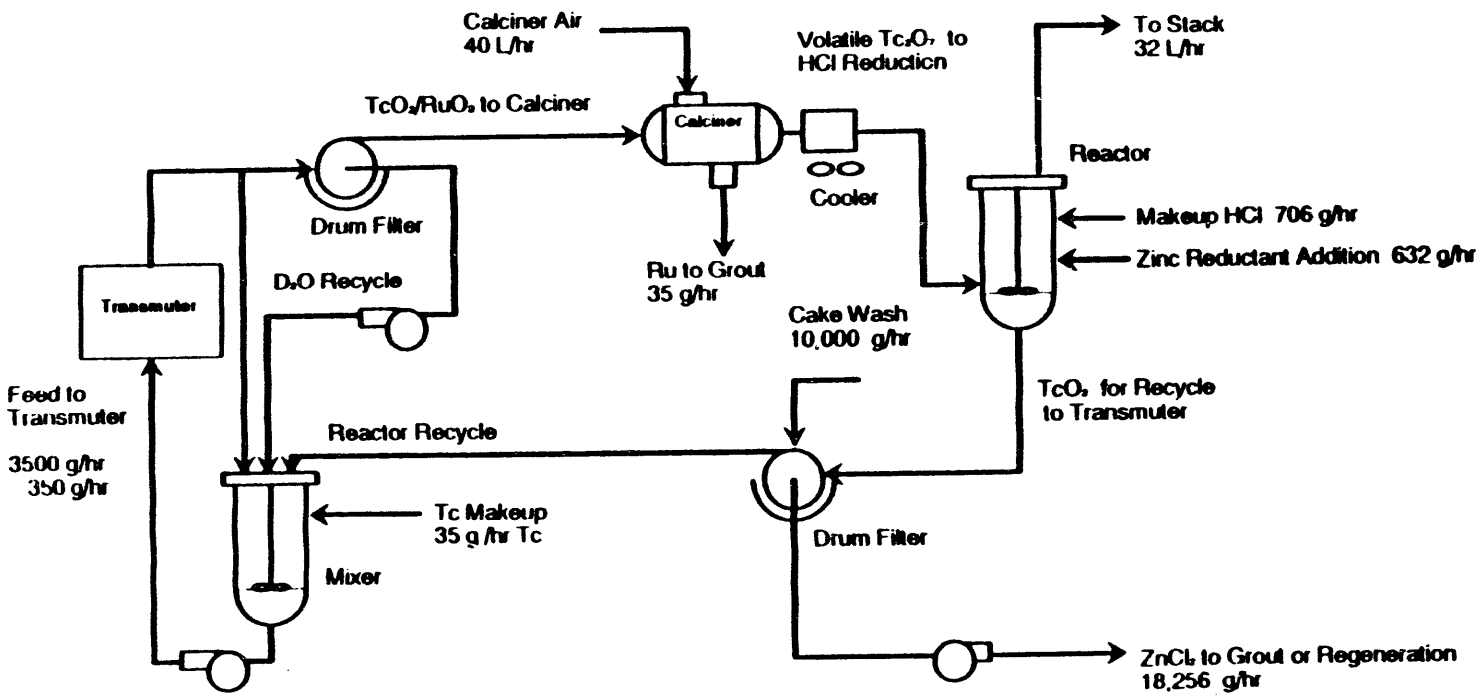
- Jet pump and separate sludge and supernate
- Dissolve sludge and recover U and Pu with PUREX, Sr recovery with hydroxyacetic acid
- PUREX raffinate to TRUEX for actinide/lanthanide recovery
- TRUEX raffinate to Tc recovery with amine solvent extraction
- Cs recovery with phophotungstic acid precipitation

Aqueous based Tc only blanket flowsheet

TcO_2 will be calcined in air at 400 deg C to form volatile Tc_2O_7 to separate Tc from Ru. An experiment is currently underway to measure the separation efficiency of this technique. Several grams of Tc and Ru oxides will be mixed and the proposed conditions used to measure the separation and the amount of cross-contamination. The most important factor is the separation between Tc and Ru in the Ru product oxide for discard. The amount of Tc will have to be extremely low to satisfy the 10 CFR 61 waste requirement.
Ref. W.T. Smith, L.E. Line and W.A. Bell, J. Am. Chem. Soc. **74** (1953) 4964

The gas stream with the Tc_2O_7 will be cooled and the Tc_2O_7 will be reduced to TcO_2 with HCl/zinc for re-entrainment in the heavy water. However, chloride could be a potentially corrosion problem with stainless-steel equipment. Therefore, other methods of reduction will be studied, such as hydroxylamine or Fe(II).
Ref. C.M. Nelson, G.E. Boyd and W.T. Smith, J. Am. Chem. Soc. **76** (1954) 348

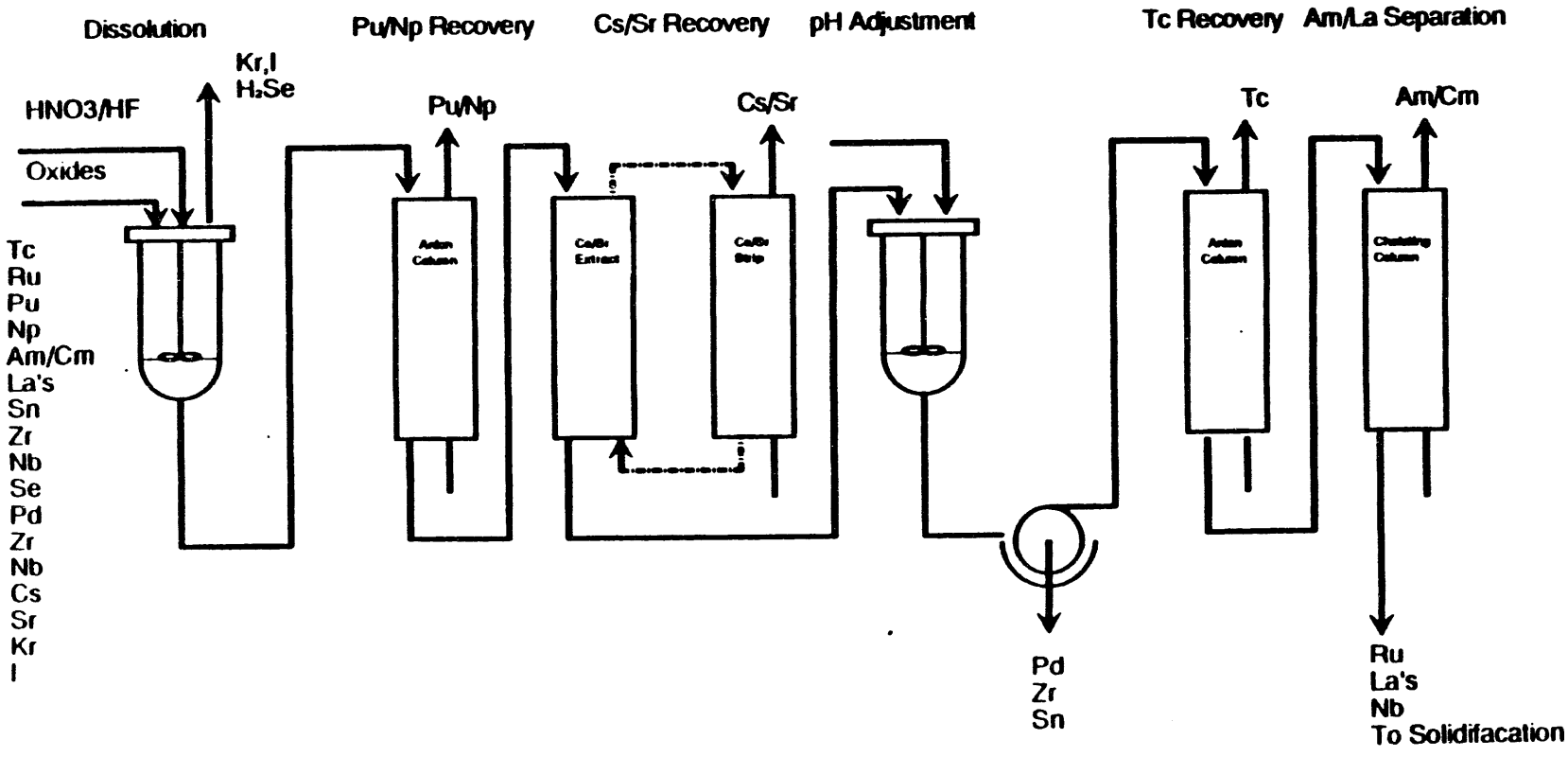
Aqueous based Tc only blanket flowsheet



Aqueous based Tc/Np blanket processing flowsheet

Ref. S.L Yarbro, S.L. Dunn and S.B. Schreiber, "Preparation of Pure Neptunium Oxide for Nondestructive Assay Standards," Los Alamos National Laboratory report, LA-11890, (July 1990)
D.B. James, "Anion Exchange Processing of Plutonium," Los Alamos National Laboratory report, LA-3499, January 1967
R.G. Shuler, et. al., "The Extraction of Cs and Sr from Acidic High Activity Nuclear Waste using a PUREX Compatible Organic Extractant," Solvent Extraction and Ion Exchange, 3(5), 567-604, (1985)
S.D. Reilly, C.F.V Mason and P.H. Smith, "Cobalt(III) Dicarbollide: A Potential Cs and Sr Waste Extraction Agent," Los Alamos National Laboratory report, LA-11695, (February 1990)
E.P. Roberts, et. al., "Recovery and Purification of Tc from Neutralized PUREX Wastes," Hanford Works report, HW-SA-2581, 1962
J.N. Mathur and P.K. Khopkar, "Ion Exchange Behaviour of Chelating Resin Dowex A-1 with Actinides and Lanthanides," Solvent Extraction and Ion Exchange, 3(5), 753-762 (1985)
B.F. Smith, G.D. Jarvinen, M. M. Jones and P.J. Hay, "The Synthesis and Actinide and Lanthanide Complexation of "Soft" Donor Ligands: Comparison Between HBMPPT and HTBMPP with TOPO Synergist for Am(III) and Eu(III) Extraction," Solvent Extraction and Ion Exchange, 7(5), 749-765 (1989)

Aqueous based Tc/Np blanket flowsheet



Liquid waste treatment flowsheet

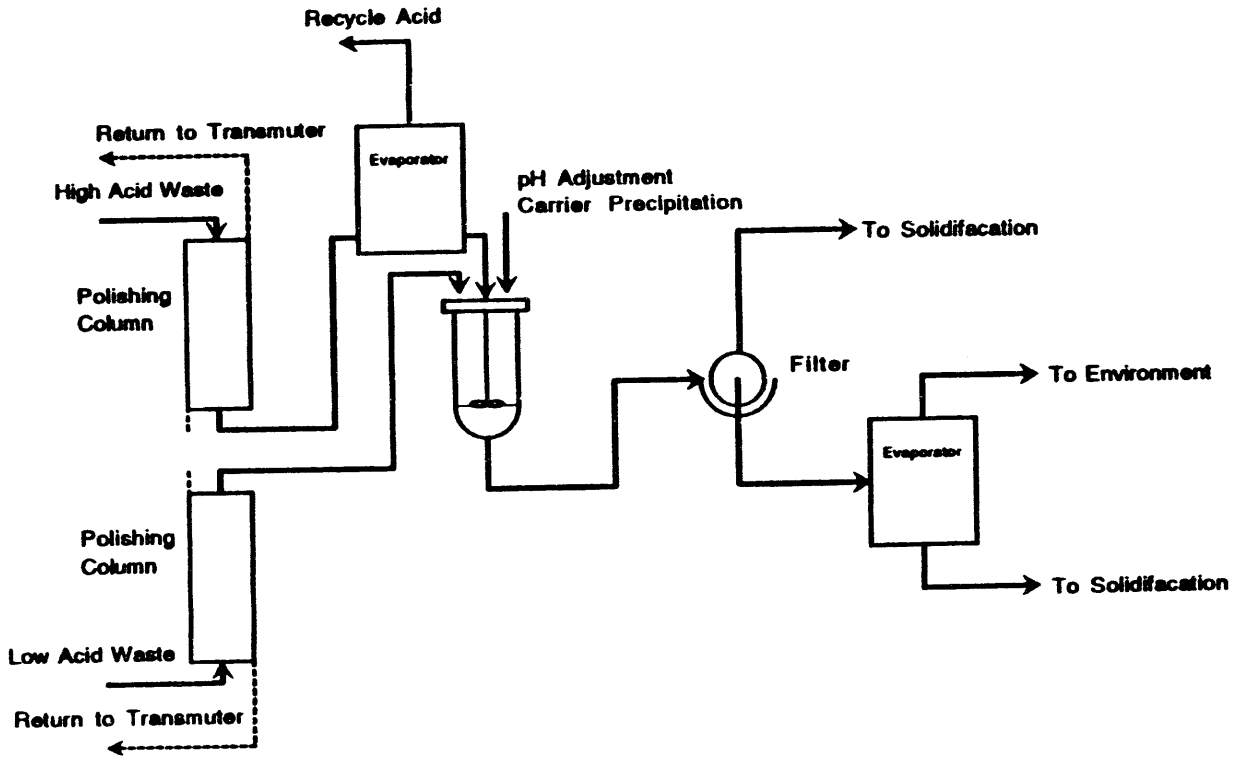
Use chelating resin columns, currently under development, to scrub actinides and lanthanides from the high-acid waste streams.
Ref. S.L Yarbro, S.L. Dunn and S.B. Schreiber, "Processing Low-Level Radioactive Solutions with Extraction Chromatography," LA-UR-89-2741, Proceedings from the 1989 National AIChE Meeting, San Francisco, CA
S.L. Yarbro, "Using CMPO-Based Levextrel Resins to Process Low-Activity Nitric Waste Streams," LA-UR-88-792, Proceedings from the 1988 Annual Actinide Separations Conference, Chicago, IL
S.B. Schreiber, S.L. Yarbro and S.L. Dunn, "Evaluation of Extractants and Chelating Resins in Polishing Actinide Contaminated Waste Streams," Los Alamos National Laboratory Report LA-11978 (October 1990)

Adjust feed acidity and use inorganic resins to scrub low acid Cs and Sr.
Ref. S.M. Robinson, W.D. Arnold and C.H. Byers, "Design of Fixed-Bed Ion Exchange Columns for Wastewater Treatment," Proceedings of the Waste Management '90 Symposium, Tucson, AZ

Use homogeneous pH control for ferric hydroxide or ferrate carrier to perform final scrub before discharge to the environment.
Ref. J. Bucholtz, Los Alamos National Laboratory, personal communication, present operating procedure at Los Alamos Low-Level Waste Handling Facility.
S.L. Yarbro, S.B. Schreiber and S.L. Dunn, Los Alamos National Laboratory, unpublished data, 1990

Use evaporation or membrane pervaporation to reduce anion concentration in solutions for discharge and recycle acid for reuse.
Ref. A.G. Nicol, B.G. Gomez, C.W. Mills, S.B. Schreiber and S.L. Yarbro, "Deployment and Operation of the Enhanced Evaporator System at TA-55," LA-UR-90-1182, 1989 American Glovebox Society National Meeting, Oakland, CA

Liquid waste treatment flowsheet



What are the engineering criteria for flowsheet design?

200 kg/yr throughput requires equipment similar in scale to the Los Alamos Plutonium Facility (TA-55). This ensures a pool of experienced engineers and scientists for design and development of the chemical processing flowsheet and facilities.

Fission products with <30 yr half-life only need 300 yr storage which is well within present engineering technology. The advantage of storing the shorter-lived fission products is that it eliminates the need for isotope separation of the cesium and reduces the requirement for remote-handling facilities. Also, there is a slight potential for an economic benefit from the heat produced from these two elements.

Class A or non-TRU (<100 nCi/g) wastes are worthy goals, but further study needs to be done to ensure that they are realistic. The major issue will be the economics associated with the processing and the amount of secondary waste produced to achieve the Class A or non-TRU waste. More development work is needed to define realistic costs and waste generation data.

What are the engineering criteria for flowsheet design?

- Throughput
  - 200 kg/yr Tc for the Tc only
  - 200 kg/yr Tc and 10 kg/yr Np for the combination
- Short-lived fission products (<30 yr half-life) to managed storage
- Long-lived fission products to transmuter
- Produce Class A or non-TRU waste (<100 nCi/g)

What separations criteria are necessary for a successful program?

The definitions of the different waste classes are specified in 10 CFR 61. For class A waste, the radionuclide content is very low. This means that decontamination factors of the order of 10^5 to 10^6 will be required for the project to be successful. Especially, the waste processing step will need to be efficient to ensure that a greater volume of waste will be produced and stored. The separations required for material entering the transmuter will be determined by the neutronics of the target and blanket design.

Ref. J.A. Rawlins, "CURE: Clean Use of Reactor Energy", Westinghouse Hanford Co., WHC-EP-0268, May 1990

What separations criteria are necessary for a successful program?

Waste Class Specifications

	Class C	B	A
Total TRU, nCi/g	100	10	10
Sr90, Ci/cu. m	7000	150	0.04
Tc99, Ci/cu. m	3	0.3	0.3
I129, Ci/cu. m	0.8	0.08	0.08
Cs137, Ci/cu. m	4600	44	1

Ref: Rawlins, J.A. ed., "CURE: Clean Use of Reactor Energy", WHC-EP-0268, Westinghouse Hanford Company May 1990 pg. 3-3

Why choose aqueous slurries for this application?

For this application, the slurries will be two orders of magnitude less concentrated than in the Oak Ridge experiments. Therefore, many of the problems they experienced with the slurries will be lessened or eliminated. Furthermore, the two phase solutions allow more concentrated storage as an oxide than as a solution. Also, the chemistry of solutions undergoing intense neutron fluxes is not well understood. With the slurry, the solid phase can be separated and the material processed with well-characterized aqueous techniques.

Ref. J.F. Flagg, *Chemical Processing of Reactor Fuels*, Academic Press, NY, pp. 328-346 (1961)

Why choose aqueous slurries for this application?

- Operating experience with thorium oxide slurries used as transmutation blankets at Oak Ridge
- Enables flexible processing by allowing storage as oxide instead of solution for cooling before processing
- Oak Ridge successfully used slurries up to 1500 g/L Th; this application will only require slurries at 10-50 g/L

Why choose ion exchange technology over conventional PUREX/TRUEX technology?

Because of many years of operating experience at Los Alamos and Rocky Flats Plant with actinides and a variety of transition metal and lanthanide contaminants. Also, ion exchange is simple to operate remotely. Ion exhange has been successfully operated in the fuel reprocessing cycle and has proven to be robust and give excellent separation of actinides from fission products. It is also the accepted method for separation of americium from lanthanides.

Ref. D.B. James, "Anion Exchange Processing of Plutonium," Los Alamos National Laboratory report, LA-3499, January 1967
S.F. Marsh and A.E. Nixon, "Review of Current Nitrate Anion Exchange Recovery and Purification Processes for Plutonium at DOE Production Facilities," Los Alamos National Laboratory report, LA-11593, August 1989
S.F. Marsh, "Improved Recovery and Purification of Plutonium at Los Alamos using Macroporous Ion Exchange Resin," Los Alamos National Laboratory report, LA-10906, May 1987

Why choose ion exchange technology over conventional PUREX/TRUEX technology?

- IX Columns are simple mechanically and cheap to operate remotely
- IX columns can provide very high concentration factors especially for dilute solutions
- IX columns can be operated to provide many theoretical plates for difficult separations
- Resins do not need continual washing as do solvents and can be incinerated for disposal

The technology for high-yield actinide and fission product recovery is available and there is *operating experience with most of the processing steps*

Many years of operating experience at Los Alamos and Rocky Flats Plant with actinides and a variety of transition metal and lanthanide contaminants.

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ATW waste problem is similar to waste problem at the Los Alamos Plutonium Facility. Los Alamos has been working on polishing wastes for several years to reduce the actinide concentration, volume and amount of solid waste generated through treatment.
Ref. S.B. Schreiber, S.L. Yarbro and S.L. Dunn, "Evaluation of Extractants and Chelating Resins in Polishing Actinide Contaminated Waste Streams," Los Alamos National Laboratory report, LA-11978 (October 1990)

Near full-scale tests (78.5" high x 76" dia. column) have been completed at Oak Ridge for removing Cs and Sr from waste water before discharge to the environment.
Ref. S.M. Robinson, W.D. Arnold and C.H. Byers, "Design of Fixed-Bed Ion Exchange Columns for Wastewater Treatment," Proceedings of the Waste Management '90 Symposium, Tucson, AZ

Technetium removal from neutralized PUREX wastes has been demonstrated at Hanford.
Ref. F.P. Roberts, et. al., "Recovery and Purification of Tc from Neutralized PUREX Wastes," Hanford Works report, HW-SA-2581, 1962

New possibilities exist for reagents for Am/La separations.
Ref. B.F. Smith, G.D. Jarvinen, M. M. Jones and P.J. Hay, "The Synthesis and Actinide and Lanthanide Complexation of "Soft" Donor Ligands: Comparison Between HBMPPT and HTBMPP with TOPO Synergist for Am(III) and Eu(III) Extraction," Solvent Extraction and Ion Exchange, 7(5), 749-765 (1989)

The technology for high-yield actinide and fission product recovery is available and there is operating experience with most of the processing steps

- Los Alamos has demonstrated 99.9% actinide recovery routinely on ion exchange columns
- Los Alamos has demonstrated 99.9995% actinide recovery on ion exchange waste streams (>10E6 total actinide recovery)
- Oak Ridge has demonstrated polishing of Cs and Sr waste streams down to discharge levels for their new Process Waste Treatment Plant (PWTP)
- Hanford has demonstrated pilot-scale Tc recovery columns with >80% Tc recovery over 25 years ago
- Los Alamos has demonstrated Am/La separation factors of >100 with a new class of "soft" donor ligand extractants

What issues remain to be resolved?

Am/La separations need to be examined and demonstrated on a large-scale. An experiment is underway to measure the separation efficiency of a new class of "soft-donor" ligand extractants for Am from the lanthanides expected to be present in the ATW processing streams.

Solidification techniques need to be examined and ultimately certified for storage. This problem can only be addressed after sufficient work has been done to adequately define the types of waste that will be produced.

Faster and more sensitive on-line analytical techniques need to be developed and certified. This techniques, along with more efficient waste polishing techniques are being agressively developed as a part of the Waste Minimization and Complex Modernization efforts at the Los Alamos Plutonium Facility.

The facility design should be carefully examined. A good model is the new hot-cell facility at Harwell in the U.K. Ref. P.E. Brown, D. Campbell and J.R. Findlay, "A Modern Alpha, Beta and Gamma Active Handling Facility for Chemical and Allied Research and Development Work," UKAEA Harwell Laboratory report, AERE R 13457 (September 1988)

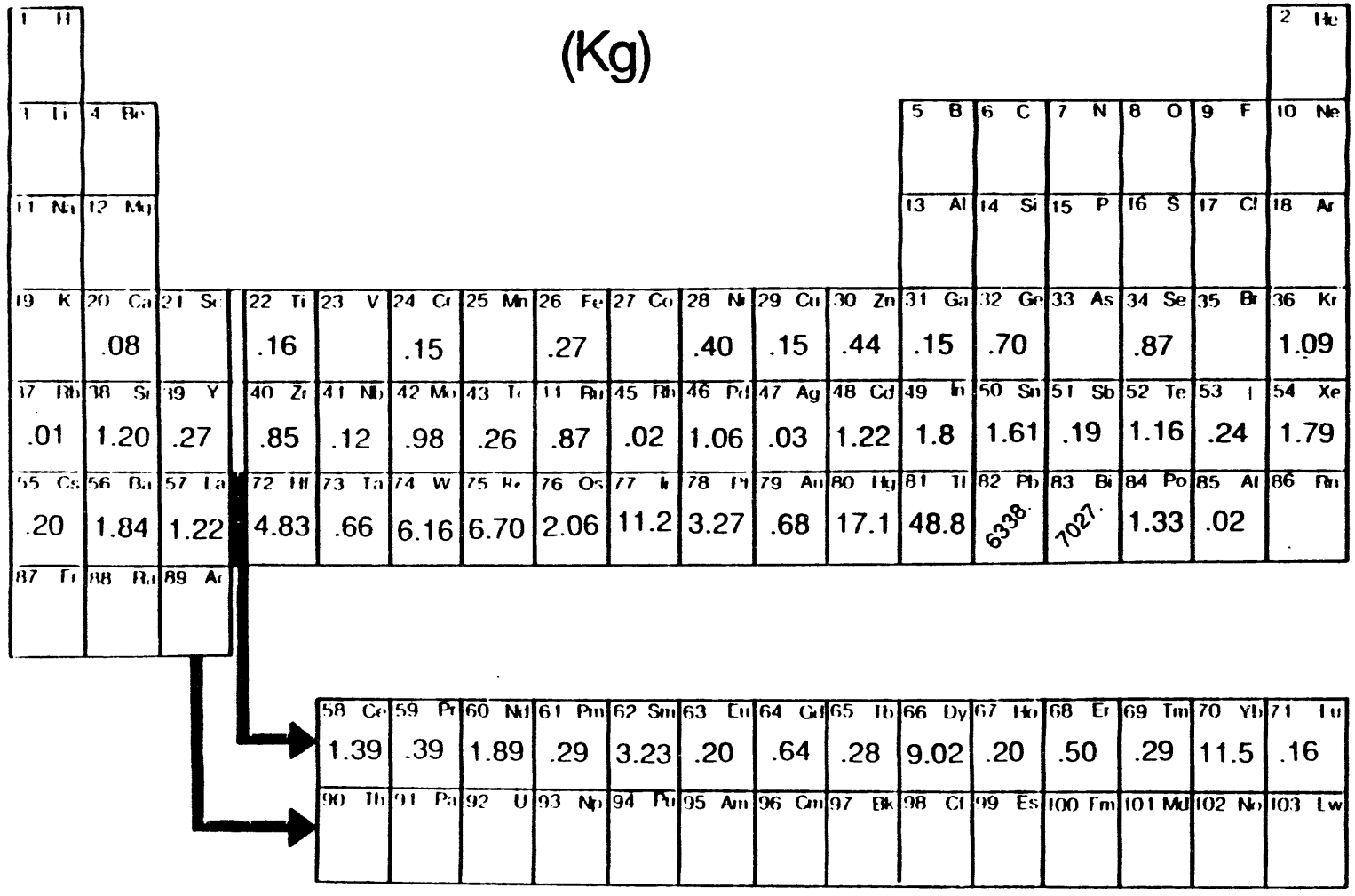
What issues remain to be resolved?

- All of the chemical processing, especially the Am/La separations, needs to be demonstrated as an integrated flowsheet
- Careful facility and flowsheet design to ensure that secondary waste generation is minimized and that the facility can be decommissioned properly
- Careful attention to safety issues, such as proper facility design to ensure low radiation exposures
- Ensure that waste forms (such as glass) can be produced which are compatible with current and future storage requirements
- Reliable cost and schedule data need to be developed

AMOUNT OF TRANSMUTED ELEMENTS FROM Pb/Bi EUTECTIC AFTER 30 YEARS

(1.6 GeV-55ma)

(Kg)



Solubilities (wt. %) At ~ 200°C

At approximately 200°C the known solubilities of the various elements in lead and bismuth are shown. Most elements (except for In, Hg, Tl, Sn, Sb) have relatively low or essentially no solubilities. This is not to imply that some solubilities may exist but that many solubilities have not been evaluated. Here is an area that needs particular focus due to the potential problems that may arise should these elements or their compounds precipitate or plate out in particularly the cold part of a recirculating system.

CHEMISTRY AND MATERIALS

ATW Materials Issues

Karl Staudhammer
Materials Science and Technology
Division

Scope

The scope of this presentation covers primarily two areas of materials concern. The first concern involves radiation effects which we know will play a dominant role in materials selection because the neutron fluences are relatively high. Secondly, we should address materials compatibility issues particularly in light of a desire for long term operation. Compatibility data from previous investigators over the last 20 years indicate that at operating temperatures less than approximately 500^{o}C for a variety of low alloy steels (particularly those containing < 2.5 wt.% Cr) have not shown any corrosion up to 24,000 hrs. While significant data exists for radiation effects on materials, as well as for compatibility/corrosion, the synergistic effects must be understood. Sufficient data exist in other material systems that indicate such a concern is warranted here.

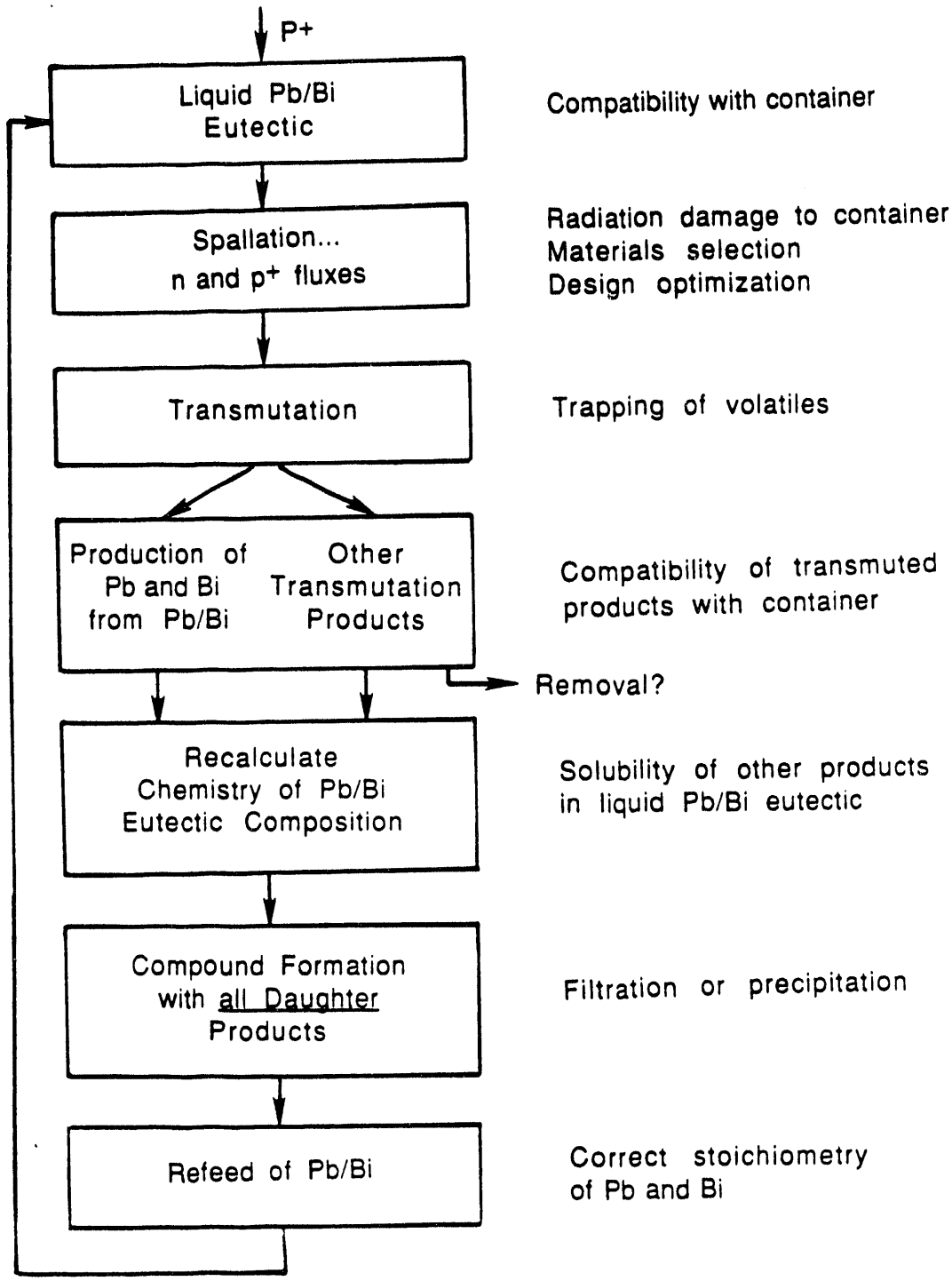
SCOPE

- RADIATION EFFECTS
- MATERIALS COMPATIBILITY
- SYNERGISTIC EFFECTS

Flow chart for the spallation Source

The flow chart presents a schematic of the pertinent issues associated with each conceptual operation/event in the spallation source. It is presented as an overview. The specific issues are presented in greater detail in the following viewgraphs.

FLOW CHART FOR THE SPALLATION SOURCE



Radiation Effects

Radiation effects concerns along with possible remedies. Both problem and remedies are microstructure dependent. Areas of concern include:

- Embrittlement the Ductile to Brittle Transformation Temperature (DBTT) is of most concern in Body Centered Cubic (BCC) metals. Some ceramics are actually toughened by radiation.

- Swelling is typically temperature dependent. Generally there are two major aspects of the swelling problem. Geometric, in which macro dimensional changes are observed, and differential swelling, which is a microscopic process that leads to mechanical failure, grain boundary separation and different swelling in multiphase systems.

- Neutron induced transmutation leading to gas formation within the metal can accelerate materials degradation.

- Welds have poorly controlled microstructure that makes them susceptible to enhanced degradation.

- Mechanical strength may improve under radiation but can be overshadowed by embrittlement problems.

Radiation enhanced creep is not a concern because operating temperature and stresses are sufficiently low. For most materials the regime would be in the diffusion controlled flow. For example, for 316 stainless steel operated at 0.3 T_m the creep rate would be $<10^{-12}$ in. / in. / sec. Materials having creep rates within an order of magnitude of 10^{-12} at low stresses will easily last the 30 year life span of an ATW system.

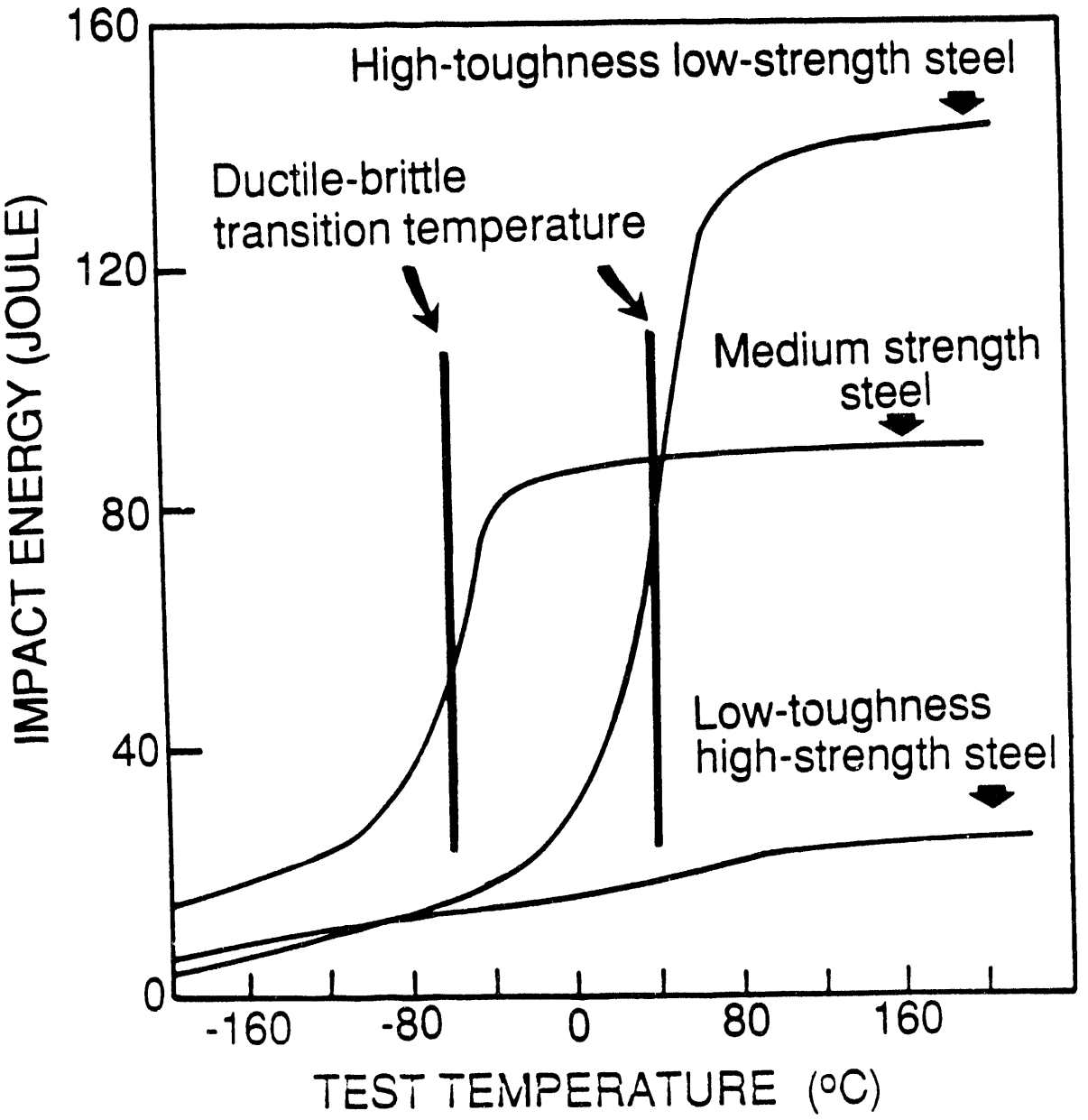
RADIATION EFFECTS

Problems	Solutions
• Embrittlement (DBTT)	Alloying
• Swelling f(T)	Design, material selection
* Geometric	Microstructural/engineering
* Differential	Graded interfaces
• Transmutation gases	Materials selection and microstructural/engineering
• Welds	Design
• Mechanical strength (often increases)	Materials selection and microstructural/engineering

Example of DBTT

Impact test data for various temperatures can reveal significant changes in toughness and fracture behavior. (After A. S. Tetelman and A. J. McEvily.) The ductile to brittle transition temperature is also a function of composition as well as texture (i.e., rolling or worked direction of the microstructure).

EXAMPLE OF DBTT

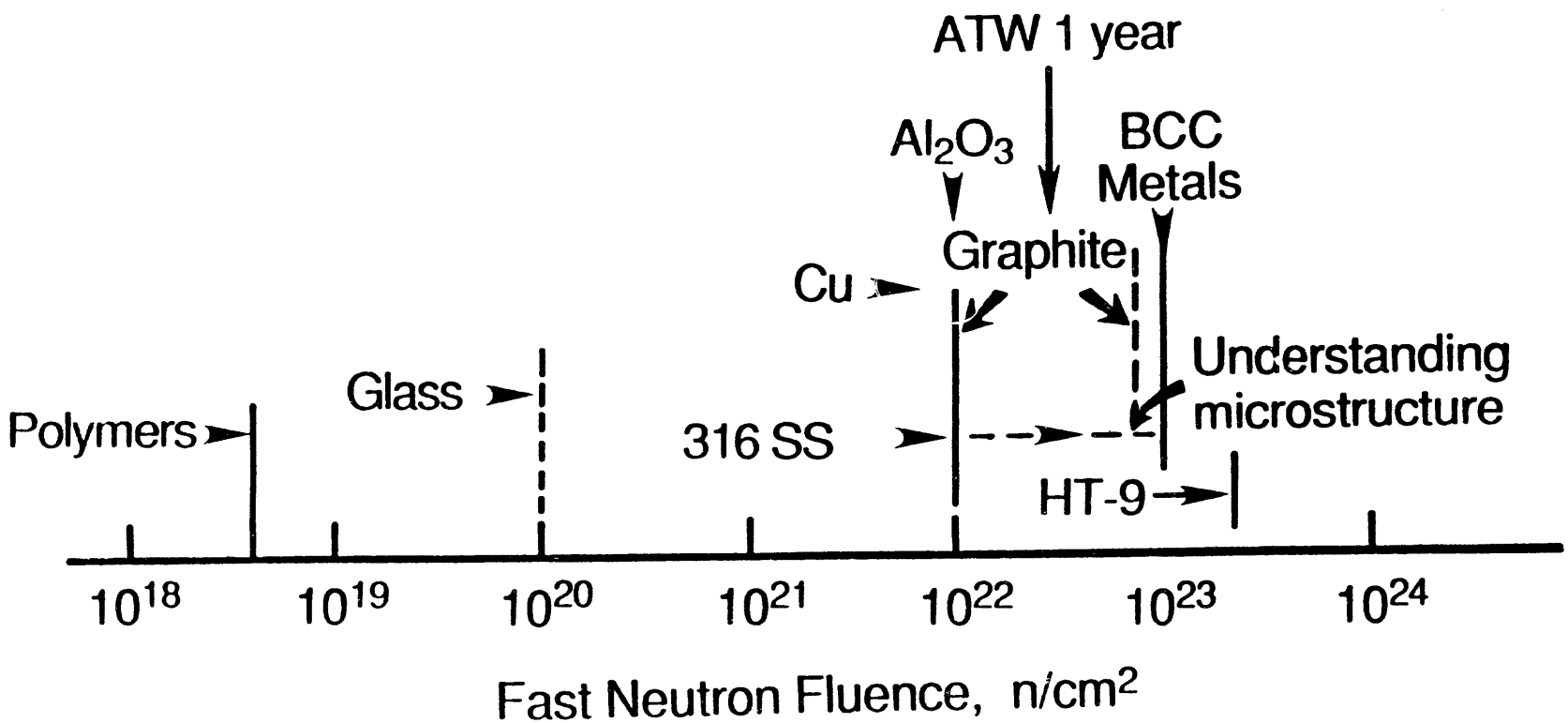


Typical Performance of Materials in a Neutron Environment

Materials respond differently to neutron fluences and have upper limits beyond which irradiation damage dominates and limits the useful service of the material. As one approaches higher and higher fluences, the number of material choices that survive these irradiation environments become very limited. From a materials point of view an order of magnitude increase in fluence at 10^{23} is not the same as at 10^{20}. At the higher fluences the data base is very limited due to the difficulty in obtaining experimental data from irradiation facilities with limited flux capacities. For the 316 SS almost an order of magnitude can be obtained in the fluence level with the appropriate manipulation of the microstructure. The average per year neutron fluence > 0.1 MeV calculated for ATW operated at 1.6 GeV - 55 mA is 4×10^{22} n/cm^2. This is over a liner region length of 20 cm. A peak fluence of 1×10^{23} n/cm^2 was calculated for a smaller portion (<10 cm length). Note: The data base shown is from fast fission irradiation in a reactor. The neutron spectrum in ATW additionally has a component of thermal and very fast neutrons > 5 MeV.

TYPICAL PERFORMANCE OF MATERIALS IN A NEUTRON ENVIRONMENT

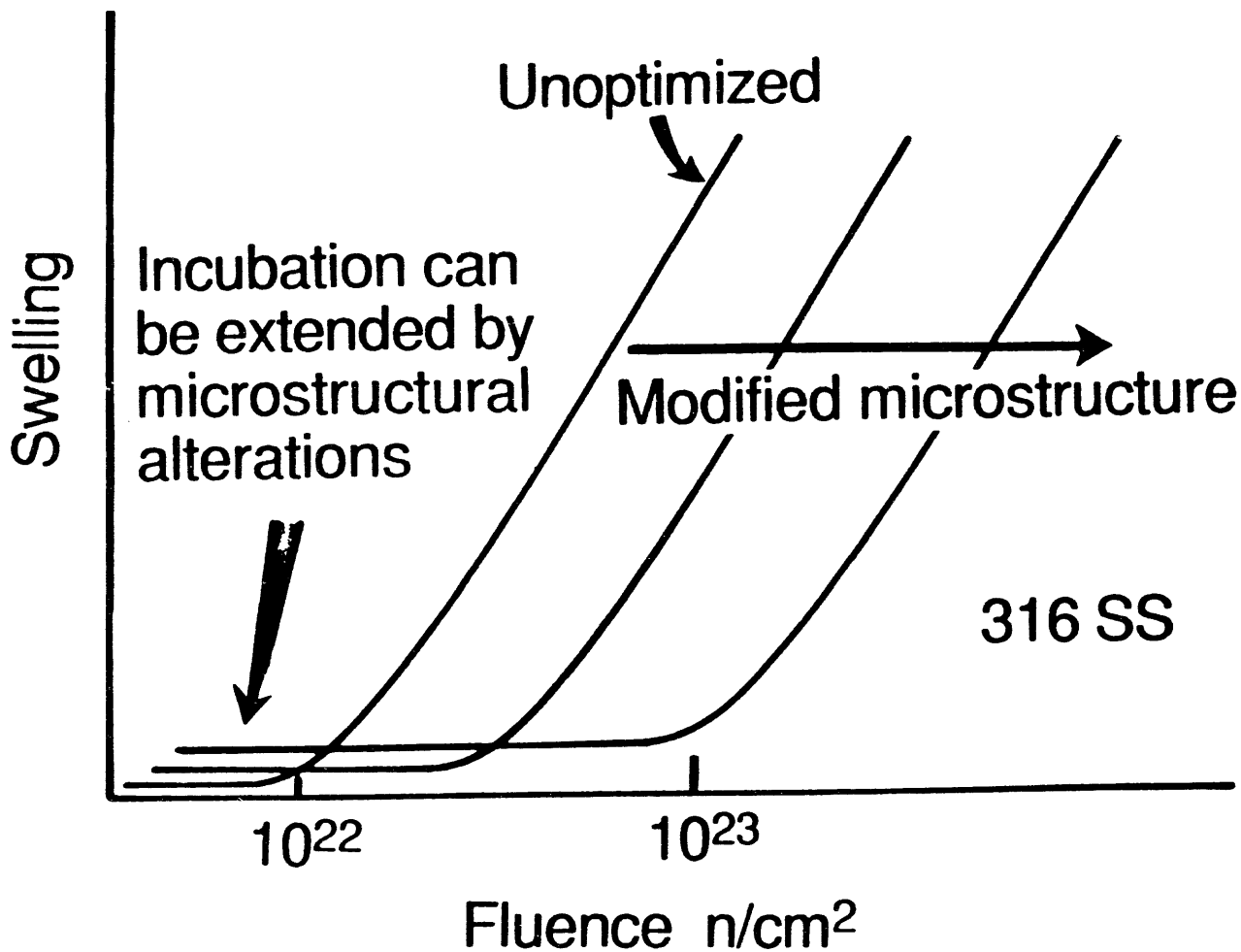
(At higher fluences, limited by data base)



Swelling vs Fluence

Swelling is a strong function of neutron fluence. With some materials such as 316 stainless steel swelling can be delayed by microstructural alteration such as cold working and other thermomechanical treatments. This allows the material to be used at higher fluences before swelling begins. As swelling sets in and continues, the mechanical properties begin to degrade. With large amounts of swelling the mechanical properties go to near zero. Each material has its own swelling behavior and is strongly temperature dependent.

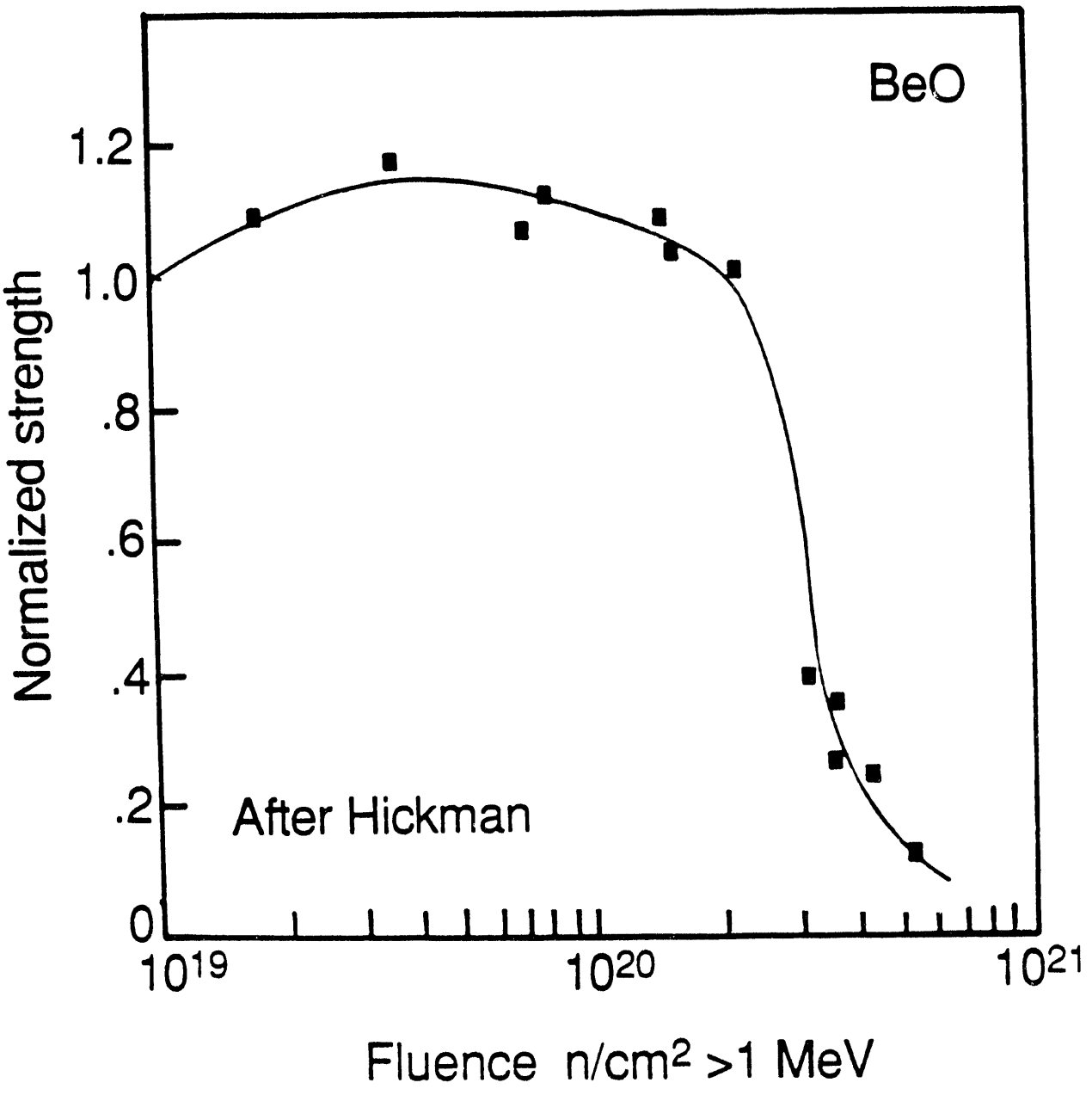
SWELLING vs FLUENCE



Strength vs Fluence

Many materials undergo a loss of strength at a critical fluence range. In the case of BeO, strength is lost due to loss of intergranular strength. In general, metals are more forgiving unless they loose their ductility. Cubic materials are preferred as a rule, due to the absence of anisotropic swelling. In the case of BeO and other specific ceramics, an improvement in strength ('radiation hardening') can be observed over a small fluence range, but very quickly then deteriorates with higher fluences.

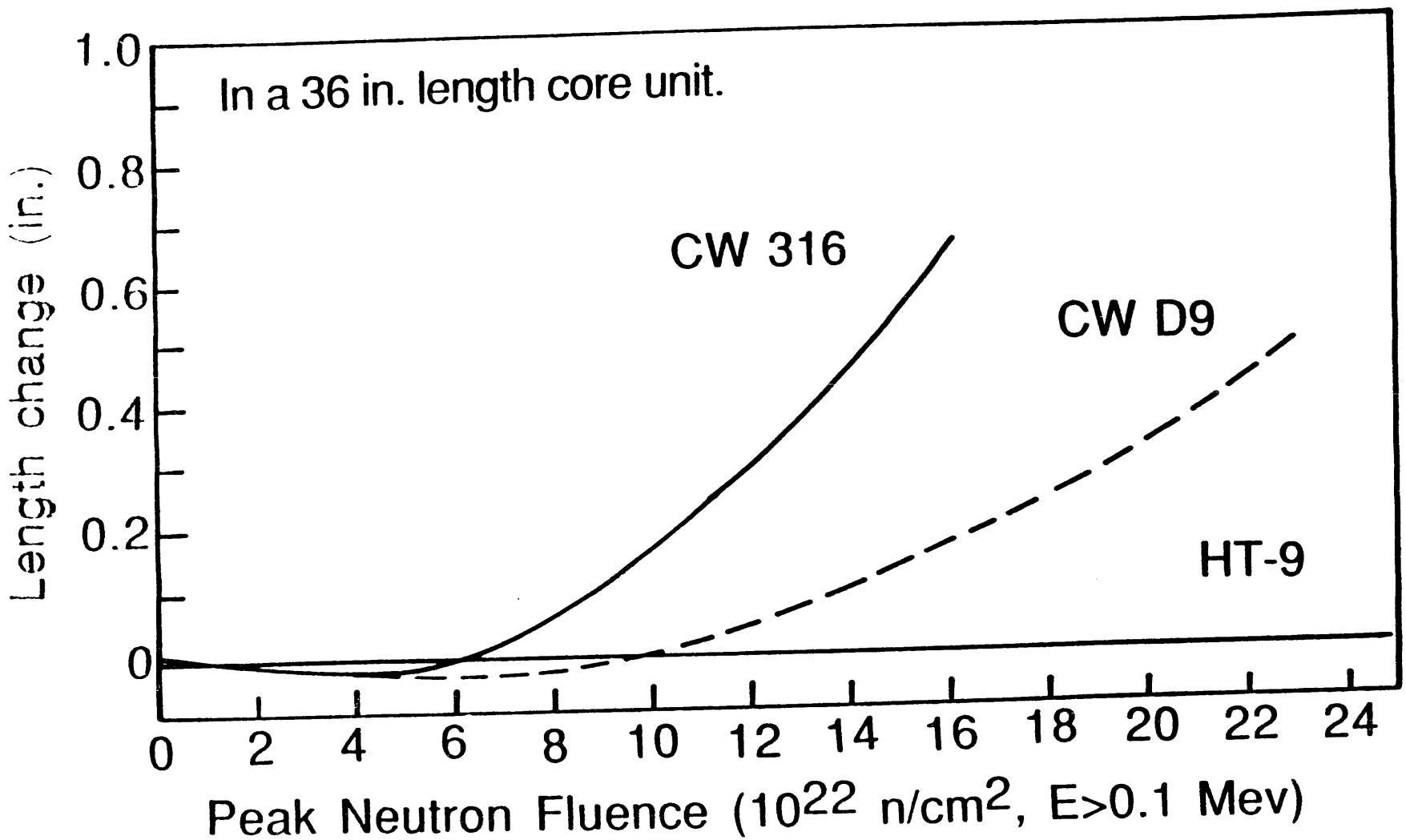
STRENGTH vs FLUENCE



Material Length Changes in FFTF

Newer alloys such as HT-9 appear to show no significant swelling by neutron influences in the range of interest to ATW and possibly beyond. The data is from W. D. Leggett III and R. D. Leggett, Westinghouse Hanford, 1987. However, HT-9 is a high chromium steel and as such would not be directly suitable as a container material for the liquid lead/bismuth. A feasible solution would be to clad HT-9 with a material having similar swelling characteristics

MATERIAL LENGTH CHANGES IN FFTF



Properties of HT-9

The alloy HT-9 is reported to have no swelling in the range of 10^{23} n/cm^2, which exceeds that of the fluence generated in ATW operated at 1.6 GeV at 55 mA for one year. This material is very sensitive to thermomechanical treatments, with proper heat treatment (i.e., austenized, quenched and aged) a relatively ductile steel can be produced. This alloy is commercially available. However, this alloy, as will be shown later, may not be suitable as a stand alone liner due to its high chromium content. Alloys of steel with high chromium contents generally, are severely attacked by liquid lead/bismuth. Consequently, some form of cladding will probably have to be utilized for this alloy.

PROPERTIES OF HT-9

- A ferritic / martensitic steel (BCC)

- Composition (wt. %)

84.81 Fe	11.69 Cr	1.00 Mo
0.51 Ni	0.71 Mn	0.49 W
0.31 V	0.24 Si	0.21 C
bal. Nb, Ti, P, S		

- Mechanical strength similar to 316 SS though somewhat lower

Strength Requirements for Spallation Liner

The strength requirements for the spallation liner are very low. Tolerance criteria are nominal and will only be required to hold a hydrostatic load. The calculated hoop stress of 680 psi is a very low stress and at 400^{o}C is only 3 % of the yield strength. The yield strength for the materials of interest are around 20,000 psi. The major point to be made is that high strength materials are not required. Nor are high temperature materials required. Though toughness requirements are modest, it is still desirable to use a material with the highest toughness available (they are more forgiving).

STRENGTH REQUIREMENTS FOR SPALLATION LINER

- High strength materials not required
- Hydrostatic load only
- Calculated hoop stress = 680 psi
  (~3 % of yield strength at 400°C)
- Toughness requirements are modest.

Use of Si_3N_4 in a Thermal Neutron Environment

Silicon nitride is a very high strength, high toughness ceramic and as such has found its way into many applications requiring these properties particularly at high temperatures. However, in a high radiation environment the material can disintegrate rather quickly. This is due to the nitrogen 14 (the natural abundant isotope). The material is irradiated, the material produces protons which stay in the lattice as hydrogen. This results in swelling and embrittlement. A unique solution which enables neutron irradiation usage, is through the use of nitrogen 15 and is called isotopic tailoring. This technique has produced silicon nitride that is not affected by similar irradiation environments. A note of caution, isotopic tailoring depending on the isotope can be very expensive, however it may indeed be a cost effective solution for limited usage components.

USE OF Si_3N_4 IN A THERMAL NEUTRON ENVIRONMENT

(A very high strength - high toughness ceramic)

$$Si_3N_4$$

$$^{14}N(n_{th},p)^{14}C$$

↑ hydrogen

Solution: Isotopic tailoring

$$Si_3N_4^{15}$$

Corrosion / Compatibility

The major concerns center around long term corrosion on the liner and supporting hardware. These concerns include not only the lead/bismuth compatibility but also that of all the daughter elements produced over the operational life of the spallation source. For lead/bismuth a limited (approximately three years) data base exists, which show no corrosion up to 550^{o}C. However, this data base does not include any daughter products which also have the potential of imcompatibility with the liner material. This point needs to be addressed. In addition, some of the daughter products are volatile and as such may also need attention, particularly, if solubility limits in lead/bismuth are exceeded. These problem areas are discussed in more detail in the following viewgraphs.

CORROSION / COMPATIBILITY

Problems	Solutions
Compatibility with container Pb/Bi	Material selection Coatings
Volatiles	Extract/trap
Compatibility with transmuted products	Materials selection/ removal

Pb/Bi Compatibility Issues

Compatibility issues, particularly for materials that potentially are required to last up to 30 years, is a major question that needs careful scrutiny. Corrosion rates which are very low or are predicted to be very low are essentially all derived from limited time tests, and must be viewed from that point. The issues of mass transport, dissolution and deposition arise from the concern that some solubilities (however small) do exist between the materials used in the spallation system and the liquid Pb /Bi. Concerns for erosion and cavitation along with embrittlement stems not only for the reasons cited above, but also from the daughter products produced over the years. Each of these points are elaborated in the following viewgraphs.

Pb/Bi COMPATIBILITY ISSUES

- Corrosion
- Mass transport
- Dissolution
- Deposition
- Erosion and Cavitation
- Embrittlement

Corrosion/Compatibility Data Base Exists for Liquid Pb/Bi

Liquid lead/bismuth has been used quite extensively for over 20 years as a spallation source and as such, data has been generated in regards to its containment. While many materials appear to work rather well, they are not necessarily usable in an irradiation environment due to the high cross sections (i.e., Ta). In general low alloy steels with > 2.5% chromium appear to do rather well and are cost effective materials.

CORROSION/COMPATIBILITY DATA BASE EXISTS FOR LIQUID Pb/Bi

- Steels with high Ni or Cr(>2.5%) are seriously attacked by liquid Pb/Bi and cannot be used

- Corrosion resistant materials
  - Iron
  - Niobium
  - Tantalum
  - Molybdenum
  - Tungsten
  - Low alloy steels

Low Alloy Steels Tested in Liquid Pb/Bi

A data base for low alloyed steel exists for several alloys that reveal essentially no corrosion for the specified times and test temperature ranges. The temperature ranges are of particular concern because the larger the difference (upper and lower temperatures), the greater the tendency for dissolution and redeposition (i.e., mass transport) of soluble elements.

LOW ALLOY STEELS TESTED IN LIQUID Pb/Bi

ALLOYS	NO CORROSION
• AISI 4130 Steel	at 550-400°C in 10,000 hrs
• RH 1081 Steel	at 550-400°C in 10,000 hrs
• 2.25Cr, 1.0Mo Steel • 1.25Cr, 0.5Mo Steel	at 500-400°C in 10,000 hrs
• 0.5Cr, 0.5Mo Steel	at 500-400°C > 24,000 hrs
• 1.25Cr, 0.5Mo Steel	at 525-425°C in 20,000 hrs

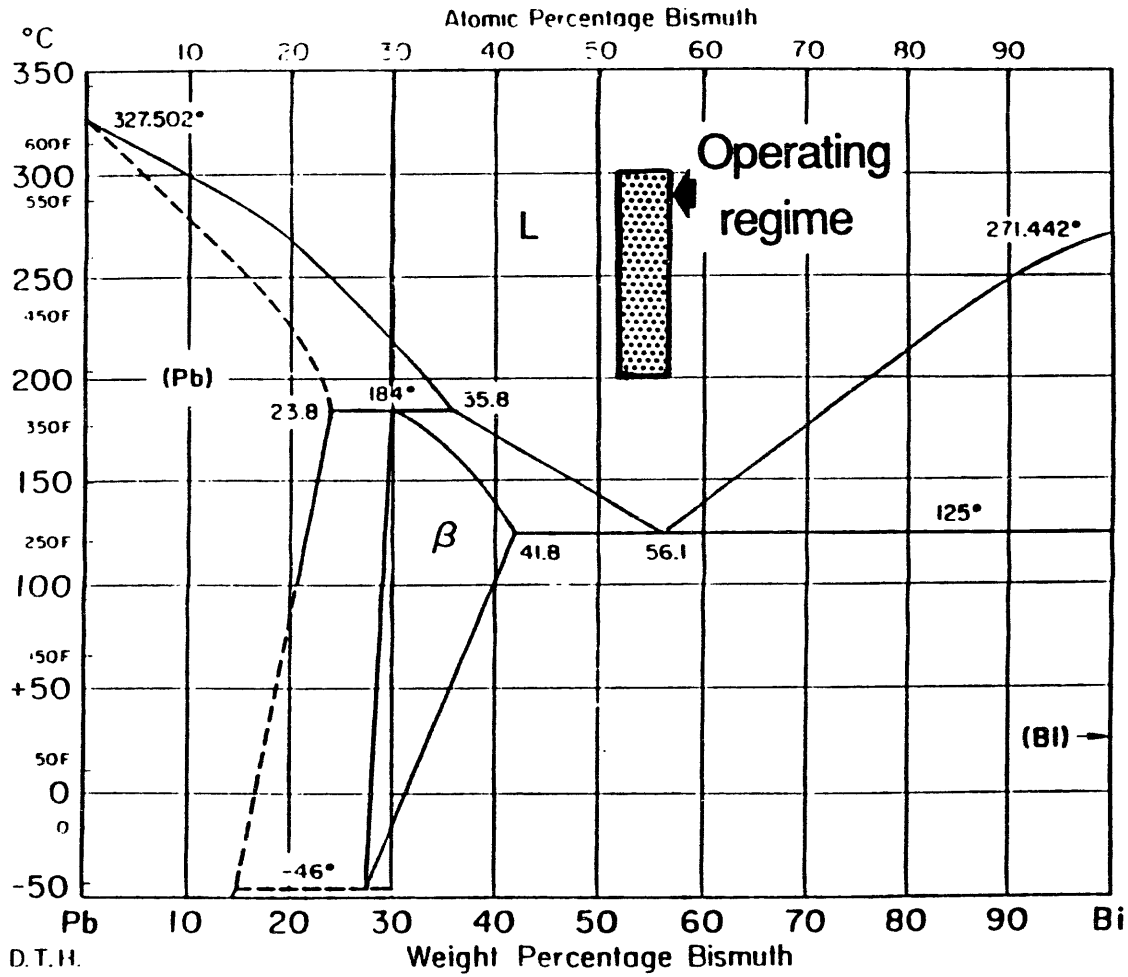
- The smaller the ΔT below 650°C, the longer the life.

- Data base from work at Chalk River, Swiss Inst. for Nuclear Science and Brookhaven.

The Lead-Bismuth Equilibrium Diagram

The lead (Pb), bismuth (Bi) equilibrium phase diagram is shown. Pertinent features of the diagram include, one phase, b, stable to 184^{o}C. A 23.8 wt.% solubility of Pb in Bi and essentially no solubility of Pb in Bi are plotted. A eutectic composition of 56.1 wt.% Bi having a eutectic temperature of 125^{o}C is also shown. The proposed ATW operation parameters are a temperature of 200-400^{o}C with a composition of 45 wt.% Pb - 55 wt.% Bi.

THE LEAD-BISMUTH EQUILIBRIUM DIAGRAM



Possible Solutions for Synergistic Effects

Based on the preliminary data at hand, possible solutions for the synergistic effects of corrosion/radiation effects can encompass several approaches. Initially, cladding of a radiation resistant material such as HT-9 with molybdenum would give the corrosion resistance required for the system. However, eventual swelling of the molybdenum may cause some problems.

A unique design solution would be to adjust the radial heat flow to create a frozen molten interface layer of Pb/Bi on the spallation container material. This would allow for the direct contact use of materials such as HT-9 in the radiation zone, thus having essentially no corrosion in the Pb/Bi as liquid contact can be eliminated.

Other possible solutions would be the utilization of cubic ceramics (they do not undergo anisotropic swelling which limits most non cubic ceramic materials). Additionally istopoically tailored materials such as the si_3N_4 mentioned earlier are also potential solutions.

POSSIBLE SOLUTIONS FOR SYNERGISTIC EFFECTS

- Cladding of radiation resistant materials with corrosion resistant materials
  - Molybdenum on HT-9
- Design
  - Frozen molten Pb/Bi interface layer
- Cubic ceramics
- Isotopically tailored materials

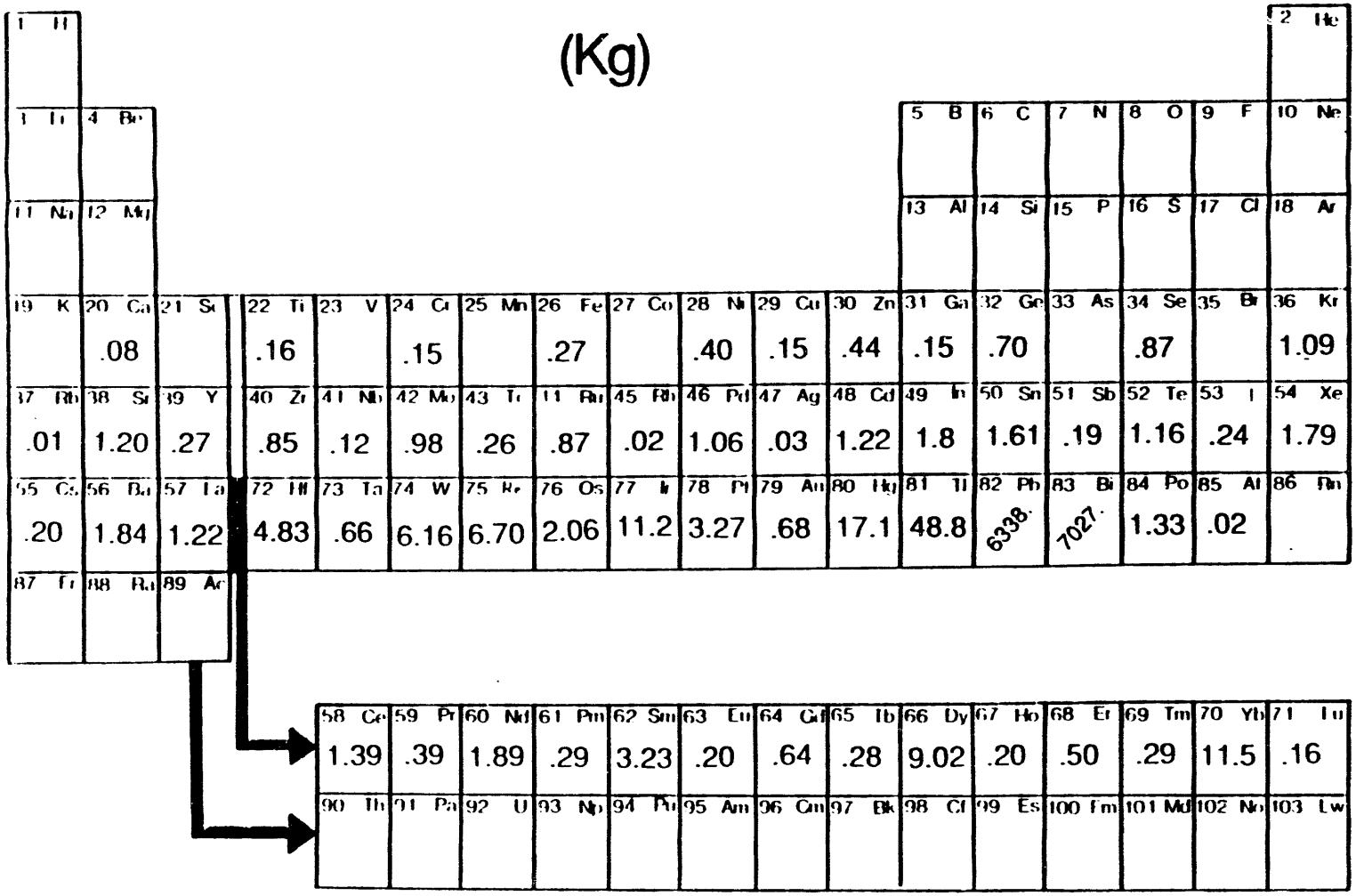
Amount of Transmuted Elements From Pb/Bi Eutectic After 30 Years

For any spallation source to operate for up to 30 years, it is necessary to know the species and amount of transmuted daughter products. The species determines the interactions that need to be accounted for and the amount, to what degree. The produced elements and amounts are calculated by the LAHET code at the operating conditions of 1.6 GeV at 55 mA. The specific concerns which were discussed previously are compatibility/corrosion, embrittlement, and to be discussed, possible compound formation.

AMOUNT OF TRANSMUTED ELEMENTS FROM Pb/Bi EUTECTIC AFTER 30 YEARS

(1.6 GeV-55ma)

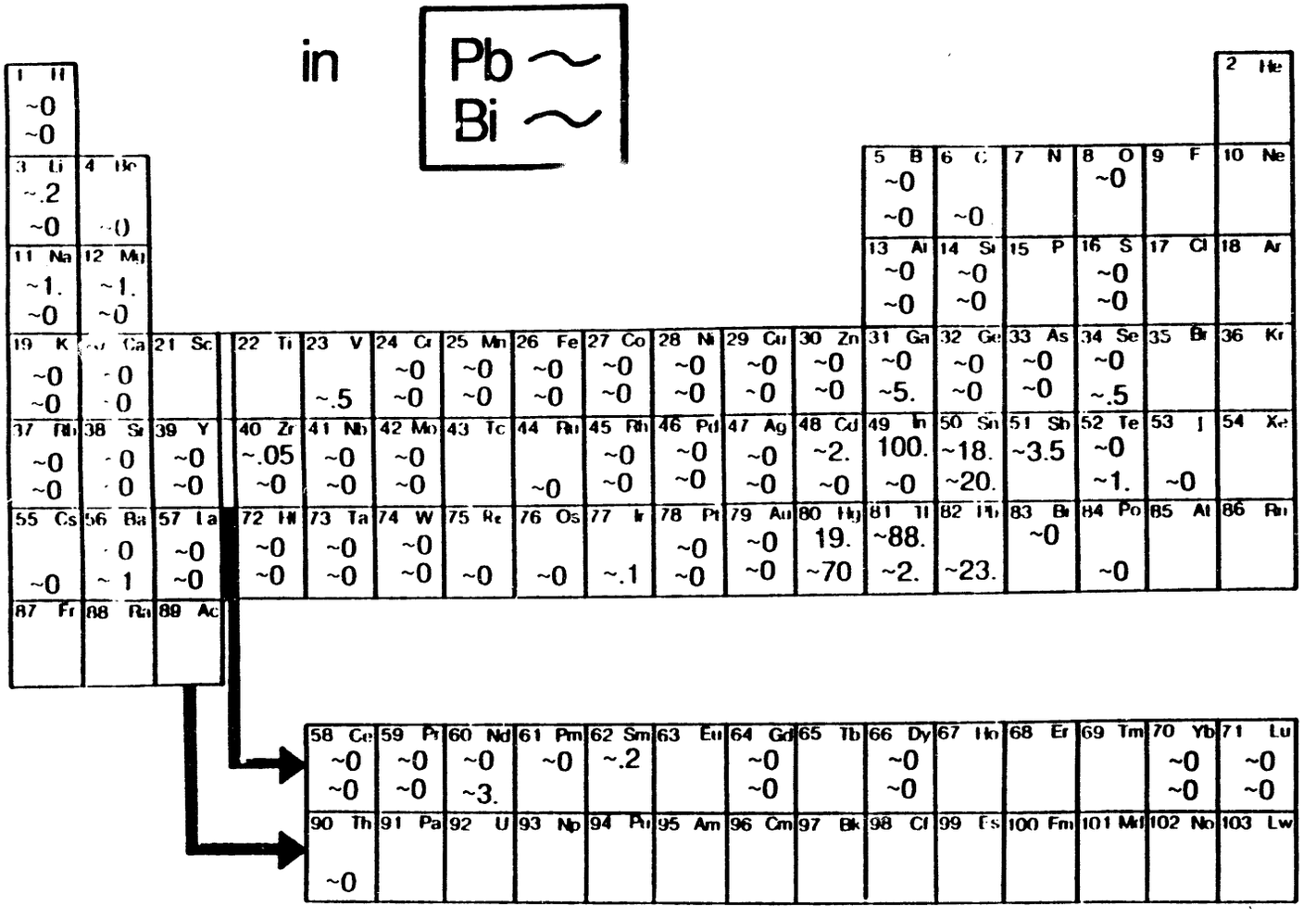
(Kg)



Solubilities (wt. %) At ~ 200°C

At approximately 200°C the known solubilities of the various elements in lead and bismuth are shown. Most elements (except for In, Hg, Tl, Sn, Sb) have relatively low or essentially no solubilities. This is not to imply that some solubilities may exist but that many solubilities have not been evaluated. Here is an area that needs particular focus due to the potential problems that may arise should these elements or their compounds precipitate or plate out in particularly the cold part of a recirculating system.

SOLUBILITIES (wt. %) AT ~ 200°C



Solubility Assessment

Solubility assessment is an attempt to put the question forth in a rational approach using existing data on binary solubilities. This assessment is only a first approximation and indeed may be far more complex, particularly when considering ternary, quaternary, etc systems. In the continual operation of such a spallation system, the possibility of preferred extraction of specific elements, such as Xe, Ar, Au may be desirable (from a nucleonics point of view). In so doing, it alters the daughter production of the remaining system. The net gain would be a reduction of the continual daughter production. Even at the 0.46% unknown or limited solubilities, it is expected that some reasonable fraction would still dissolve in the liquid lead / bismuth. In any event, we must address the possibility that we may have insoluble particles that must be "gathered" at some point in time to prevent potential problems of erosion, plugging, galling, etc. In principle, a simple slip stream containing precipitation / or filtration capabilities (via mechanical or thermal deposition) should suffice.

SOLUBILITY ASSESSMENT

(1.6 GeV - 55 mA)

- Total amount of daughter products at the end of 30 years is 154 kg which is 1.23% of the initial target mass.

- However, elements with known solubilities in Pb or Bi (>0.1) i.e., Tl, Hg, In,... stay in solution

- Thus, elements with low (<0.1) or unknown solubilities amount to 62 Kg which is only 0.46%.

- The average rate of daughter product production is ~2 Kg/yr and per year is only 0.015 % of the initial target mass.

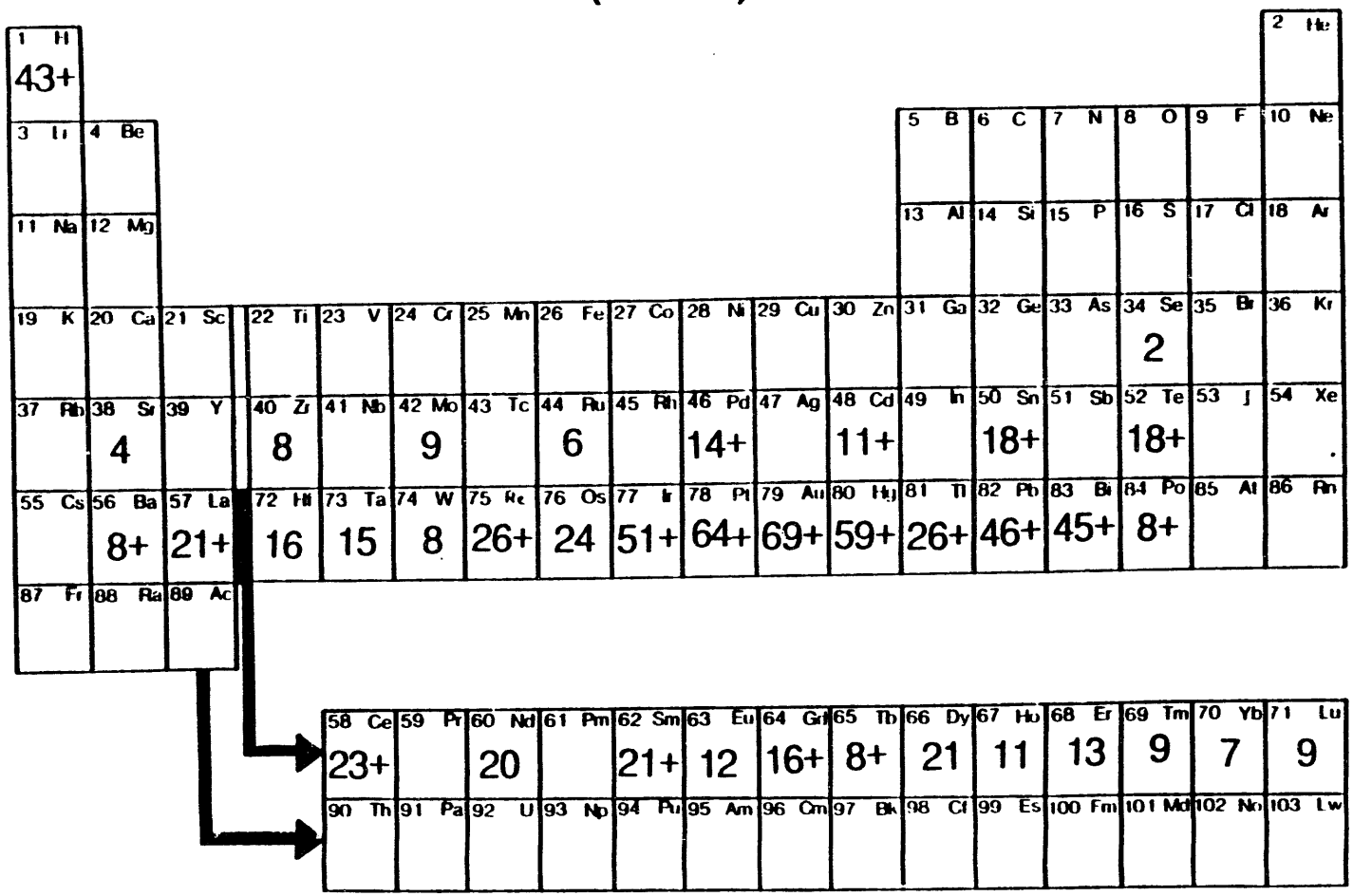
- Must address insoluble particle presence.

Possible Binary Compounds

In trying to assess the possible binary compounds that could exist, a literature survey was made of all the known compounds between lead, bismuth and all the major daughter products (in excess of approximately 1 kg). These are individually listed by each element. This chart is not meant to imply that all of these compounds will form but if compounds were to form, the lowest free energy would dominate and thus could limit further compound formation. The problem is much more complex in that ternary, quaternary and/or higher order compounds could form for which data does not exist.

POSSIBLE BINARY COMPOUNDS

(790+)



Binary Compound Melting Points (^{o}C)

This viewgraph is an attempt to put in perspective the data on known binary compounds that exist between Pb, Bi, and the nine most abundant daughter products. This was done in an effort to highlight potential areas of concern, i.e., high melting point compounds (numbers >400^{o}C), limited solubilities (Im - immiscible elements, many at low temperatures have essentially no solubilities in one another) and elements that are soluble in one another, i.e., SS, but are solid above 400^{o}C. Other designations such as 2C, 4C, 1C, etc. refer to number of compounds in that binary system, though melting point information has not been determined. The symbol E and P refer to a simple eutectic and peritectic equilibrium systems. The concern brought forth in this table is the compounds with melting points > ~ 400^{o}C. These compounds (especially > 1000^{o}C) could eventually pose problems in the design of a recirculating system if not taken into account. The footnote on binary, ternary and quaternary refer to the number of possible combinations in which compound formation could occur.

BINARY COMPOUND MELTING POINTS (°C)

(For the 11 most abundant elements)

	Pb	Bi	Tl	Hg	Au	Pt	Ir	Os	W	Hf
Bi	180									
Tl	P	303 213								
Hg	140	P	14.5							
Au	418 254	373 241	E	420 402 310						
Pt	915 795 360	775 665 660	685	480 245 160	SS					
Ir	1C	1420 1440	-	-	Im	SS				
Os	-	Im	-	-	Im	Im	SS/ Im			
W	Im	Im	-	Im	Im	Im	2540 2490	2945		
Hf	-	-	-	-	>1000 >700	5C	2465 2400 1920 1720	2C	2650	
Yb	2C	6C	4C	-	-	-	1C	3C	-	-

Possibilities: Binary - 55, Ternary - 1980, Quaternary - 57,420, ...

Target Mass by Element vs Time

Based on a time evolution rate of daughter products produced in a liquid Pb /Bi spallation source run at 1.6 GeV - 55 mA, the following table was generated to allow for an experimental simulation as a function of time to be performed. In such a simulation, incremental amounts of each element (in its relation to its production in a spallation source) would be added and heated in a pressure vessel at high temperatures (~1000^{o}C) to promote metallurgical reactions to proceed. This would be repeated, for example as listed in the table, for each two year time interval only listed performed in a one week interval. Subsequently, at the end (30 years or any interval of interest in between) samples can be extracted from the liquid phase and precipitated solids to determine, via analytical techniques, the distribution of the elements. Also solubility limits can be ascertained which would allow or promote deposition and/or compound formation if exceeded. RF sputtering techniques have been devised to simulate chemical stoichiometry and homogeneous distribution throughout a powder mix of Pb and Bi.

TARGET MASS BY ELEMENT vs TIME

wt.%

Δ change from earlier years

ele \ yr	0	2	4	6	8	10	12	14	16	18	20	30
Pb	44.49	44.429	44.477	44.538	44.569	44.644	44.687	44.717	44.735	44.758	44.733	44.583
		-.063	.050	.061	.031	.075	.043	.030	.018	.003	-.006	-.150
Bi	54.51	55.288	55.001	54.684	54.523	54.029	53.915	53.609	53.112	52.823	52.543	51.332
		.788	-.287	-.317	.161	-.494	-.314	-.306	-.297	-.289	-.280	-.211
Tl		.100	.213	.341	.409	.627	.781	.938	1.097	1.258	1.420	2.220
			.113	.128	.068	.218	.154	.159	.159	.161	.162	.800
Hg		.067	.133	.200	.234	.336	.404	.471	.539	.605	.671	.985
			.066	.067	.034	.102	.068	.067	.068	.066	.066	.314
Au						.016	.017	.019	.020	.021	.022	.027
							.001	.002	.001	.001	.001	.005
Pt		.011	.024	.036	.042	.059	.066	.074	.081	.088	.093	.114
			.013	.012	.006	.015	.009	.008	.007	.007	.005	.021
Ir						.013	.017	.021	.025	.029	.034	.058
							.004	.004	.004	.004	.005	.024
Os		.008	.015	.021	.024	.032	.036	.040	.044	.047	.050	.063
			.007	.006	.003	.008	.004	.004	.004	.003	.003	.013
W						.014	.022	.026	.029	.032	.034	.046
							.008	.004	.003	.003	.002	.013
Hf						.021	.025	.029	.033	.036	.040	.055
							.004	.004	.004	.003	.004	.015
Yb		.003	.006	.010	.012	.018	.023	.028	.033	.038	.043	.074
			.003	.004	.002	.006	.005	.005	.005	.005	.005	.031

Research Needs

The areas that would require research attention focuses around the materials problems in an environment of high energy radiation combined with liquid metal compatibility. A large data base exists in the literature. However, the database needs to be examined in greater detail to better define the experimental directions as applied to the material needs for ATW. The question of daughter product compatibility (in spite of their low percentages) also needs further attention, as the number of compounds and their impact on the circulating spallation system is an unknown.

RESEARCH NEEDS

- Materials Optimization - Corrosion resistance that has the ability to withstand the neutron environment and vice versa.
- Liquid metal embrittlement.
- Transport phenomena in the spallation hardware.
- Daughter product compatibility and possible compound formations.

Summary of Spallation Target Materials Issues

Self Explanatory

SUMMARY

- There are many complex issues with synergistic implications. We will require a dedicated technological research effort to address and resolve them.
- The existing data base of compatibility/radiation effects needs further development.
- Many existing technologies have been demonstrated that can be applied to the ATW concept.

OVERVIEW

Overview of Advanced Application

Edward Arthur
Theoretical Division

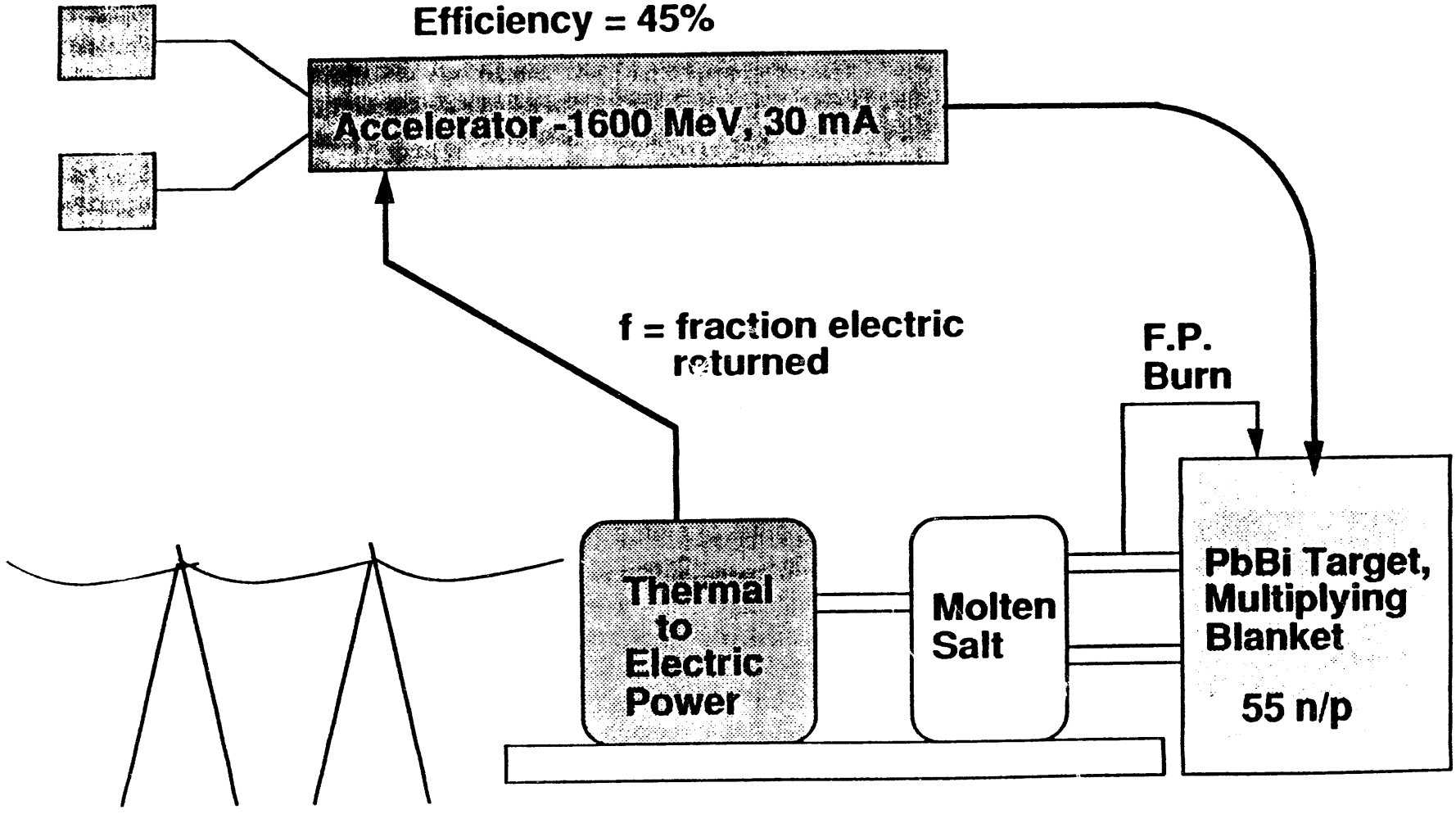
Contents

- The Idea
- Fundamental Enablers
- Features

The Basis for the Advanced ATW Concept

The generic system components that form the basis for the advanced application of ATW -- fission energy with a minimal long-term, high-level waste stream - are shown here. We utilize an accelerator of the class that has been assumed for our defense waste application of ATW. This accelerator would have an overall efficiency of 45 percent. The beam from it is directed into a PbBi spallation target surrounded by a multiplying blanket operated at a k_{eff} of 0.8. As part of the system, we utilize a molten salt loop to handle the fission power and to extract high-grade heat. This loop would operate around 700^0 C so that the overall efficiency for thermal to electric power is 45 percent. Electricity would be generated, part of which would be returned to run the accelerator. The remainder (a majority) would be provided to the commercial grid. Finally, aqueous material flow loops would be used to circulate fission products having half lives greater than 11 years for destruction by transmutation.

The Basis for the Fission Power With a Minimal HLW Stream Concept



Thermal to Electric Efficiency = 45%

Enablers for the Advanced Concept

The principal enabler of the advanced ATW concept is the fundamental impact that spallation neutron generation can have on fission systems. As will be explained in the next presentation, use of an accelerator to produce spallation neutrons which are then used to drive a multiplying blanket effectively increases the number of neutrons per fission by fifty percent. In addition, the features of the ATW system as discussed -- high flux, low inventories -- are fundamental to the conceptualization of a fission source capable of transmuting its long-lived fission product waste. Finally, the feature of continuous material flow enables us to achieve cesium isotope separation using natural decay features of xenon precursors. Utilization of this phenomena is not possible under standard reactor operating conditions.

Enablers

- Fundamental impact of spallation physics on a fission system

50% increase in #n/fission

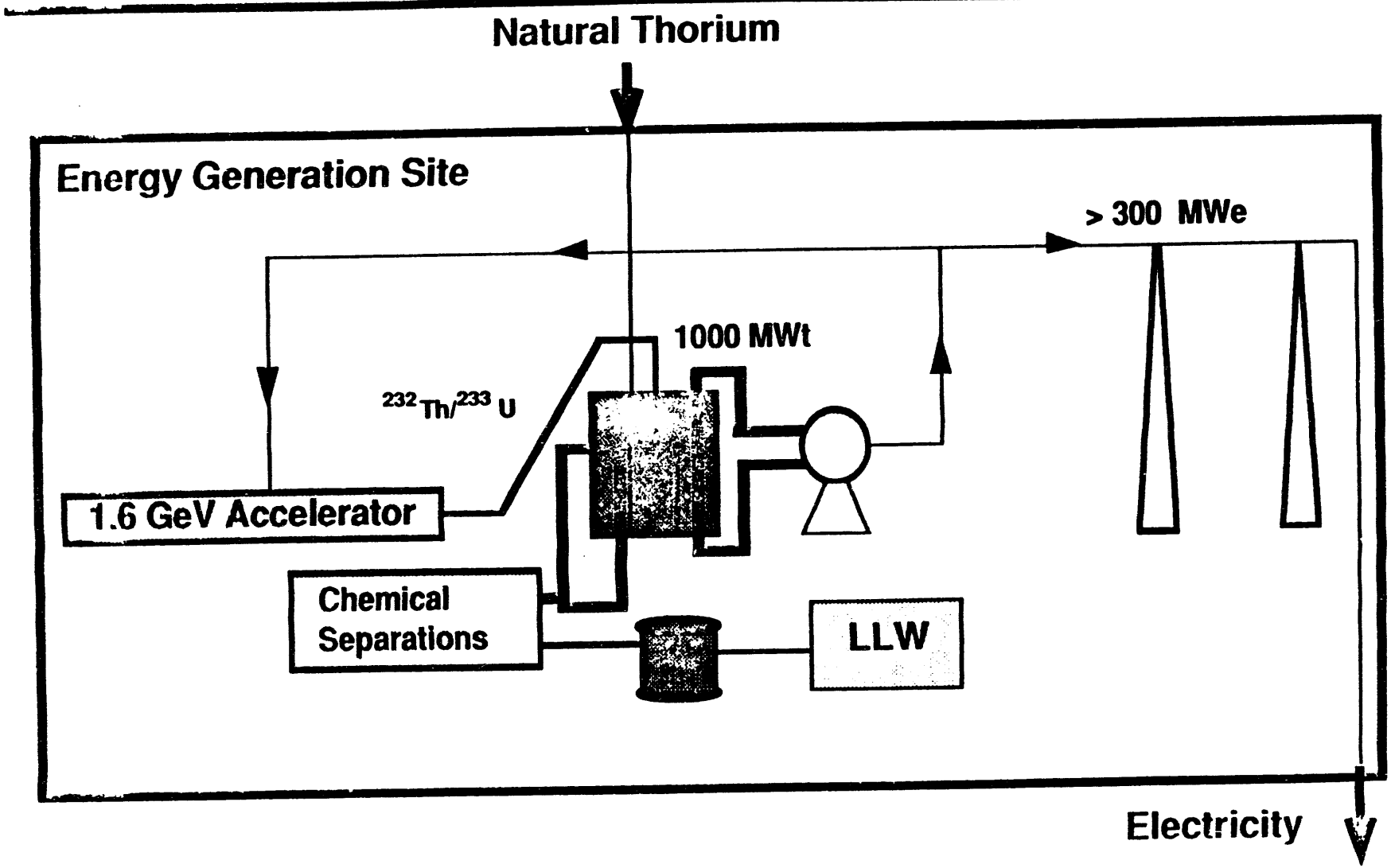
- High flux, dilute system features
- Continuous materials flow and extraction

The Advanced ATW Concept - Fission Energy With A Minimal Long-Lived, High-Level Waste Stream

This schematic illustrates the main features. Natural thorium is introduced into the blanket where ^{233}U is bred, *in situ*, from it. This ^{233}U is reintroduced into the system in a molten salt loop which is operated at a thermal power level of 1000 MW. Heat from the molten salt loop is converted to electricity, 48 megawatts of which is needed to power the accelerator. The remaining amount (> 300 Megawatts) can be supplied to the commercial grid.

During this process, fission products created during ^{233}U fission are extracted and reintroduced back into the system for transmutation. Transmutation of approximately ten percent of the products produced during fission is sufficient to ensure that a minimal high level waste stream results from the systems operation. Fission products with half lives greater than 11 years would be transmuted. The remainder would be extracted from the system and allowed to decay during short-term storage on site.

The Advanced ATW Concept



Features

Our initial analysis of this system, based on neutron economy arguments, indicate that net power production in excess of 300 MW_E would occur. The overall efficiency of the system is around 33 percent. This high efficiency is possible because of the 45 percent thermal-to-electric conversion efficiency of the molten salt.

Features

- Thorium fueled
- ^{233}U breeding ($^{233}Th \longrightarrow ^{233}Pa \longrightarrow ^{233}U$)
- Molten LiF/BeF_2 salt for high thermal to electric conversion
- Multiplying blanket @ 1000 MW_T
- Net Power Production $\geq$ 300 MW_E

Features (Continued)

In the system we would transmute all fission products with half lives greater than 11 years. The system utilizes three flow loops for this fission product transmutation (an aqueous loop), ^{233}U breeding (aqueous), and ^{233}U fission (molten salt).

Features (Cont)

- Transmutation of all fission products with $\tau_{1/2} \geq 11$ years (8 isotopes)
- Two classes of flow loops
  - Aqueous - ^{233}Pa breeding
    Fission product burn
  - Molten Salt - ^{233}U fission

Three Separate Flow Loops

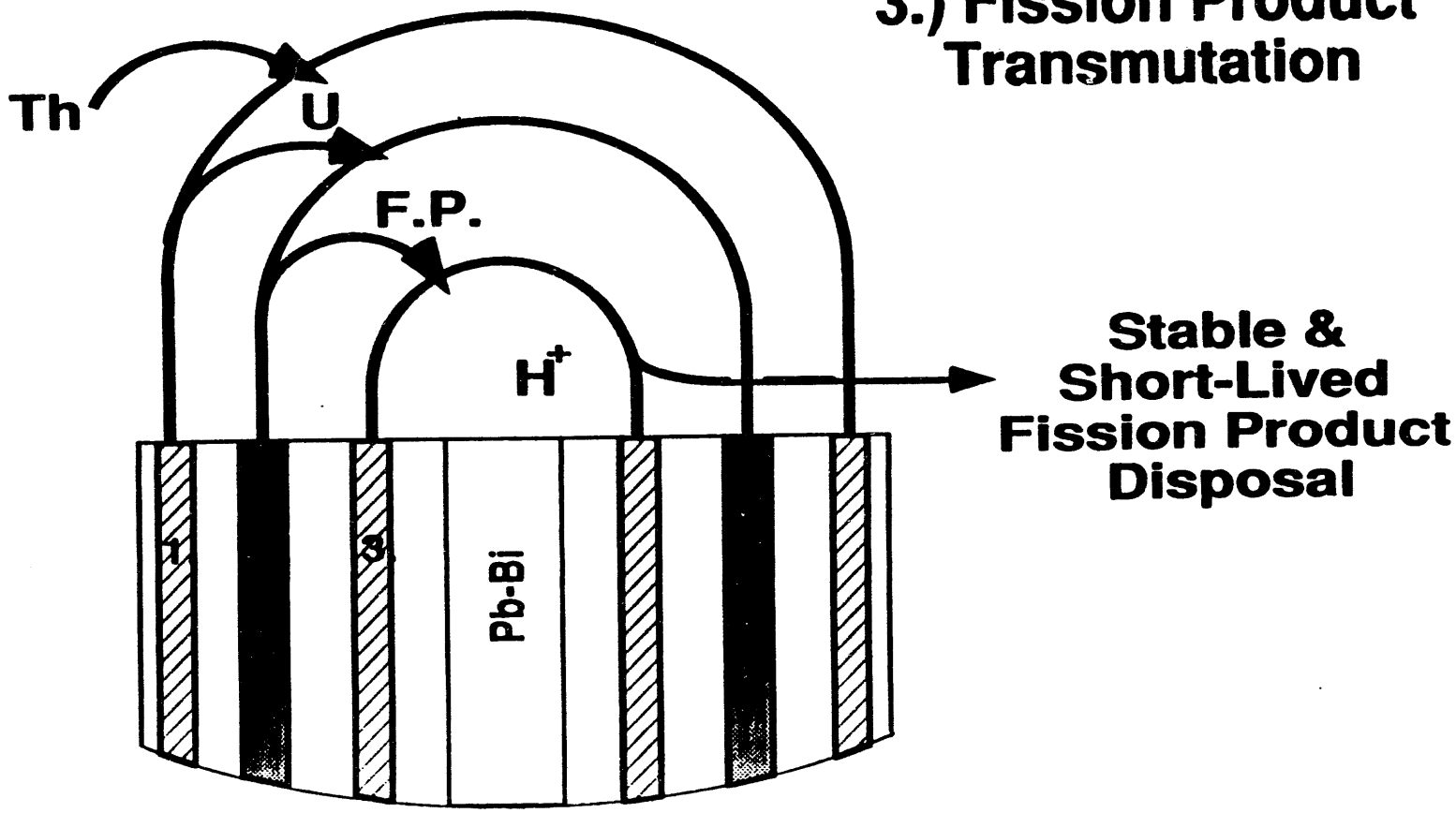
The flow loops described earlier are illustrated schematically.

Three Separate Flow Loops Are Present in the ATW Advanced Concept

1.) U233 Breeding

2.) U233 Fission

3.) Fission Product Transmutation



Fission Products to Be Transmuted in an Advanced System

This list illustrates the fission products to be transmuted in this concept. These comprise the overwhelming majority of longer-lived, high-level waste produced in the system. Higher actinide contributions are virtually nonexistent because of the use of the thorium-^{233}U fuel cycle.

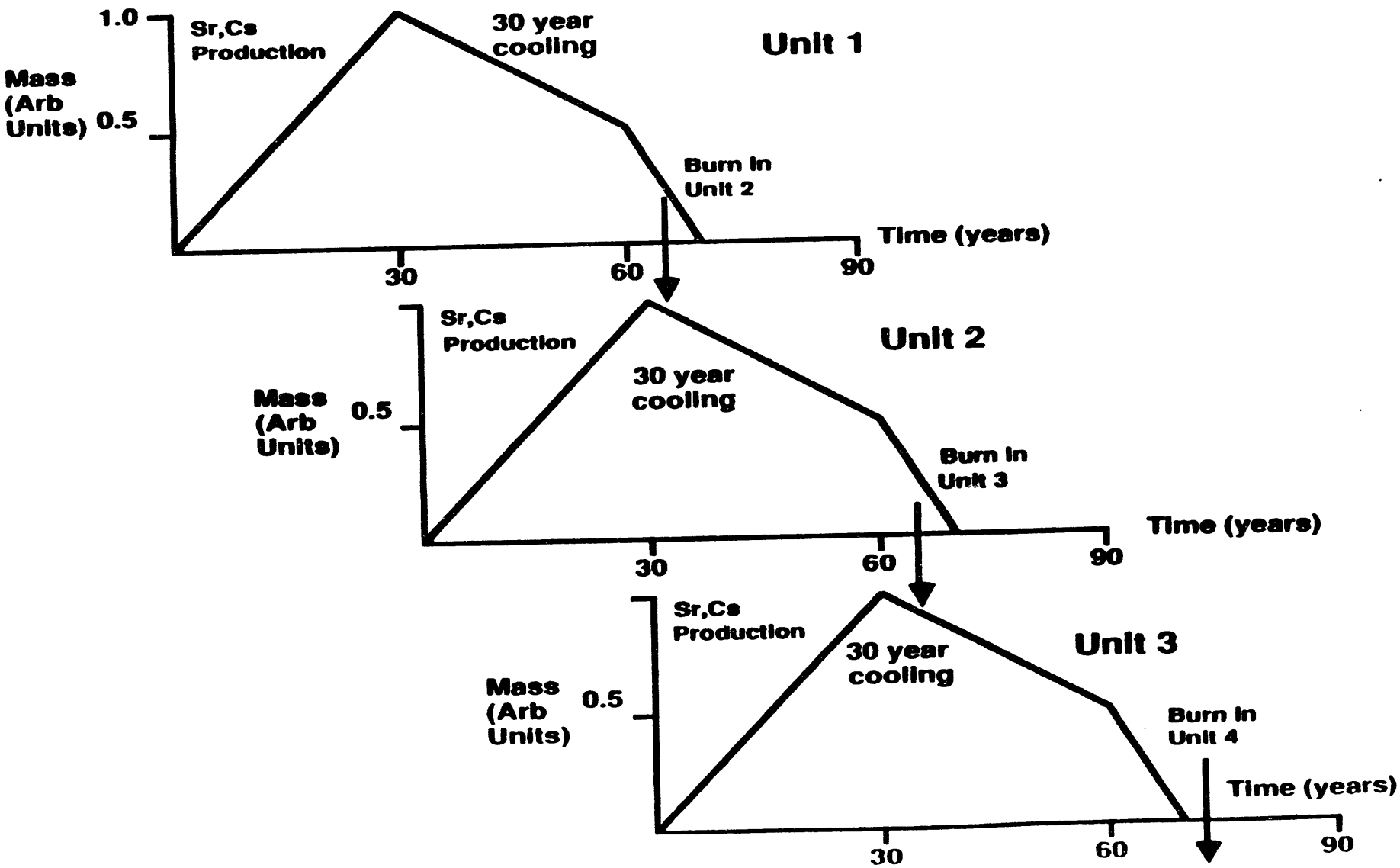
The Following Fission Products Would Be Transmuted or Burned Internally

Isotope	Half Life (Years)
Se79	65,000
Sn126	100,000
Cs135	3,000,000
Sm151	90
Tc99	200,000
I129	16,000,000
Sr90	30
Cs137	30

Burning of Sr and Cs Inventories

The approach used to transmute ^{90}Sr and ^{137}Cs is illustrated here. During the 30 year lifetime of the system these waste products are produced in the fashion shown. The wastes produced by one system would be cooled for one half life (30 years) to reduce their inventory by a factor of two. During this cooling period a second facility would be brought on-line. The inventory from the first system remaining after cooling would be introduced into the second system for transmuting. This inventory would be transmuted over a period of time of about 5 to 10 years. Concurrently, inventories building up from the second system operation would be cooled and introduced into a third system for transmutation.

Burning of Sr,Cs Inventories Would Occur After a 30 Year Cooling Period



Summary

An advanced energy production system based upon the ATW system would use natural thorium as a fuel. This could ensure a long-term energy supply because of large reserves of thorium that are expected to last thousands of years. During operation of such a facility, the major high-level waste would be burned concurrently, so that a minimal long-lived, high-level waste stream would result from the system.

The ideas embodied in the system described here are direct extrapolations of technologies developed in areas associated with the near-term ATW application to defense waste transmutation. The principal areas of development would be associated with molten salt loops utilized for ^{233}U fission.

Summary

- Use of natural thorium
- Long-term energy supply
- Minimal high-level, long-lived nuclear waste stream
- Direct extrapolation of near-term ATW technologies

ADVANCED TECHNOLOGY

Fission Energy Without a High Level Waste Stream

Charles Bowman
Physics Division

Objectives

In our concept this system would convert the thorium and/or uranium resources to fissile material and burn it to produce electric power.

This power can then be used, in part to drive the accelerator. The accelerator/target system produces a surplus of neutrons which can be used to convert fertile to fissile material as well as to burn fission products with half lives greater than 11 years. The transmutation of these fission products minimizes the high level waste stream associated with other concepts. The system is far sub-critical and, therefore, avoids the possibility of criticality excursions.

Fission Energy Without A High Level Waste Stream Objectives

Inexhaustible energy supply

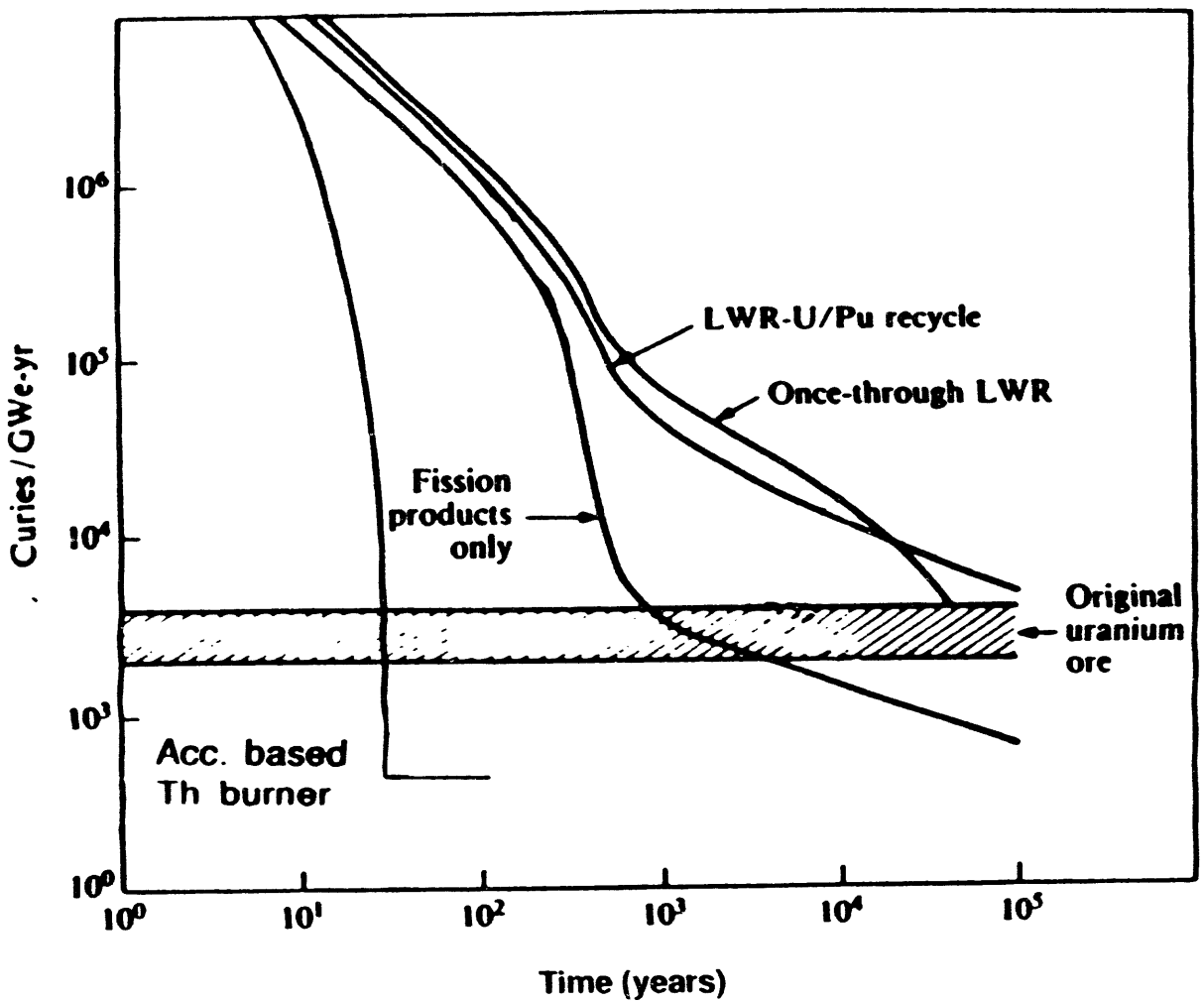
No criticality excursions

Minimal high level waste legacy

Fission Waste Decay Rates

This graph shows radioactivity vs time for spent fuel from commercial power reactors. The top-most curves show the decay rate for the once-through light water reactors and the decay rate with reprocessing and burning of the plutonium. The band shows the radioactivity of the original uranium ore, which is a goal for the concept described here. The transmutation of the higher actinides along with a few long-lived fission products would result in the curve labeled "fission products only" which reaches the ore objective in several hundred instead of about 100,000 years. We propose a device which generates nuclear power, transmutes all of the higher actinides, and also transmutes all fission products with half-lives greater than 11 years, so that the activity reaches the objective in a time comparable to the human life span.

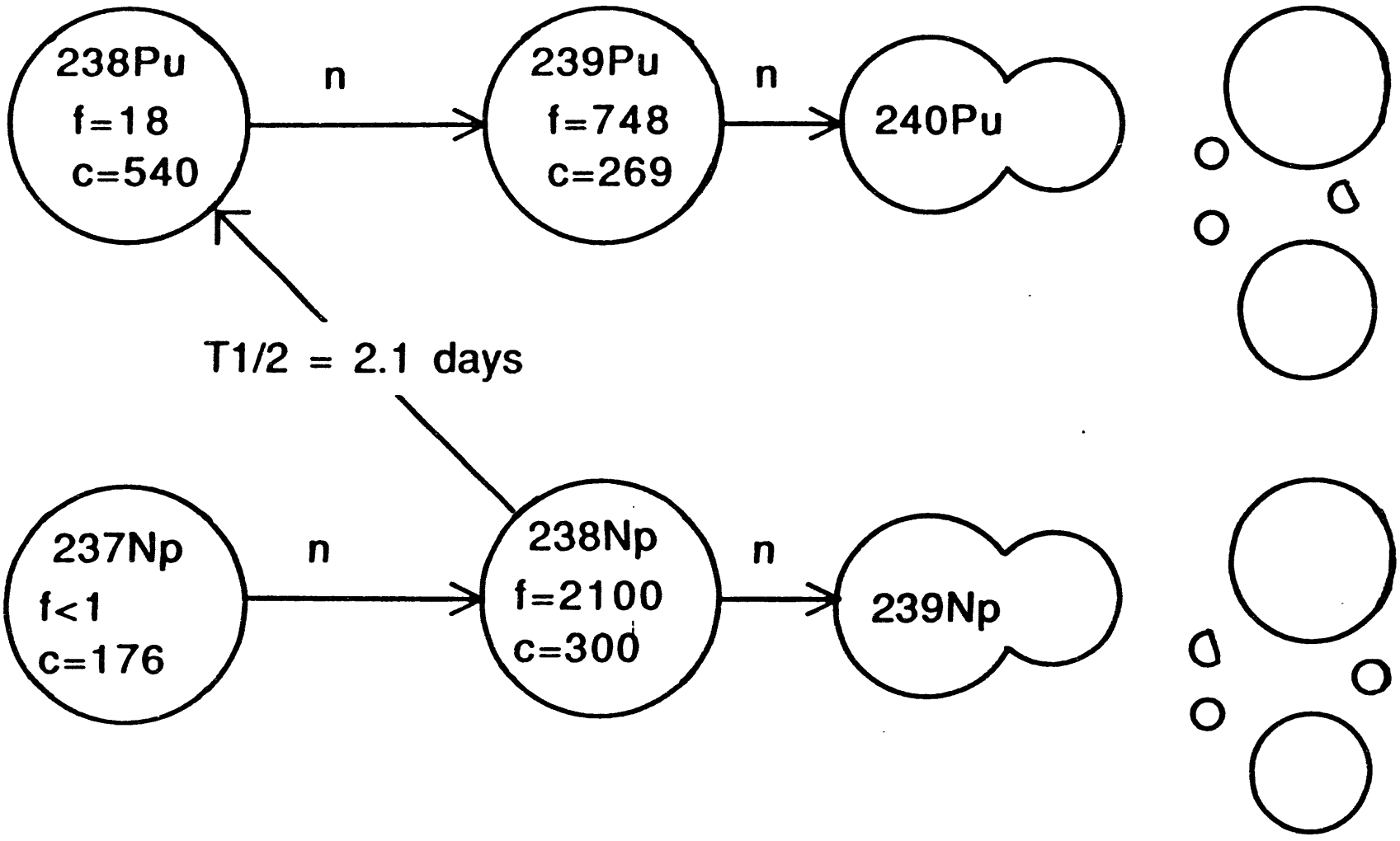
Fission Waste Decay Rate



High Flux Thermal Fission of Higher Actinides

The transmutation using thermal neutrons of higher actinides such as ^{237}Np, which has no thermal fission cross section, is illustrated. In the figure, f refers to fission cross section and c to capture cross section. In a low thermal flux the ^{237}Np nucleus absorbs a neutron to ^{238}Np which decays, in spite of its high cross sections, before a subsequent absorption to ^{238}Pu. The ^{238}Pu fission cross sections are low compared to capture so that most of the material is transmuted to ^{239}Pu. Upon absorption of a neutron in ^{239}Pu, 3/4 of the compound nuclei formed fission with the release of two fragments and about 2.8 neutrons. By counting neutrons absorbed we see that a total of three neutrons were absorbed while only 2.8 X 3/4 = 2.1 neutrons were liberated. Since more neutrons were absorbed than emitted, a chain reaction based on this process cannot be self-sustaining and ^{237}Np is a poison in a low flux. However if the flux is near 10^{16} n/cm^2-s, the ^{238}Np nucleus absorbs another neutron before it decays to ^{238}Pu. It then fissions with the production of about 2.8 neutrons. Since only two neutrons were absorbed in this high flux process, more neutrons were generated than absorbed so that in the high flux ^{237}Np is a fuel rather than a poison.

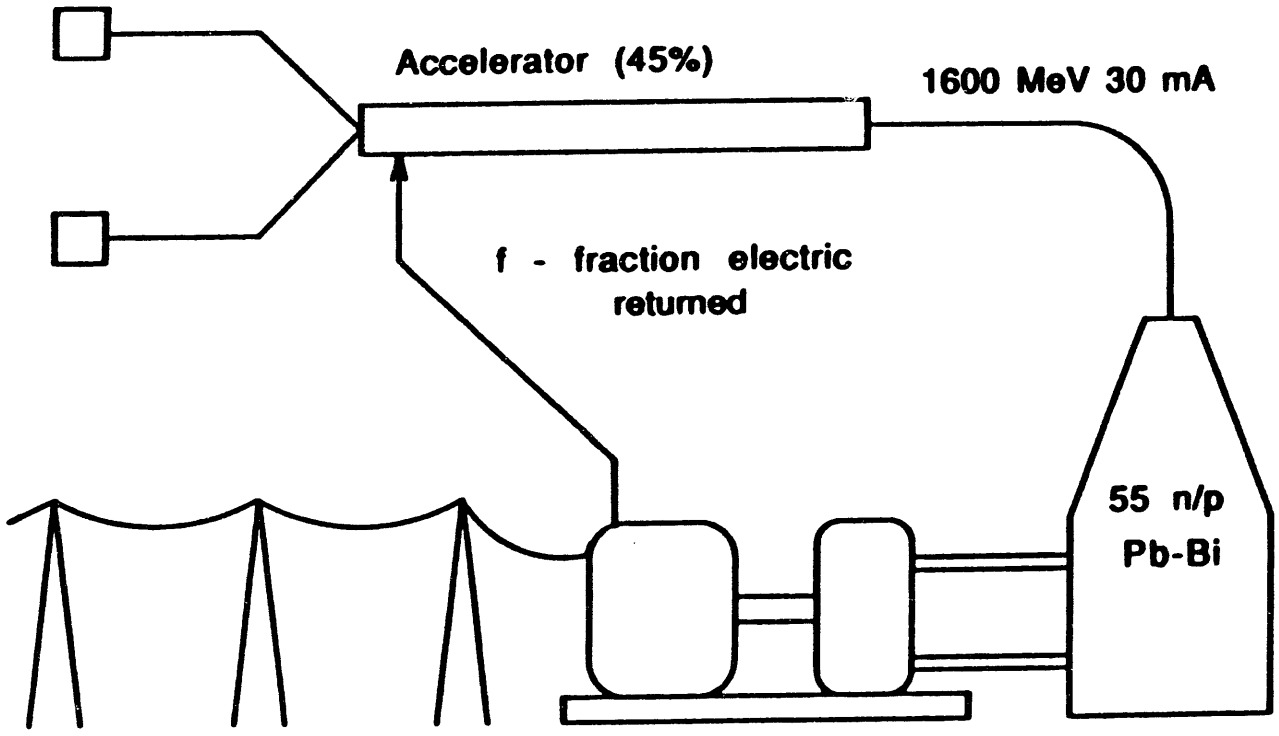
High Flux Fission of Higher Actinides



Accelerator-driven Target Blanket with Power Generation

A 1.6 GeV proton accelerator generates a high flux beam that impacts a molten Pb-Bi target, producing 55 neutrons per proton. These neutrons induce fission in fissile material carried by molten salt. The high temperature of the molten salt makes possible a high thermal-to-electric conversion efficiency of 44 %. Some electric power is fed back to the accelerator, which operates with an electrical efficiency of 45 %.

Accelerator-Driven Target-Blanket with Power Generation



Fundamental Impact of Accelerator Neutron Sources

The average number of neutrons from fission is a fixed number, typically about 2.5, and all nuclear reactor design has had to work within the limitations prescribed by this number. Introduction of the accelerator as shown in the last transparency changes things fundamentally. If all the electric power generated from the target is fed back into the accelerator the effective number of neutrons per fission may be increased by about 50 %. The transparency shows that 500 fission events in the target, taking into account the efficiencies, will produce 12.4 1600-MeV protons which in turn will produce 680 neutrons. The 500 fission events will produce 500 X 2.5 = 1250 neutrons. The total of 1930 neutrons is 50 % larger than the neutrons produced in fission alone. These extra neutrons may be used for purposes impossible to achieve with a reactor alone-such as destroying fission product waste. Not all of these neutrons are required for burning the fission product waste produced in the target; the electric power not used by the accelerator can be sent to the power line.

Fundamental Impact of Accelerator Neutron Sources

Example: 500 fission events

Heat energy	100,000 MeV
Electricity conversion (44%)	44,000 MeV
Beam power (45%)	19,800 MeV
# 1600 MeV protons produced	12.4
@ 55 n/p number of neutrons produced	680 neutrons
@ 2.5 neutrons/fission	1250 neutrons
For 500 source fissions ⟶	1930 neutrons TOTAL

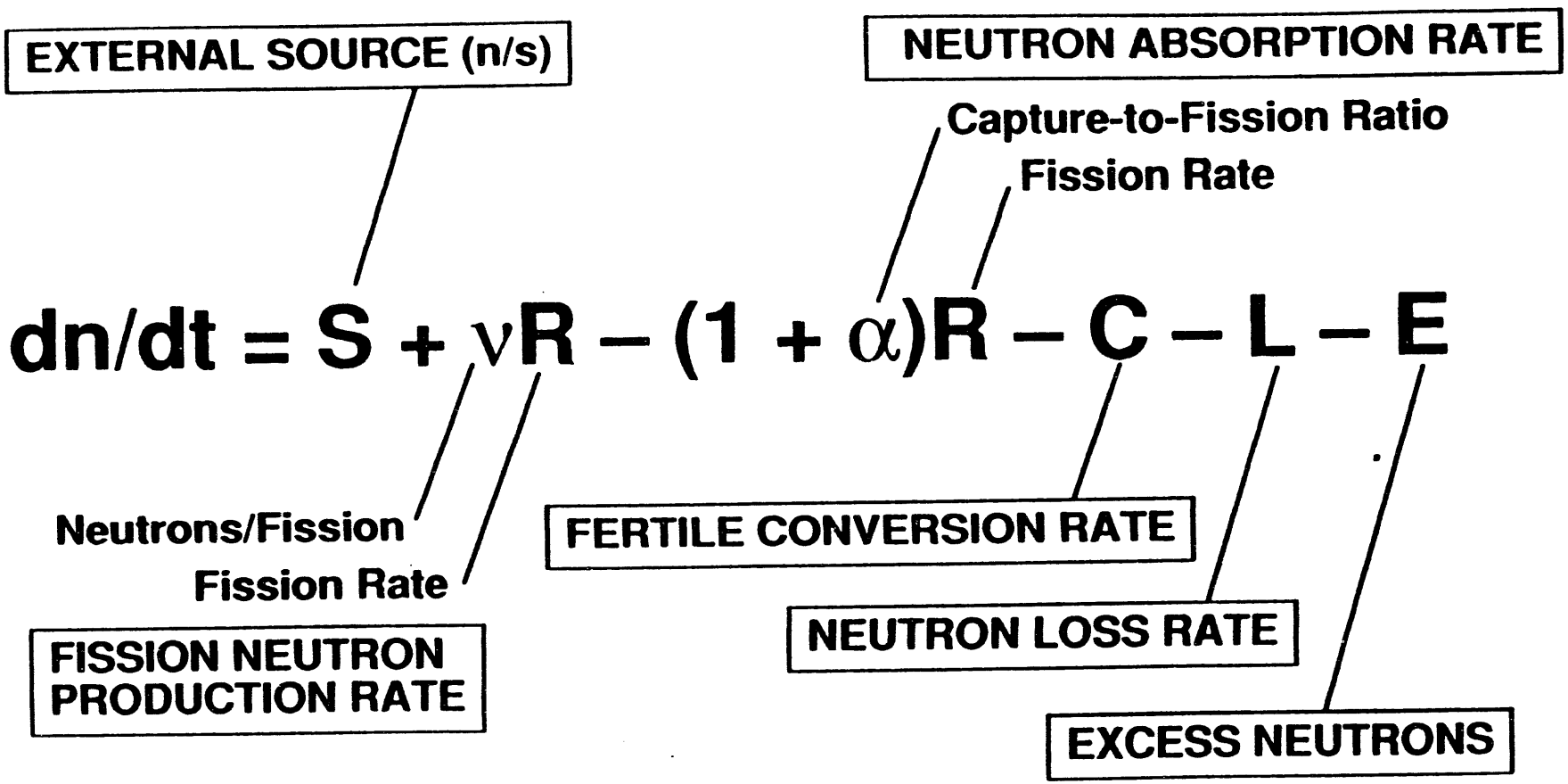
The effective neutrons/fission is 3.8 versus 2.5 in a standard reactor system - a 50% increase.

Neutron Economy Equation

This equation with simple and straightforward modifications can be applied to any reactor or accelerator-driven system for evaluating performance.

The time dependence of the total number of neutrons/cm^3, n, in an infinite medium assuming thermal neutrons only is given by six terms as explained. The term C is the loss of neutrons from conversion of fertile to fissile material-for example ^{232}Th to ^{233}U. It is the same as the neutron absorption rate term for inducing fission. The term L is the loss rate from parasitic capture, leakage, absorption in control rods, etc. Since we use molten salt in this system, we assume that we can achieve the same neutron loss rate as was achieved in the Molten Salt Breeder Reactor program at ORNL in the 1960's, which was 13 % of the S + νR neutron source terms. The last term E is the excess neutrons available for burning fission product waste. Note that there are no cross sections, only ratios of cross sections. For the steady state, the equation can be set equal to zero and for a given fission rate the accelerator-produced neutron source term can be calculated for providing the number of excess neutrons required for waste burning. Knowing the efficiencies already given it is possible to determine the amount of power which must be used for the accelerator. This equation is applied in the next two transparencies.

Neutron Economy Equation

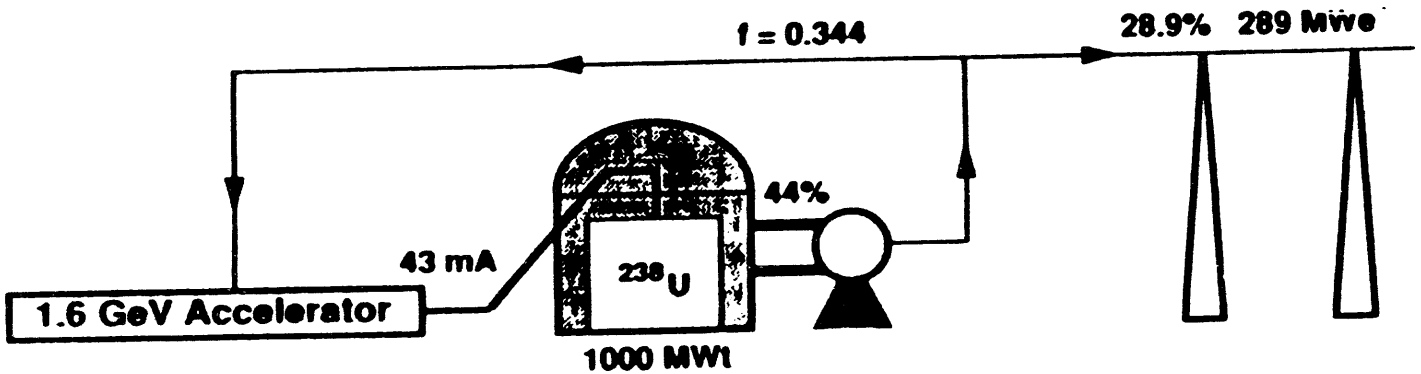


Accelerator-transmuter Performance Derived from Neutron Economy

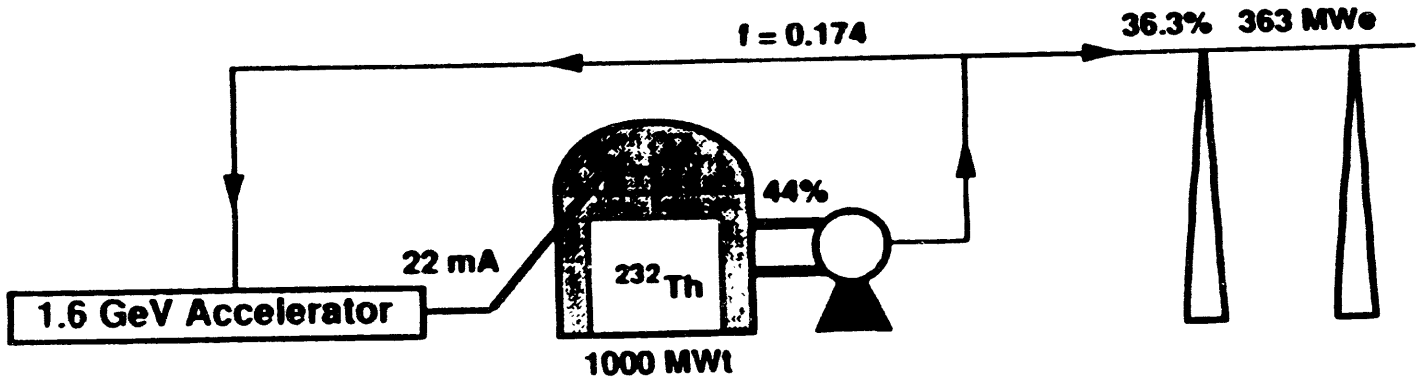
For both examples in the figure it is assumed that all of the long-lived fission products ($T_{1/2}$ > 11 years) generated in the target-blanket are transmuted with the ^{137}Cs and ^{90}Sr decaying for 60 years before transmutation begins for them. The upper system is for the ^{238}U-^{239}Pu cycle. It is assumed that the accelerator drives the target at a power of 1000 MWt which is converted to 440 MWe. The neutron economy equation says that the accelerator must produce 43 mA of current to produce this much fission power and burn its waste as well. Therefore, the fraction 0.344 of the 440 MWe must be diverted to the accelerator. The remainder of the power, 289 MWe can be put into the power line. The overall efficiency for power production, including the reduction for powering the accelerator, is therefore 28.9 %. For the ^{232}Th -^{233}U cycle, the accelerator need be only half as large and the overall efficiency is 36.3 %. The ^{233}U system is better only because the thermal capture-to-fission ratio is more favorable for ^{233}U than for ^{239}Pu.

Accelerator/Transmuter Performance Derived from Neutron Economy

1. Energy Production from ^{238}U Without Long-Term Fission-Product Storage



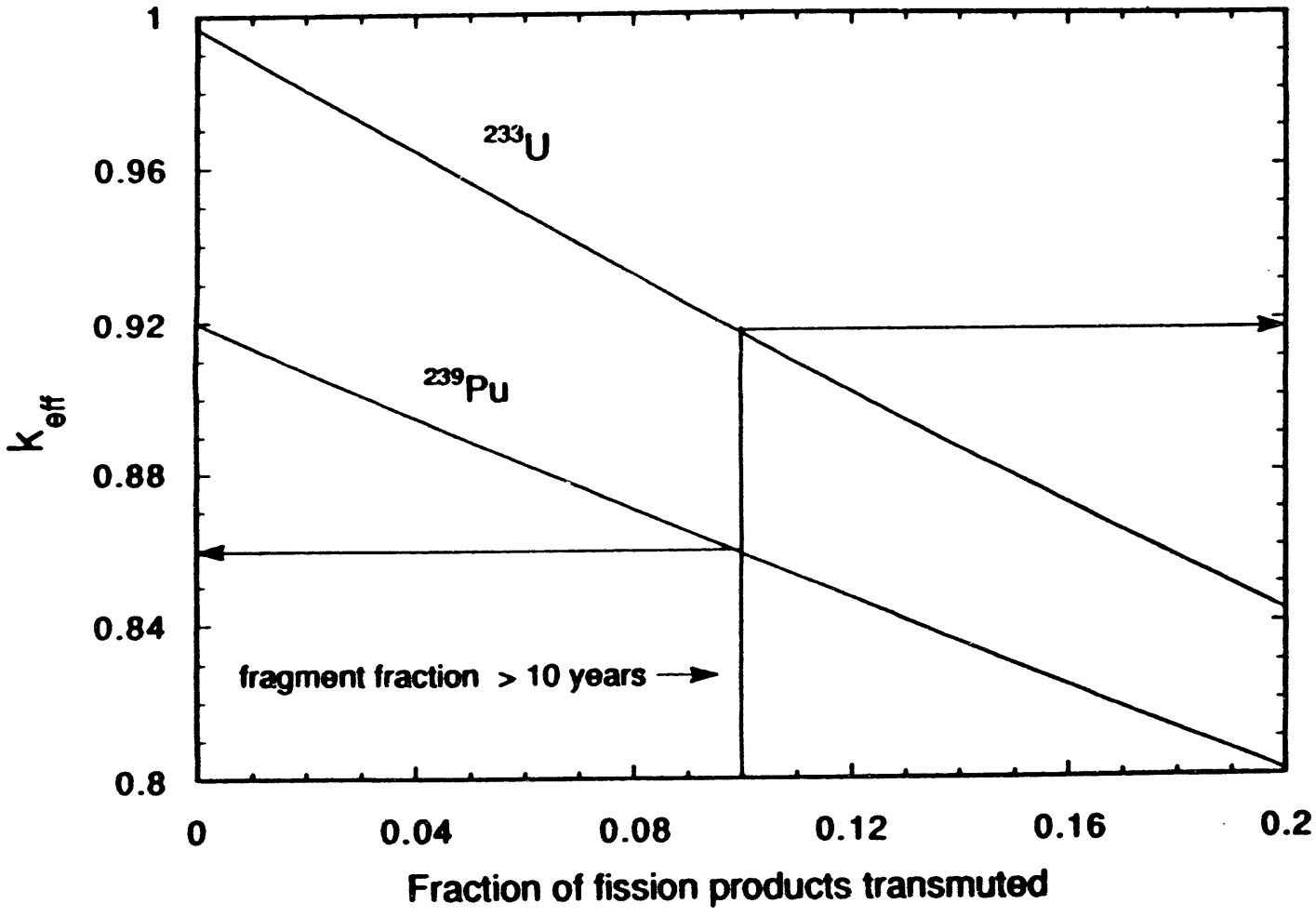
2. Energy Production from ^{232}Th Without Long-Term Fission-Product Storage



Dependence of k_{eff} on Fraction Transmuted

This figure shows the system multiplication constant k_{eff} for ^{238}U and ^{232}Th derived from the neutron economy equation. (An operating reactor has a k_{eff} of 1.0000.) If no fission products are burned, the ^{233}U system is almost critical; in fact the MSBR reactor at ORNL achieved a breeding ratio of about 1.02. On the other hand, for ^{238}U and ^{239}Pu the value for k_{eff} is only 0.92 for no fission product burning. These results reflect the well known facts that a thermal ^{233}U reactor will barely breed and that a ^{238}U and ^{239}Pu thermal breeder reactor isn't even close to practical. For burning all fission fragments with $T_{1/2} > 11$ years, one must burn about 10 % of the fission fragments and we see that this means a maximum k_{eff} of 0.86 for ^{239}Pu and 0.92 for ^{233}U. The corresponding multiplication factor (M) can be estimated using the equation $k_{eff} = M/(M-1)$. This figure shows that no thermal breeder reactor can burn its own waste. The accelerator therefore provides the vital function of supplementing the neutron production so that the system can breed at a high power level and burn its own waste. A new function is obtained by introducing the accelerator which is not possible with a reactor alone.

Dependence of k_{eff} on Fraction Transmuted



Equations for High Thermal Flux Burning of ^{237}Np

The dynamics of transmuting the higher actinide waste nuclide ^{237}Np is given for an infinite medium via these four equations. In particular, the third one describes the burning of a fission product nucleus which we might take to be ^{99}Tc. After a transient turn-on period the flux can be shown to settle down to a steady state for most values of the atomic densities N of the various nuclides involved. The equations can then be set equal to zero and solved for the flux and burn-up rates per cm^3 for the fission product R(fp) and for ^{237}Np, R(^{237}Np). The results are given in the next transparency.

Equations for High Thermal Flux Burning of ^{237}Np

$$dN(^{237}Np)/dt = -N(^{237}Np)\cdot\Phi\cdot\sigma_a(^{237}Np) + R(^{237}Np)$$

$$dN(^{238}Np)/dt = -\lambda(^{238}Np)\cdot N(^{238}Np) - N(^{238}Np)\cdot\Phi\cdot\sigma_a(^{238}Np) + N(^{237}Np)\cdot\Phi\cdot\sigma_a(^{237}Np)$$

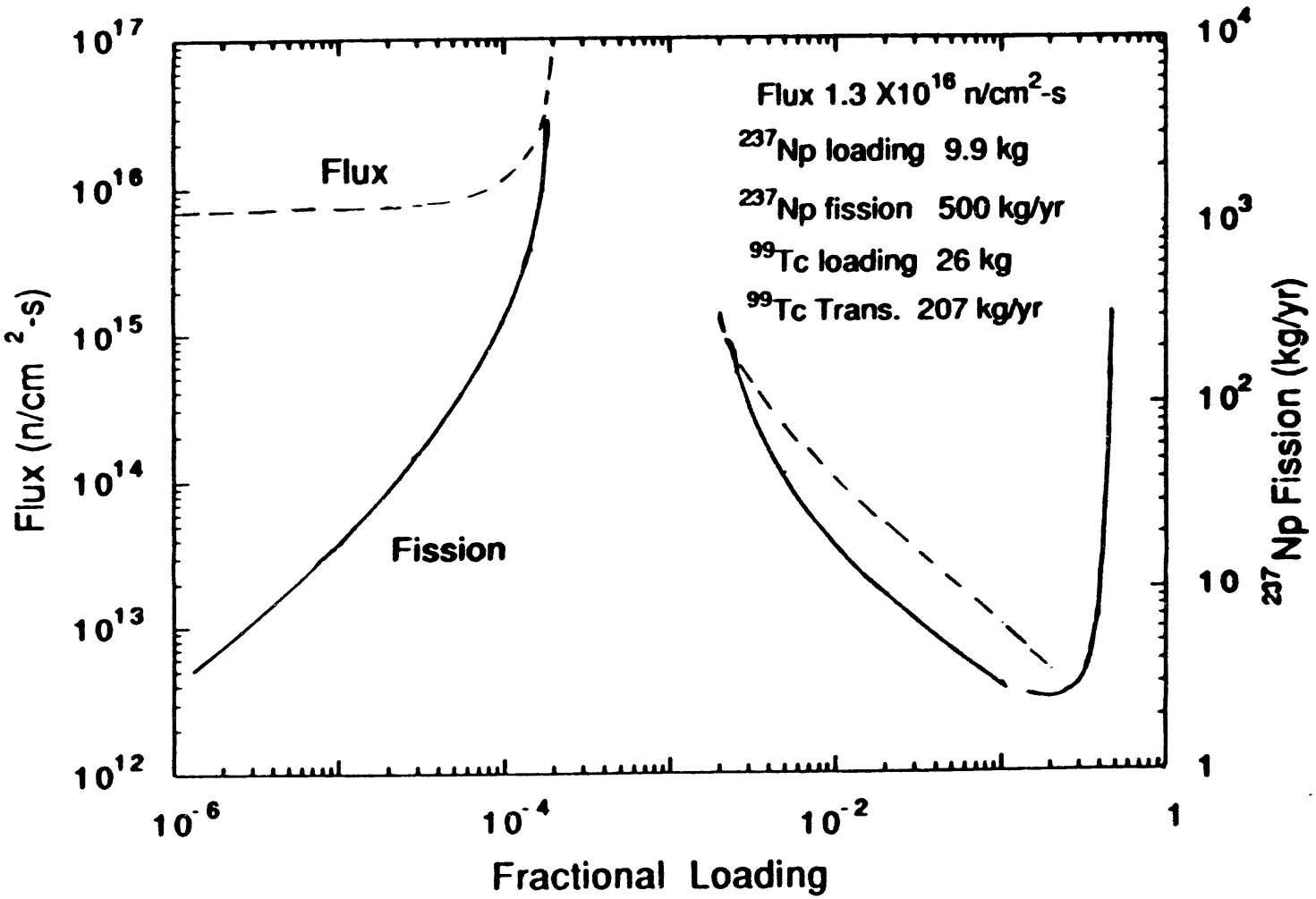
$$dN(fp)/dt = -N(fp)\cdot\Phi\cdot\sigma_a(fp) + R(fp)$$

$$1/v\cdot d\Phi/dt = N_0 - \Phi\cdot[\, N(^{237}Np)\cdot\sigma_a(^{237}Np) + N(fp)\cdot\sigma_a(fp) + N(^{238}Np)\cdot\sigma_a(^{238}Np)\,\{1 - \nu(^{238}Np)/(1+\alpha(^{238}Np))\}\,]$$

Inventory and Transmutation Rate

The flux is shown as the dashed curve referred to the left-hand ordinate as a function of the fractional loading, which would be NP metal for a fractional loading of one. The solid curve shows the rate of Np destruction by fission referred to the right-hand ordinate. The flux at the smallest load is about 7 X 10^{15} n/cm^2-s as provided by the accelerator operating at 50 ma and driving a system of a few m3 volume. As the fractional loading is increased starting from 10^{-6} the Np burning rate increases from the value of about 3 kg/yr. When a fractional loading of 10^{-4} is approached, the neurons from ^{238}NP fission begin to enhance the flux (and the transmutation rate). At a loading above 2 X 10^{-4} the system approaches a supercritical regime which exists for this idealized problem formulation. (The presence of absorbing materials in the problem would limit the magnitude of this increase.) Consider now a fractional loading of 0.1. In this condition there is so much capturing material present that the accelerator can maintain a flux of only about 10^{13} n/cm^2s. Any ^{238}Np transmuted from ^{237}Np decays in this low flux before it can be fissioned. Therefore the Np acts as a poison and the neutron flux is reduced rather than enhanced. As the fractional loading is decreased from 0.1, the depression of the flux is less and near 2 X 10^{-3} the system passes into the supercritical range. Note that as the fractional loading is increased from 0.1 the system starts to become fast critical. This is the regime in which fast reactors and all earlier-considered accelerator-driven assemblies for waste burning operate. We suggest an operating fractional loading near 10^{-4} where the flux is about 1 X 10^{16} n/cm^2-s and where the Np transmutation rate is about 500 kg/yr with an inventory of about 10 kg. The same Np transmutation rate for the fast reactor is achieved with an inventory larger by more than a factor of 1000! Substantial transmutation rates at low inventory for the fission product are also given in the figure for a modest loading.

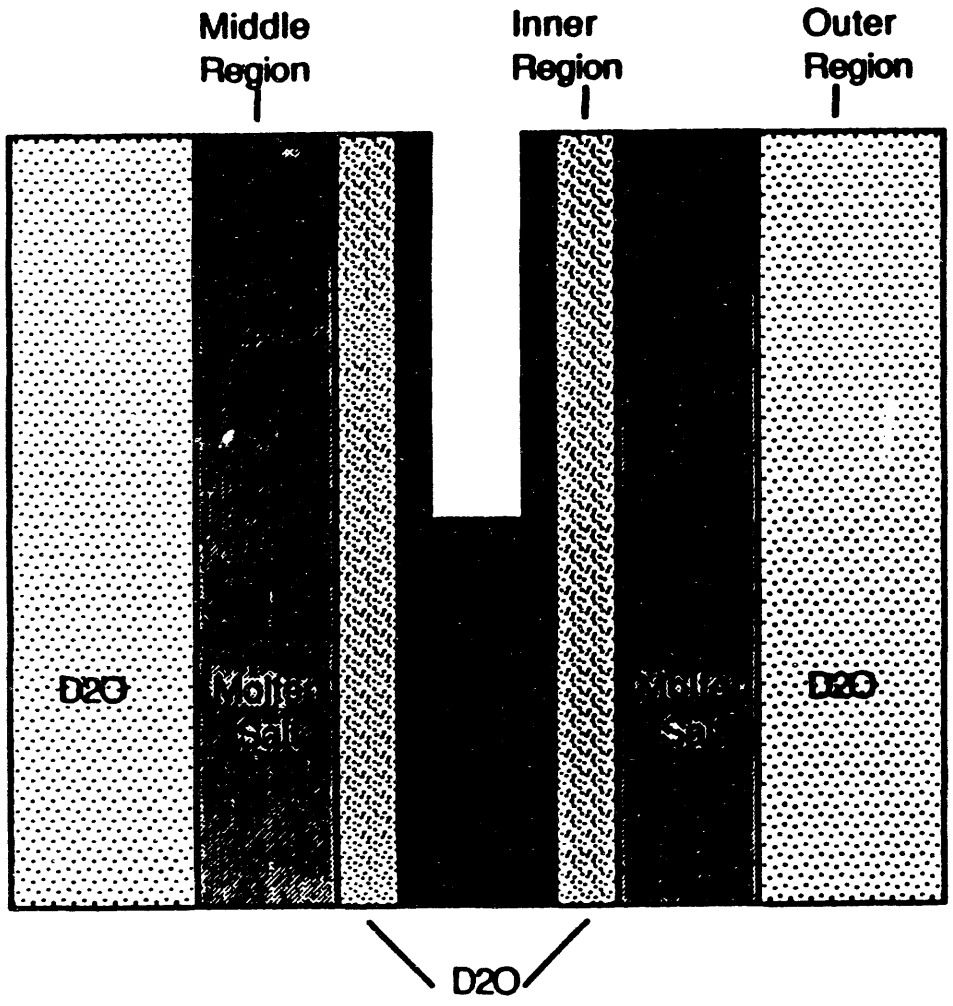
Inventory and Transmutation Rate



Target-blanket Geometry

The blanket for the Th energy producer is divided into three parts. The Th is converted to ^{233}Pa in the outer region where the flux is lowest (and for where high flux is not required) and is removed immediately so as to avoid any fission of ^{233}U. The ^{233}Pa decays outside of the system to ^{233}U and this is fed back into the molten salt as $^{233}UF_4$ along with any other actinides which are to be fissioned. Fission power is generated here and the fission product removed continuously. The inner region where the flux is highest is used for burning the 10 % of the fission products with half-lives greater than 11 years.

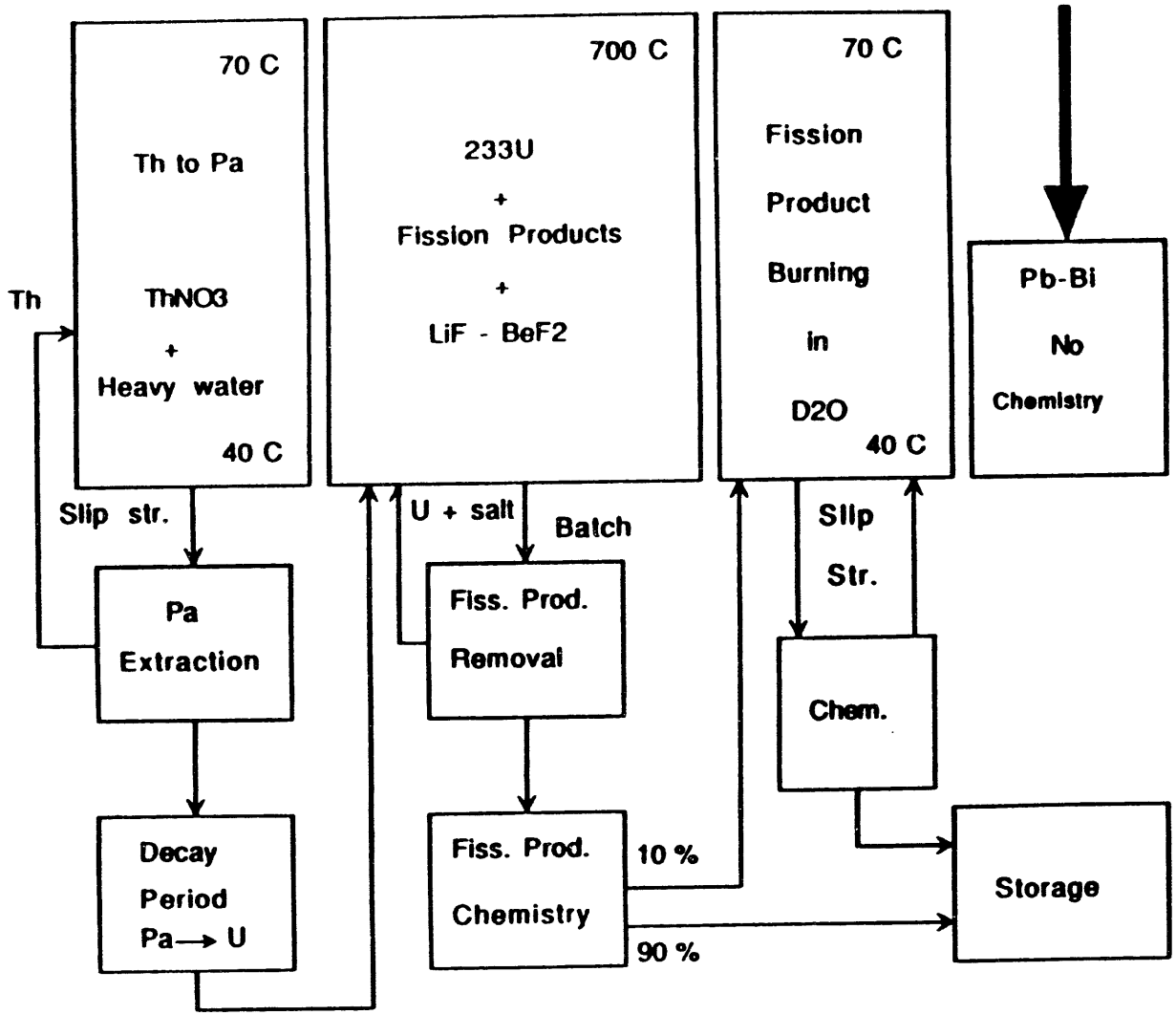
Target-blanket Geometry



Blanket Chemistry

This transparency shows the progression of original Th material through the conversion to Pa followed by decay to U which is burned in the molten salt. The fission products are separated from the molten salt and the stable or short-lived products are sent to storage. The 10 % of the fission products which must be burned (6 nuclides) are returned one by one to the inner region of highest flux for transmutation. Upon transmutation the product nuclei are also sent to storage. Prompt removal of the Pa from the outer blanket via a continuous chemistry on a slip stream is necessary to assure that there is no fission of the ^{233}U in the outer region. Prompt removal of the transmuted fission product in the inner region is necessary in order to prevent the waste of neutrons in capture on already transmuted nuclei.

Blanket Chemistry



PWR Fission Products with $T_{1/2}$ > 11 Years

This transparency includes for reference all of the nuclides from a commercial power reactor with half lives greater than 11 years. For the accelerator-driven Th burner there will be no ^{151}Sm, because of its high cross section or ^{135}Cs because of the high cross section of its precursor ^{135}Xe (over two million barns). We propose not to burn the 93 Zr because of the low energy (30 keV) of its radiation or the ^{107}Pd because of its low energy radiation (30 keV). Both metals also have very stable metallic or alloy states. The latter two nuclides also could be burned if necessary at some reduction in overall power efficiency. This leaves only six fission products which need to be transmuted.

PWR Fission products # with $T_{1/2}$ > 25 Years

Nuclide	Kg/yr	Half life (yrs)	Cross Section (barns)	Atoms/yr ($X 10^{25}$)
^{79}Se	0.19	$6.5 X 10^4$	? (10)	0.13
^{90}Sr	13.5	29	0.9	8.97
^{93}Zr*	23.3	$1.5 X 10^6$	2.5	15
^{99}Tc	25.1	$2.1 X 10^5$	20	15.2
^{107}Pd*	7.4	$6.5 X 10^6$	1.8	4.1
^{126}Sn	0.91	$1 X 10^5$	0.14	0.46
^{129}I	5.87	$1.6 X 10^7$	27	2.73
^{135}Cs+	9.47	$3.0 X 10^6$	8.7	4.22
^{137}Cs	30.9	30	0.25	13.5
^{151}Sm+	0.38	90	15200	0.16
			Total*	$41.0 X 10^{25}$

These amounts are annual production for a PWR running at 3 GW thermal with fuel burned to 33,000 megawatt-days per ton requiring the removal of 33 MTU spent fuel per year.

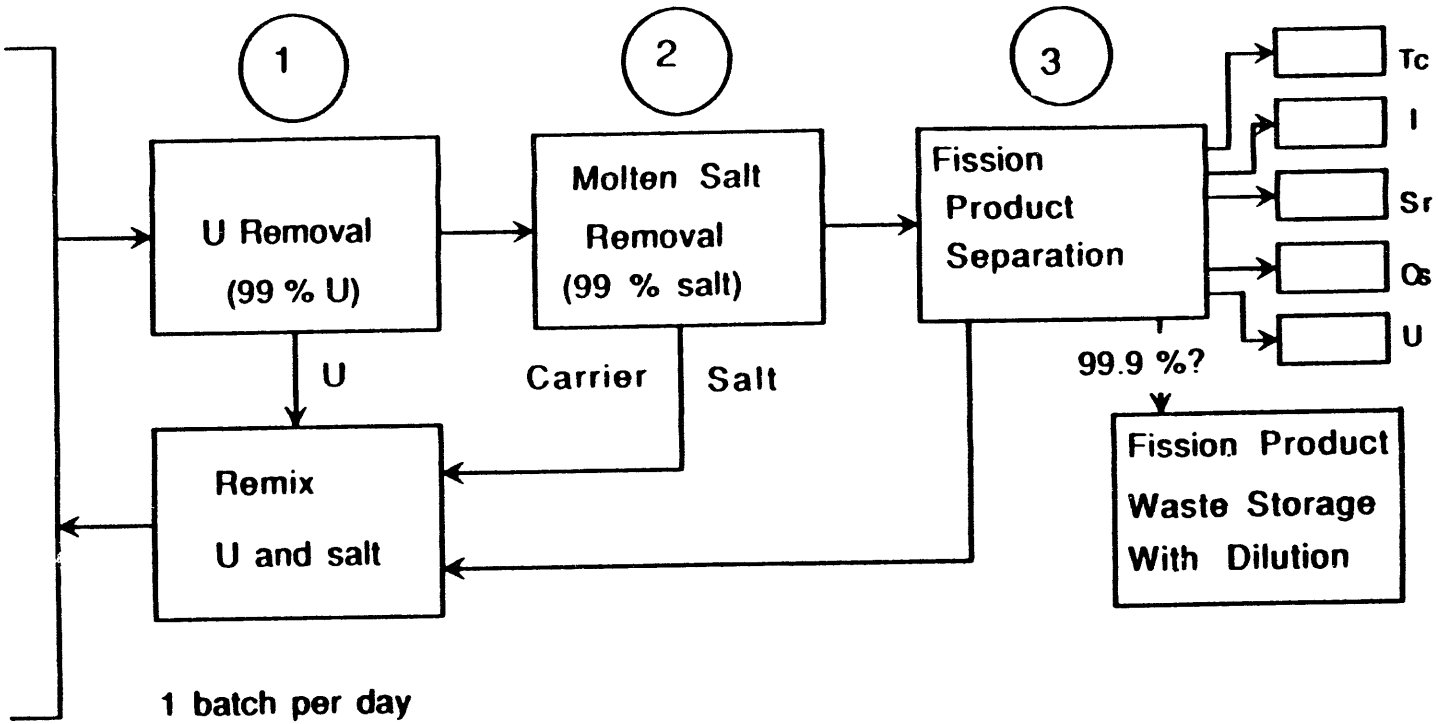
* These nuclides have very weak β- and γ radiations and are not transmuted but diluted as described in the text. They are omitted from the sum.

+ Burned internally when the flux is very large. They are omitted from the sum.

Molten Salt Chemistry

We propose a batch process in which 10 % of the salt is removed each day. For a 3000 MWT system this would correspond to a daily production of about 3 Kg of fission product. The purpose of this transparency is to identify three distinct steps in the molten salt chemistry. The first is the removal of UF_4 from the molten salt. This separation should be effective at the level of about 99 % of the uranium removed if possible and the material returned immediately to the blanket. Contamination of the separated uranium with a high level of fission product or carrier salt is not a problem. The second step is the separation of the carrier molten salt from the fission product. This chemistry process should remove nearly all of the salt, although contamination with the fission product is not a serious issue. These two steps yield the fission product mixture. From this, one must efficiently separate the six nuclides to be transmuted. This third step is by far the most difficult, but since it contains only fission products a variety of chemical processes can be used. This chemistry can proceed off line at a leisurely rate.

Molten Salt Chemistry



1 batch per day
10% of salt per batch
1 kg of U removed
3 kg of fission products removed
3000 MWT

Energy Production from 232Th with Waste Transmutation and Isotope Separation

Chemical separations of fission products to be transmuted will produce not only the isotope of interest but all of the stable isotopes of a particular element as well. We want to avoid wasting neutrons in the transmutation blanket by having them absorbed by stable nuclei, possibly leading to unstable products. This transparency addresses the role of isotope separation on the transmutation of the fission product. Using the neutron economy equation in combination with the series of differential equations for transmutation from one species to another, one finds that with no isotopic separation the overall power production efficiency is only 17%. Isotopic separation of the Cs alone almost doubles the efficiency to 30%. Isotopic separation for all of the other species has little additional impact since the efficiency only increases to 32%. If Cs is burned with isotopic separation after 30 years of decay, the efficiency increases a little more. We conclude that isotopic separation of Cs is vital but that isotopic separation of the other species is not worthwhile.

Energy Production from ^{232}Th With Waste Transmutation and Isotope Separation

Isotope Separation (IS) Condition	Overall Eff. (%)	Power to Acc. (%)	I (ma)
Burning All			
No IS	17	61	75
IS Cs only	30	31	38
IS all	32	28	35
IS Cs only after 30-year decay	34	23	28

Effect of Isotope Separation on Transmutation of ^{233}U Fission Products

The second column of this transparency shows the amounts in atoms per year produced in a 3000-MWt system of an isotope along with the total production of that element. The estimated number of neutrons required to transmute each of the nuclides assuming no isotopic separation is given in the third column. Note that the Cs offers the most serious problem. The ^{79}Se and the ^{126}Sn are not important since the amounts are relatively small. One sees that the situation for Cs is much improved after isotope separation but that the overall impact of isotope separation for the other nuclides is small.

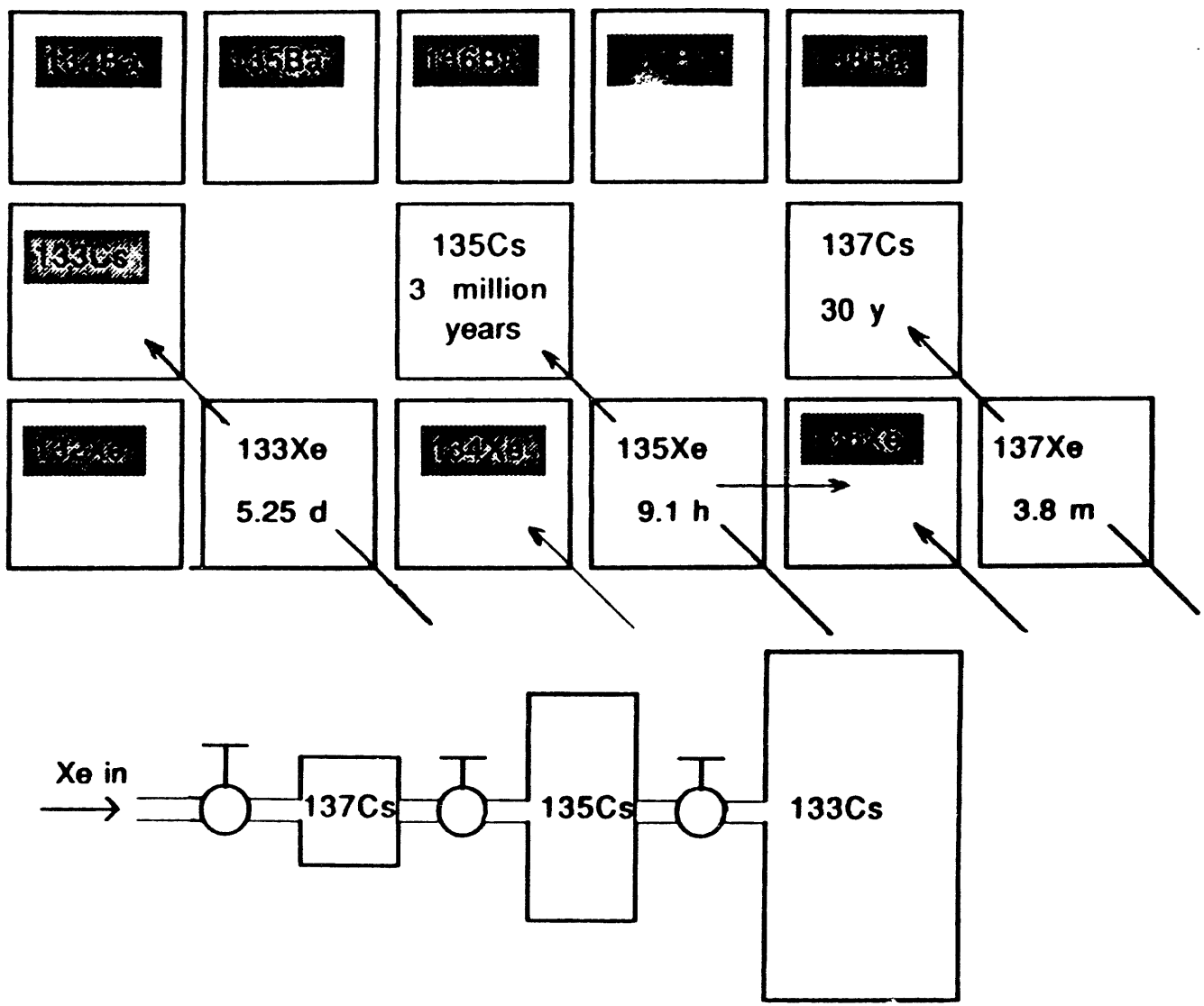
Effect of Isotope Separation (IS) on Transmutation of ^{233}U Fission Products

Isotope	Production (A/yr X $^{23}10$)	Neutrons Without IS	Neutrons With IS
Se	245	1.48	1.15
^{79}Se	36.6		
Sr	2988	1.05	1.00
^{90}Sr	1660		
Tc	1186	1.00	1.00
^{99}Tc	1186		
Sn	83	1.93	1.02
^{126}Sn	61		
I	478	1.08	1.00
^{129}I	359		
Cs	4605	4.1	1.00
^{135}Cs	1520		1.48
^{137}Cs	1626		

Cs Isotopic Separation by Xe Decay

The continuous flow system allows one to take advantage of a natural isotope separation process based on the half lives of the Xe precursors. The stable nuclides in this region are shown with the shaded labels. Only a tiny fraction of the Cs isotopes are produced directly in fission. The great bulk of them comes from beta decay along the mass chain. The mass chains for 132, 134, and 136 all end on stable Xe isotopes. However, odd mass chains all pass through unstable Xe isotopes which (however) have half-lives that differ by more than a factor of ten. Therefore the Xe, which is emitted promptly from the salt, could be sent into a system of decay volumes separated by appropriate flow control. Since the half-lives differ so greatly, excellent separation can be obtained through this decay separation process. It is important to note in addition that ^{135}Xe is the notorious isotope giving rise to xenon poisoning in thermal reactors through its 2,500,000 barn thermal capture cross section. The ^{135}Xe is therefore transmuted in the high flux in a few seconds to ^{136}Xe. In our high flux system there will be no ^{135}Cs product except for that associated with interruptions of operation of the system. Note that for reactors using solid fuels as fuel assemblies, this decay separation scheme cannot be implemented.

Cs Isotopic Separation by Xe Decay



Irradiation Volume for Fission Product Burning as Aqueous Salt

This transparency addresses the question of whether there is adequate high flux volume in the system to accomplish the required fission product transmutations. Lower cross section isotopes require a longer irradiation period. The system therefore must offer enough flux-volume product to allow transmutation. We have assumed the use of the soluble compounds given in column 1 with the solubility given in column 3. The volume occupied by the annual production in a 3000-MWT system of a fission product is given in column 5. The sixth column gives the time in years required for transmutation so that the fraction 1/e of the isotope remains untransmuted. The last column shows the volume required for transmutation to the 1/e level derived as the product of columns 5 and 6. We see that a total of 590 liters is required of flux at the 10^{16} n/cm^2-s level. If the Cs and Sr are allowed to decay for 30 years before transmutation, the required volume decreases to 402 liters. The high flux volume in the blanket is about 1000 liters.

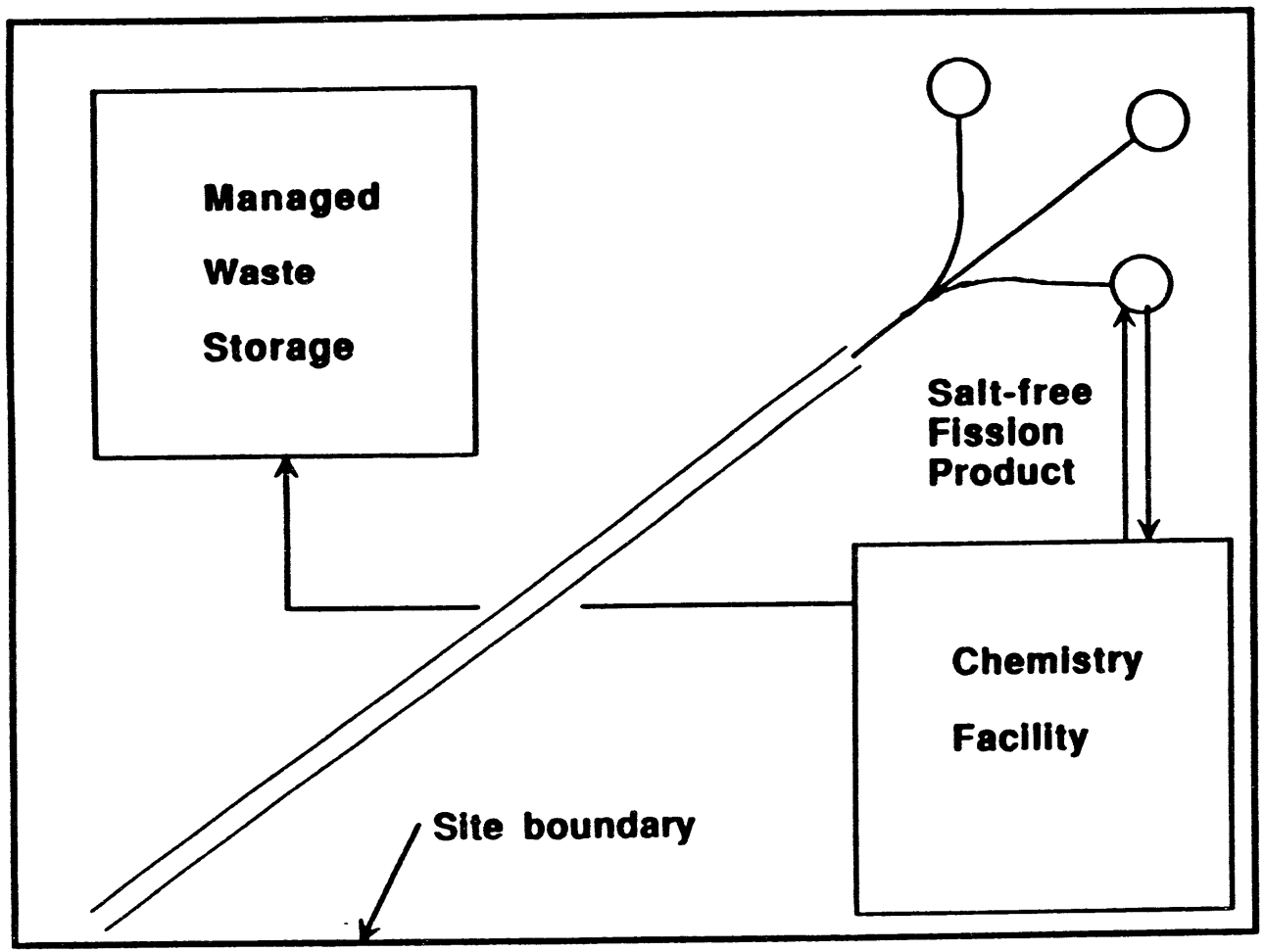
Irradiation Volume for Fission Product Burning as Aqueous Salt

Compound	Mass (kg/yr)	Sol. (g/liter)	Mol.Wt.	Volume (liters/yr)	Sat. Factor (yrs)	Total (liters)
$^{90}Sr(NO_3)_2$	34.2	1000	214	81	3.2	259
$^{107}PdSO_4$	3.2	?(5000)	203	1.3	1.8	2.3
$^{79}SeO_2$	3.4	825	111	59	0.32	19.0
$^{126}SnSO_4$	1.7	330	222	9	21	189
$^{99}Tc(NO_3)_4$	19.5	?(5000)	347	13.7	0.16	2.2
$^{7}Li^{129}I$	10.0	4330	136	2.4	0.12	0.3
^{137}CsF	37.0	3670	156	11.5	9.9	114
^{135}CsF	33.0	3670	154	10.3	0.37	3.8
$^{151}Sm(NO_3)$	8.3	?(5000)	445	4.9	0.00015	0.0
					Total	**590**
						(402 30 y)

Energy Park Layout

The most economic implementation of this concept would include a single accelerator operating at the highest technically practical level driving, by beam multiplexing, more than one target blanket system. A single facility for fission product chemistry would serve all target-blanket systems. It might be necessary for each target-blanket system to have its own U and carrier salt removal systems. The short-lived or stable fission product is sent to the on-site managed storage facility where it is diluted if necessary to the U ore activity level. As low level waste it might be stored here, moved to a repository, or returned to the mine.

Energy Park Layout



Comparison of Reactor and Accelerator-driven Systems

Issue	Reactor	Acc. Driven Sys.
Criticality	Yes	No
LOCA Concern	Alternate cooling	Fluidsdrainage
Afterheat	1	1/10
Control Rods	Yes	No
Fission products burned	No	Yes
Geologic repository	Yes	No
Radioactivity leakage	Low	Low
Fluid Circulation loops		4
Fuel fabrication	Yes	No
Internal structure	Complex	Simpler
Reloading interruption	Yes	No
Materials damage	Satisfactory	Greater
Radioactive Inventory	High	Low

Study Issues

Target-Blanket Neutronic Performance
- Accurate neutron proton measurement
- Thermal and higher cross sections for spallation products
- Resonance cross sections for Cs, Sr and others
- Thermal and higher energy cross sections for structural mat.
- Spallation product distribution measurements
- Detailed neuuronic performance calculations

Chemistry
- 233Pa from 232 Th
- 233 U and molten salt from fission products
- Extraction of Sr, Cs, Pd, Zr, I, Tc, Se, Sn from fiss. prod. mix
- Transmuted fission products from untransmuted
- Reflux at Pb-Bi surfact
- Chemistry among spallation products

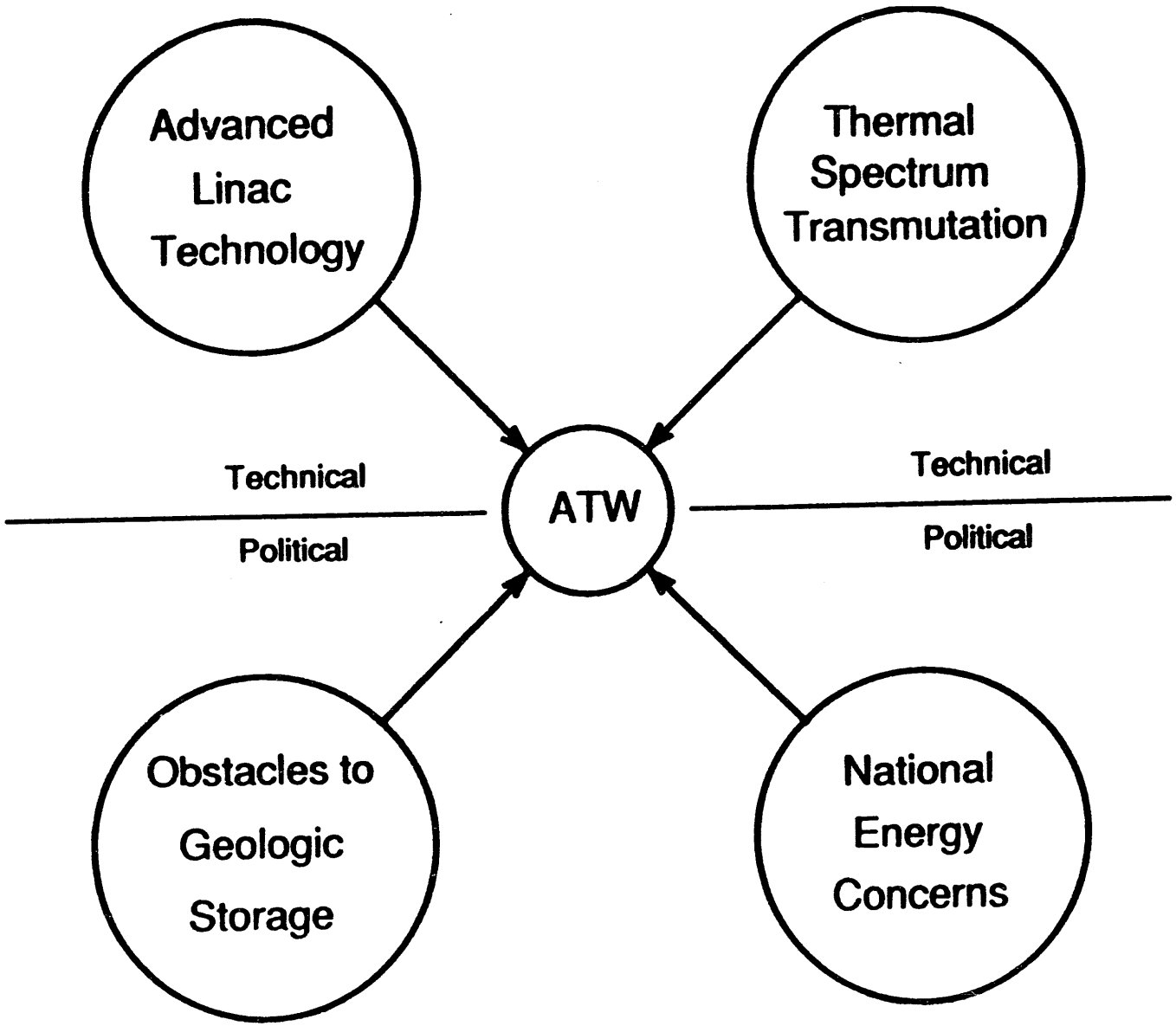
Materials selection
- Pb-Bi containment
- Molten salt containment
- Heavy water containment (zircalloy)

Accelerator Research
- Improved klystron
- Injector demonstration
- R-f modulation

Safety
- Target-blanket, chemistry accelerator

Detailed demo design
- Accelerator, target-blanket, chemistry

Project Timeliness



Summary

Accelerator increases the number of neutrons per fission

Neutron economy allows energy production from Th and U with waste burning

High transmuuation rates with low inventory

Three-step molten salt chemistry

Cs isotope separation by natural decay

No other isotope separation required

Low-level (U-ore) waste only

Could Lead To

Inexhaustible energy supply

Nuclear runaway avoidance

Low Level waste only

Thorough Study Required

ADVANCED CHEMISTRY

Molten Salt Chemistry for the Advanced ATW Concept

Scott Kinkead
Isotope and Nuclear Chemistry Division

Stable & Short-Lived Fission Product Disposal

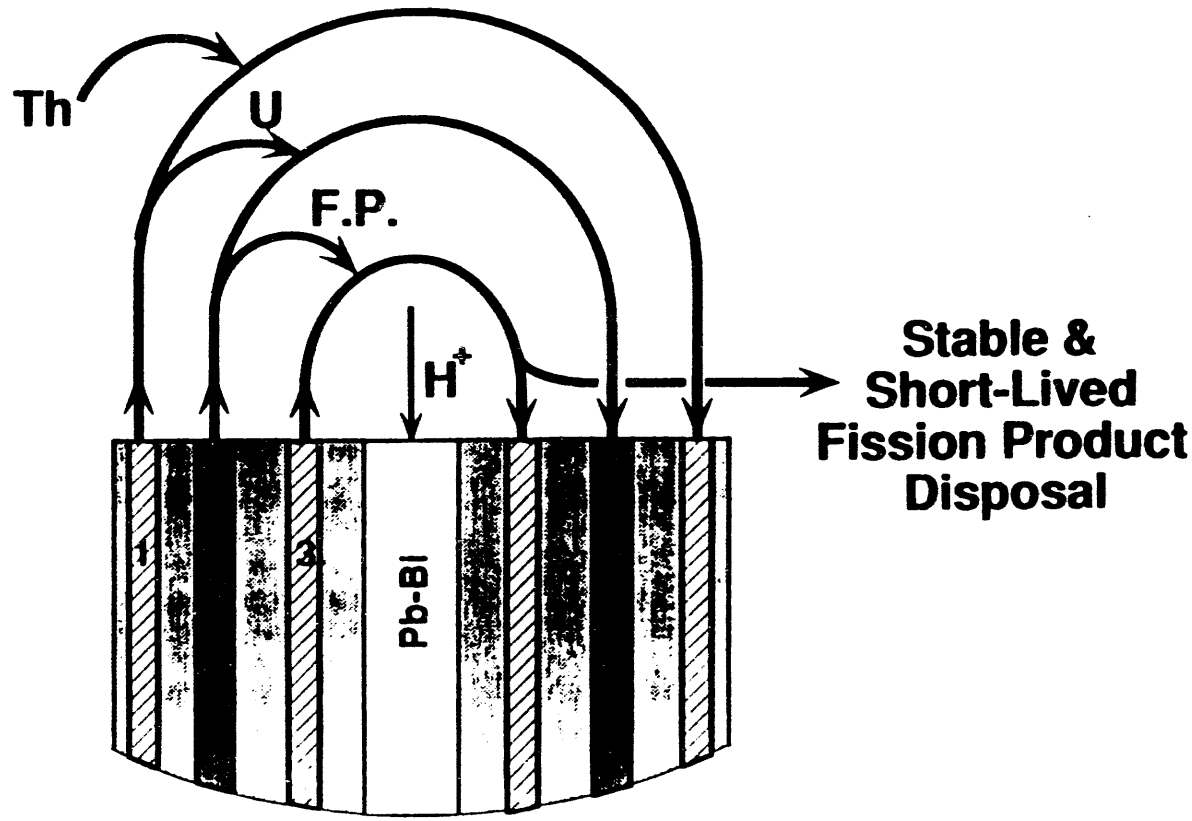
Modern chemical processing is essential for energy production without a high level waste stream. The system has three interconnected flow loops, each with optimized feed and processing systems. The first loop is aqueous based and is dedicated to the conversion of ^{232}Th to ^{233}U. The second loop is based upon a molten mixture of ^{7}LiF and BeF_2. It will accommodate the fissioning ^{233}U and support the fission product stream. The peak temperature of the loop is approximately 775° C to provide high electrical efficiency. The third loop is also aqueous based and is dedicated to the transmutation of long-lived fission products received from the molten salt loop. The various fission products will be transmuted in sequential campaigns. This procedure will simplify the chemical processing plant required to remove stable products from that loop.

Stable & Short-Lived Product Disposal

The Energy Without a High Level Waste Stream Concept has

Three Separate Flow Loops: 1. ^{233}U breeding;

2. ^{233}U Fission; and 3. Fission Product Transmutation



Objective

The objective of our advanced concept is energy production with a minimal high level waste stream. To accomplish this, the fission loop must generate sufficient energy to run the high energy proton accelerator as well as producing electrical energy for the power grid. Using a high neutron flux generated from spallation of lead-bismuth and thermalized in heavy water, large scale nuclear energy can be produced with far less than critical inventories of fissile materials.

Approach

Conventional nuclear reactors, using pressurized water, boiling water, or gas cooling operate at thermal efficiencies in the range of 20 to 40%, largely dictated by thermodynamic considerations of the cooling process. Because of their thermal properties and liquid range, molten salts are capable of operating at thermal efficiencies of 44%.

Concept

New designs in components permit construction of accelerators with unprecedented levels of energy and current; all the while maintaining high duty factors necessary for operation of a power production station.

Objective

To develop a process which will permit GW power generation from sub-critical inventories of actinides using a large, accelerator-generated thermal neutron flux.

Approach

Molten salt-based processes, because of their high operating temperatures, possess higher energy production efficiencies than many water- and gas-cooled reactors (44 vs. 20-40%).

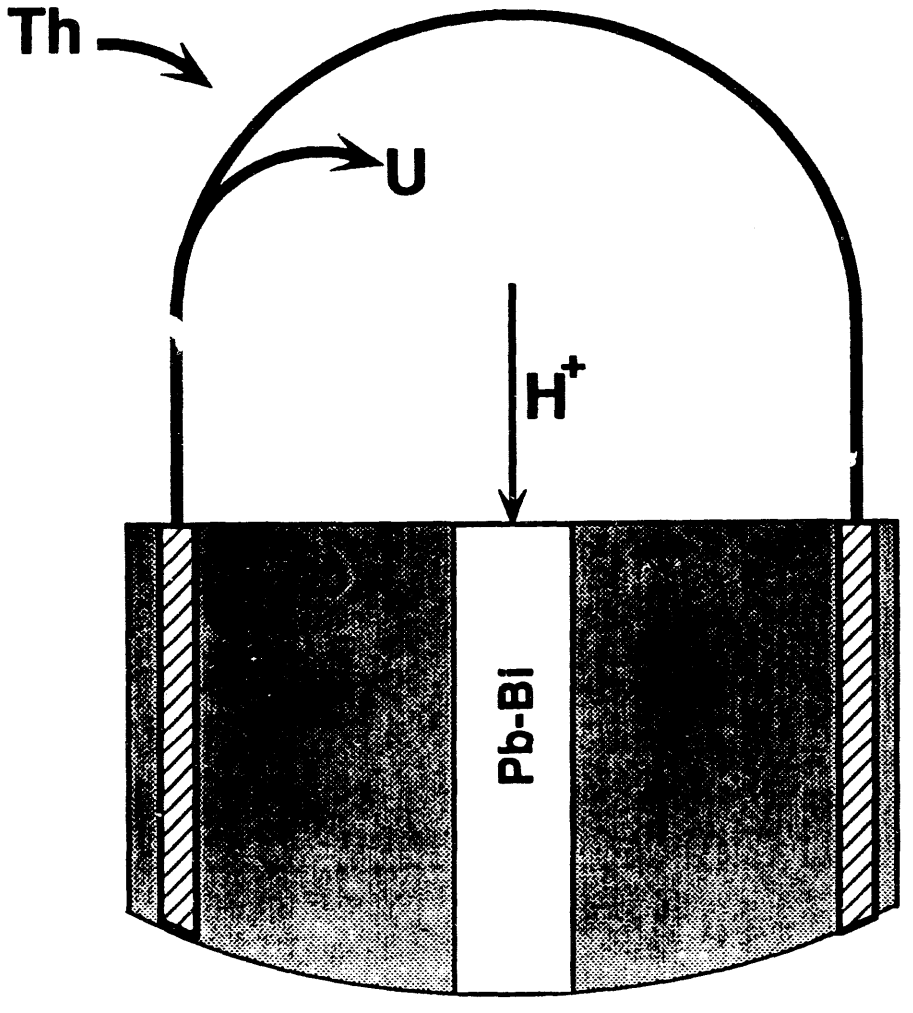
Concept

New spallation target design allows neutron flux of up to $10^{16} n/cm^2/sec$. Large thermal and epithermal neutron flux permits power generation from dilute actinide blankets.

Loop 1: ^{232}Th - ^{233}U Thermal Neutron Breeder

To simplify the complicated nature of the chemistry which is required for the accelerator production of energy concept, we will address the features, requirements, and technical issues for each loop separately. To take advantage of the large thermal neutron flux produced from the accelerator, the first loop will convert fertile ^{232}Th into ^{233}U fuel.

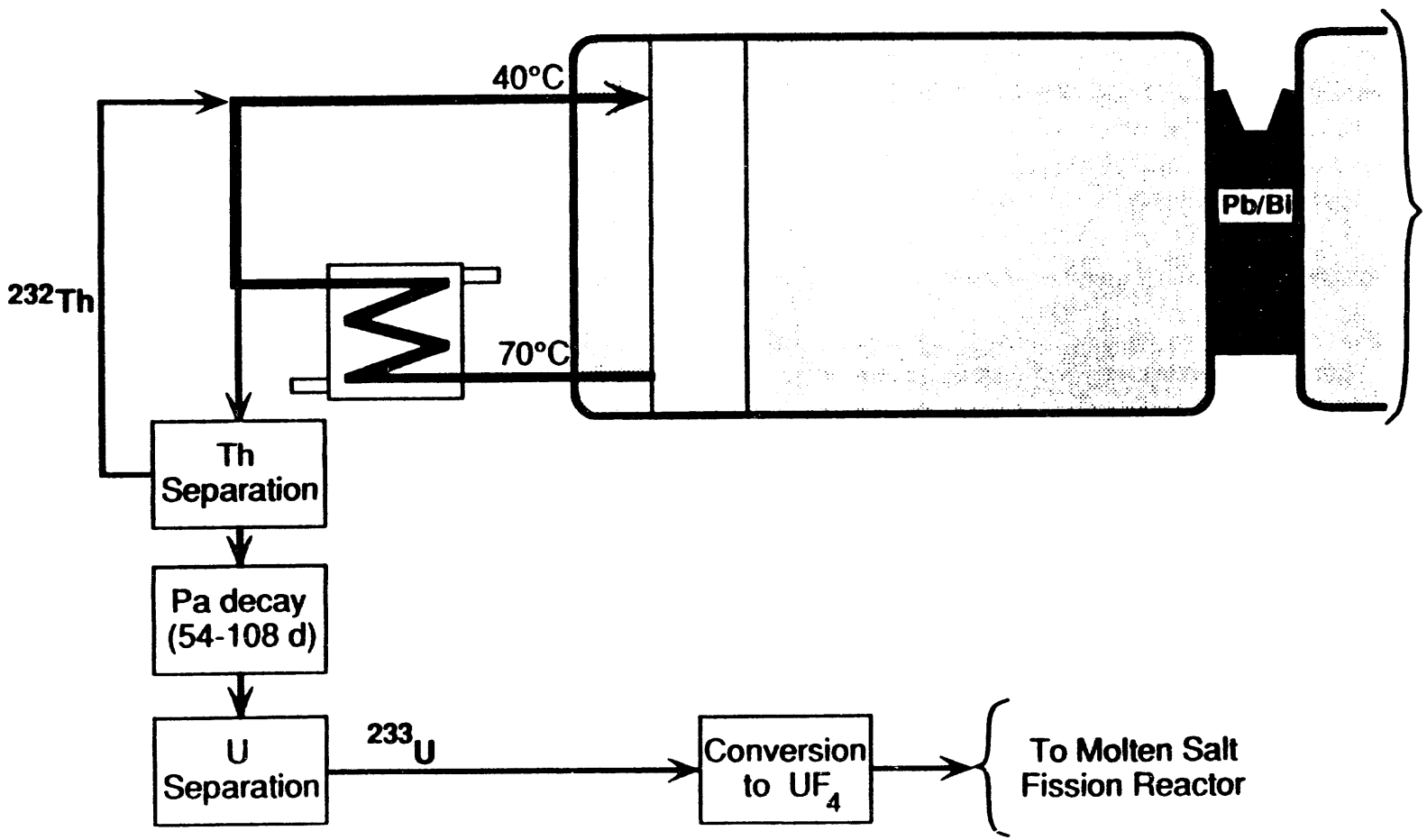
Loop 1: ^{232}Th - ^{233}U Thermal Neutron Breeder



ATW $^{232}Th/^{233}U$ Transmuter

In this schematic, we show a simplified flowsheet for processing the mixture of ^{232}Th, ^{233}Pa, and ^{233}U. Because of the large thermal neutron flux in the region of the Th/U breeder system (10^{15} n/cm^2/s), production of the intermediate ^{233}Pa will be at a higher rate than found in, for example, the two-region thermal breeder reactor (TBR) developed at ORNL. Even with a more dilute blanket than that found in the TBR (1-200 g/l vs. 1 kg/l), production of the ^{233}Pa intermediate should be at least equivalent to the TBR rate of 350 g/day of ^{233}Pa. The low-grade heat produced in the transmutation process is removed through a heat exchanger and discarded. Chemical processing of the mixed Th/Pa/U product is based on the vast differences in the aqueous chemistry of protactinium, thorium and uranium. After a suitable period to allow the bulk of the ^{233}Pa ($t_{1/2}$ = 27 d) to decay to fissile ^{233}U, the uranium is separated and converted to the tetrafluoride for fuel in the molten salt fission loop.

ATW $^{232}Th/^{233}U$ Transmuter



Features of the $^{232}Th/^{233}U$ Thermal Breeder Loop

Selection of the chemical species for the Th/U breeder is predicated on achieving a facile, high level of separation of Th from the Pa and U products. An important consideration for such a high flux system is using counterions which contain elements transparent to thermal neutrons. For example, the use of $^{15}NO_3^-$ provides a suitably transparent counterion whereas $^{14}NO_3^-$ has a high cross-section for (n,α) reaction. The addition of fluoride ion is mandated by the ready hydrolytic polymerization and precipitation of protactinium. As in the Thorex process Pa and U can be separated from Th, after complexation of the fluoride ion with aluminum, with conventional techniques such as extraction with TBP into organic solvents, or ion exchange.

In the ORNL molten salt reactor experience, the decay period for ^{233}Pa is 270 days, to allow >99.9% of the available ^{233}U to be formed. Because of the economics of storing available fissile material, it is desirable to process the Pa more frequently to remove the U as rapidly as possible.

One possible process for separating uranium from ^{233}Pa is the volatilization of UF_6; due to its short half life (as well as the expected lower vapor pressure of PaF_5), $^{233}PaF_5$ should be completely decomposed by radiolysis (c.f. $^{238}PuF_6$) and thus not be volatilized.

Features of the $^{232}Th/^{233}U$ Thermal Breeder Loop

- Aqueous (deuterated) thorium [e. g. Th(NO3)4] blanket. Addition of DF to prevent Pa fraction/hydrolysis

- Th species chosen for rapid, facile separation of ^{233}Pa from ^{232}Th ion exchange, solvent extraction (Thorex), etc.

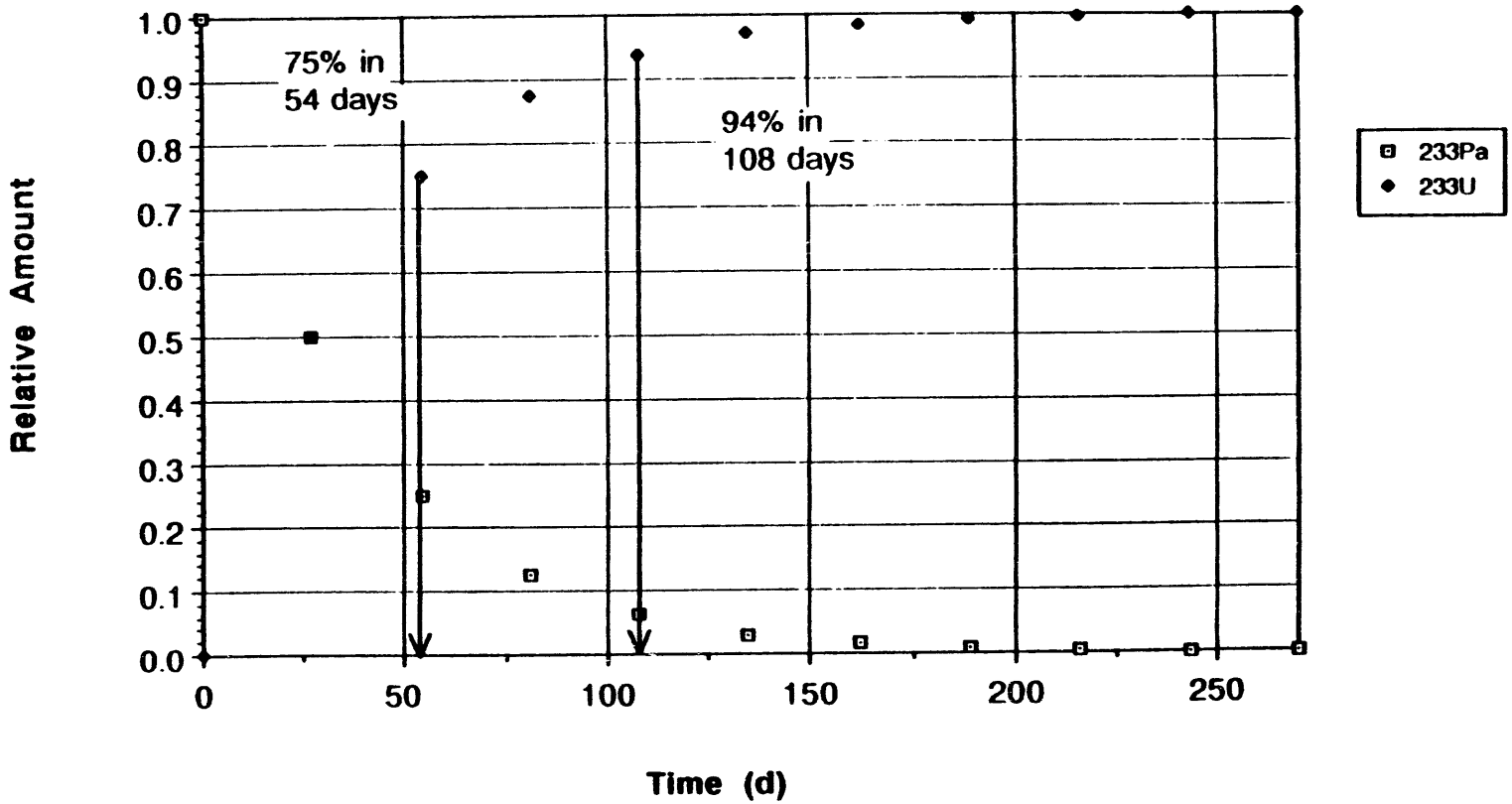
- Cooling period to allow a large fraction of ^{233}Pa ($t_{1/2}$ = 27 d) to decay to ^{233}U

- Uranium separation (from ^{233}Pa) and conversion to UF_4 (e.g., fluoride volatility process)

^{233}Pa Decay/^{233}U Growth

Although 99.9% of ^{233}Pa decays to ^{233}U in 270 days for a fixed-size batch of protactinium, 94% of the available uranium is formed in only 108 days. Thus, waiting an additional 162 days provides only 5.9% additional ^{233}U. If U/Pa separations are fairly complete so that significant quantities of Pa are not lost through transmutation/fissioning in the molten salt cycle, and radiolytic concerns are adequately addressed, shorter decay cycles are economically viable.

^{233}Pa Decay/^{233}U Growth



$^{232}Th/^{233}U$ Breeder Technical Issues

Many of the issues raised in the Th-U breeder loop have been addressed to varying degrees in earlier programs such as the ORNL TBR program. A technical challenge unique to the accelerator-based energy production concept is the exposure of materials to high neutron flux levels. In an aqueous process operating at low pressures and temperatures, low nickel stainless steels meet the requirement of thermal neutron transparency, while being readily available and possessing adequate corrosion resistance.

The high specific activity of ^{233}Pa (256 KeV β^-) induces radiolysis in solvents and ion exchange resins unless sufficient decay times are built in. In the TBR program, a 60 day decay interval prior to processing reduced solvent exposure in the blanket material (~400g/day ^{233}Pa) to approximately 0.25 watt-hr/liter, an acceptable level for processing. A related issue is the radiologically mandated remote processing and handling of the $^{233}Pa/U$ material. While this is a concern, there is abundant experience at the national laboratories for remote handling of radionuclides.

Along similar lines, large amounts of uranium fluorides, particularly the higher activity ^{233}U isotope, introduce concerns due to increased neutron dosages due to the well-known (α,n) reaction of fluorine. This reaction is sufficient to increase the dosage by ten-fold in Pu processing over oxides or chlorides, and must be dealt with using adequate shielding or remote operations.

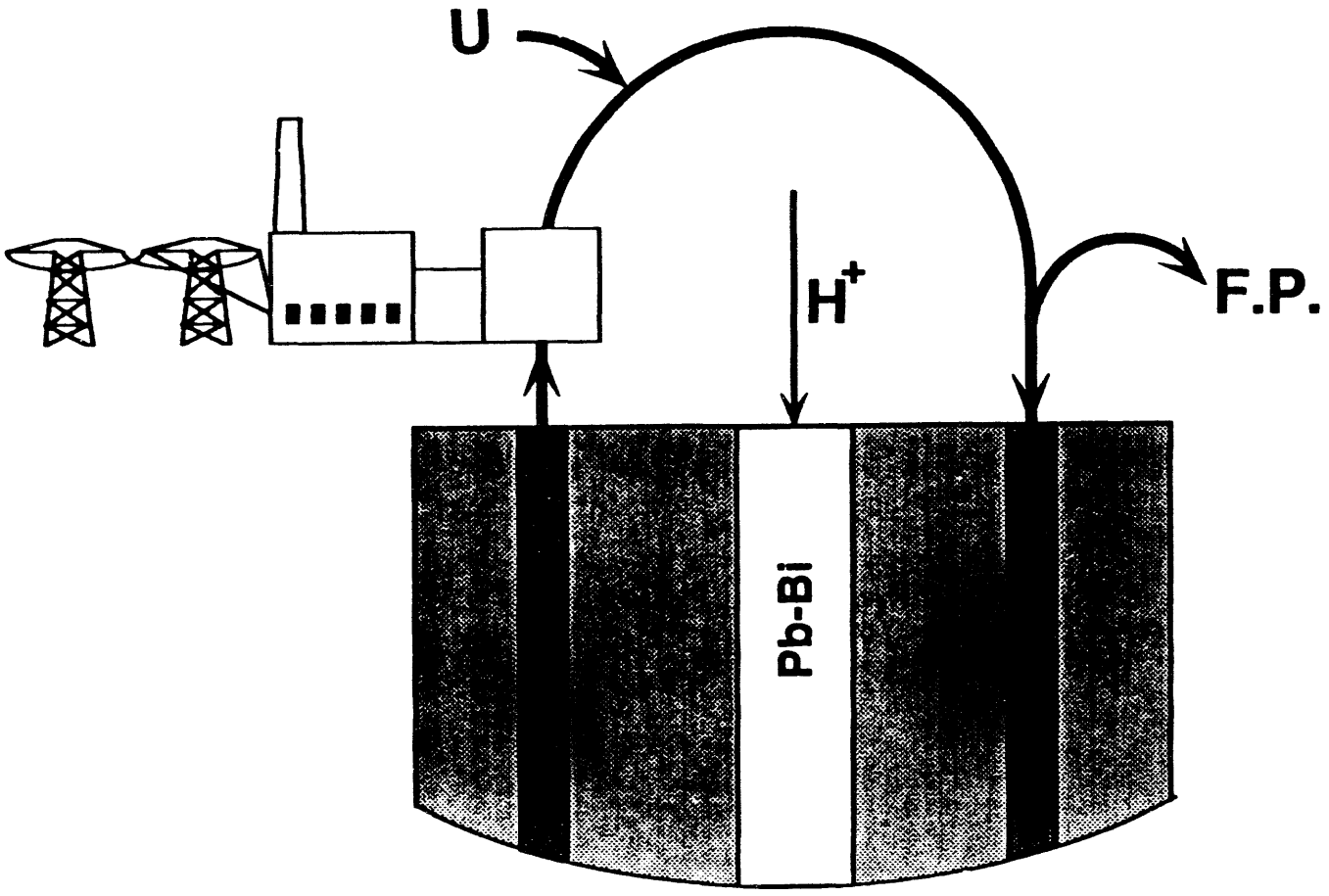
$^{232}Th/^{233}U$ Breeder Technical Issues

- Effect of thermal neutron flux on materials (up to 10^{15} n/cm^2/s.)
- Radiolysis of process materials (solvents/resins) by ^{233}Pa (256 KeV β^-)
- Radiological safety/handling concerns for ^{233}Pa Extensive remote operation experience at national laboratories
- Fluoride volatility processing of ^{233}U may increase neutron exposures due to (α,n) reactions of fluorides

Loop 2: ^{233}U Molten Salt Fission Loop

Loop 2 represents the key feature of the energy production without a long-lived waste-stream concept: In this loop fission energy is extracted from ^{233}U with a high thermal efficiency circulating media, and the heat energy is transferred through a second salt media to a steam generating plant. Meanwhile, the fuel, salt, and fission products are processed separately.

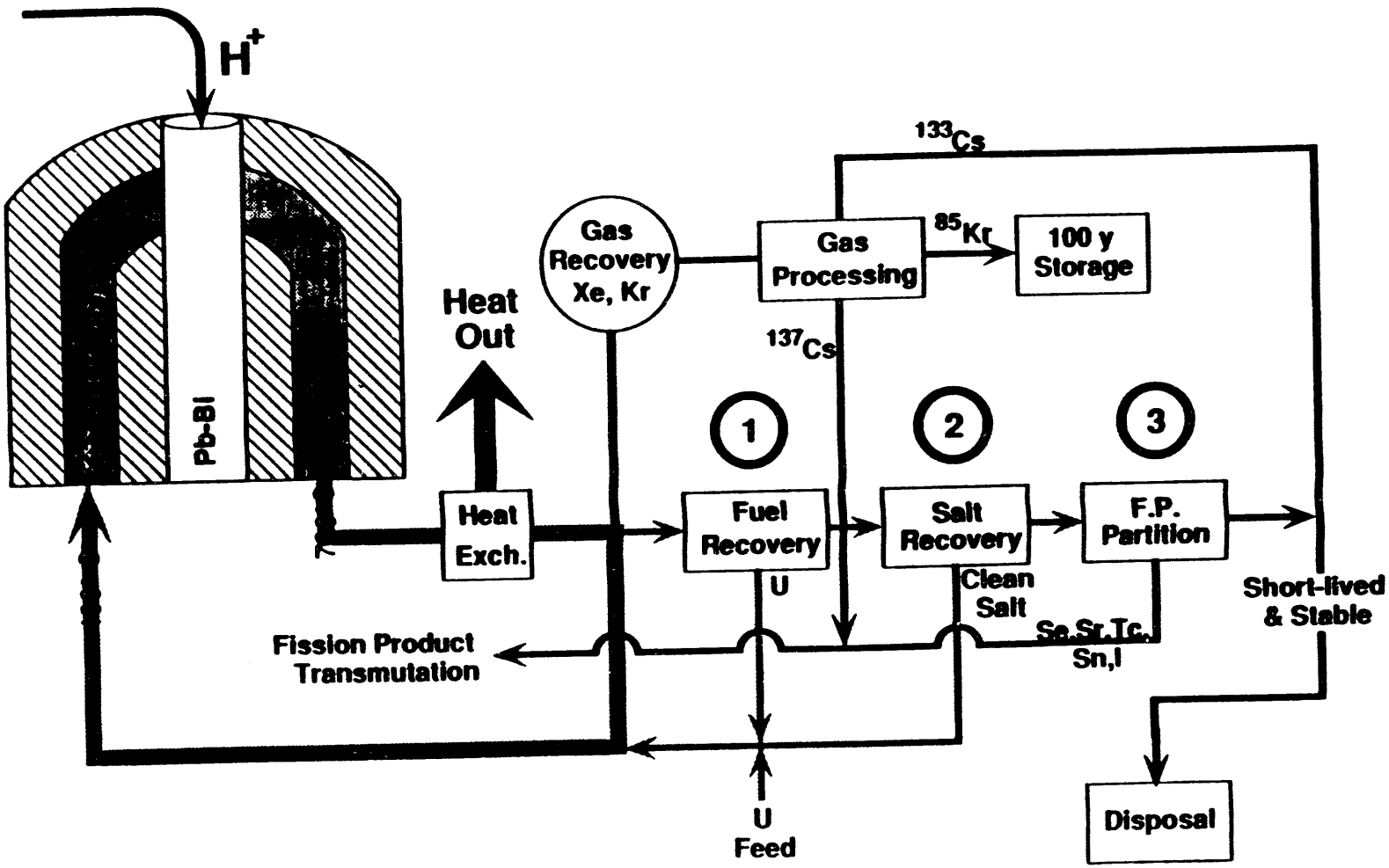
Loop 2: ^{233}U Molten Salt Fission Loop



Chemical Processing of the Molten Salt Fission Loop Relies on Three Key Steps

In this schematic we show only the principal features of the molten salt processing system as we have defined it thus far. Beginning with the high temperature (775 °C) side of the transmuter these are: the heat exchanger, gas sparging/recovery operation, fuel recovery process, salt recovery process, and finally the fission product partitioning sequence. Central to the processing of the molten salt is the uranium fuel recovery, molten salt recovery, and lastly, the fission product partitioning. Because of the dilute loading of actinides, and the subsequent small concentrations of fission products (approximately 3 kg/day), as well as the presence of short-lived fission products.

Chemical Processing of the Molten Salt Fission Loop Relies on Three Key Processes



Features of the Molten Salt Fission Process Loop

Much of the technology for the fission process loop has been adapted from the ORNL molten salt reactor experiment (MSRE) and studies for the molten salt breeder reactor (MSBR). A key feature of the molten salt loop is the use of $^{7}LiF/BeF_2$ salt as a solvent/coolant for the fission reactor. The secondary coolant, consisting of $NaF/NaBF_4$ to transfer heat to the steam plant, was also developed for the ORNL program. A critical difference between this program and the MSRE program is the use of reduced actinide loadings. Rather than 12 % Th and 0.3 % U, our preliminary studies indicate that power generation is feasible using no thorium and only 0.1 % uranium, due to the enormous thermal fluxes produced in the spallation source. Batch, rather than continuous processing of the salt on a daily basis (~10 %/day) permits isolation of the small quantity of fission products produced (~ 3 kg/day) and subsequent storage or processing.

Features of the Molten Salt Fission Process Loop

- Molten salt matrix ($^7LiF/BeF_2$) for high thermal efficiency
- Coolant system $NaF/NaBF_4$ for heat transfer to steam generator
- Actinide loadings of ~0.1% are sufficient at high flux for power generation: MSRE required 12% Th, 0.3% U fuel
- **Batch** processing of salt (10%/day) to remove fission products

Features of the Molten Salt Fission Process Loop (cont.)

A feature of the MSRE experience was the low solubility of gaseous fission products (Xe and Kr) from the reactor. In addition to maintaining a low neutron poison factor, highly radioactive ^{85}Kr is removed within 60 seconds of its formation. After cryoseparation of the two elements, Kr can be absorbed on heated graphite/charcoal to a density approaching that of liquid Kr for 107 year storage. As discussed on the following page, the xenon isotopes can be milked at appropriate intervals to separate the Cs daughters. Because of its large thermal neutron cross-section, nearly all of the ^{135}Xe will be transmuted to stable ^{136}Xe prior to leaving the reactor.

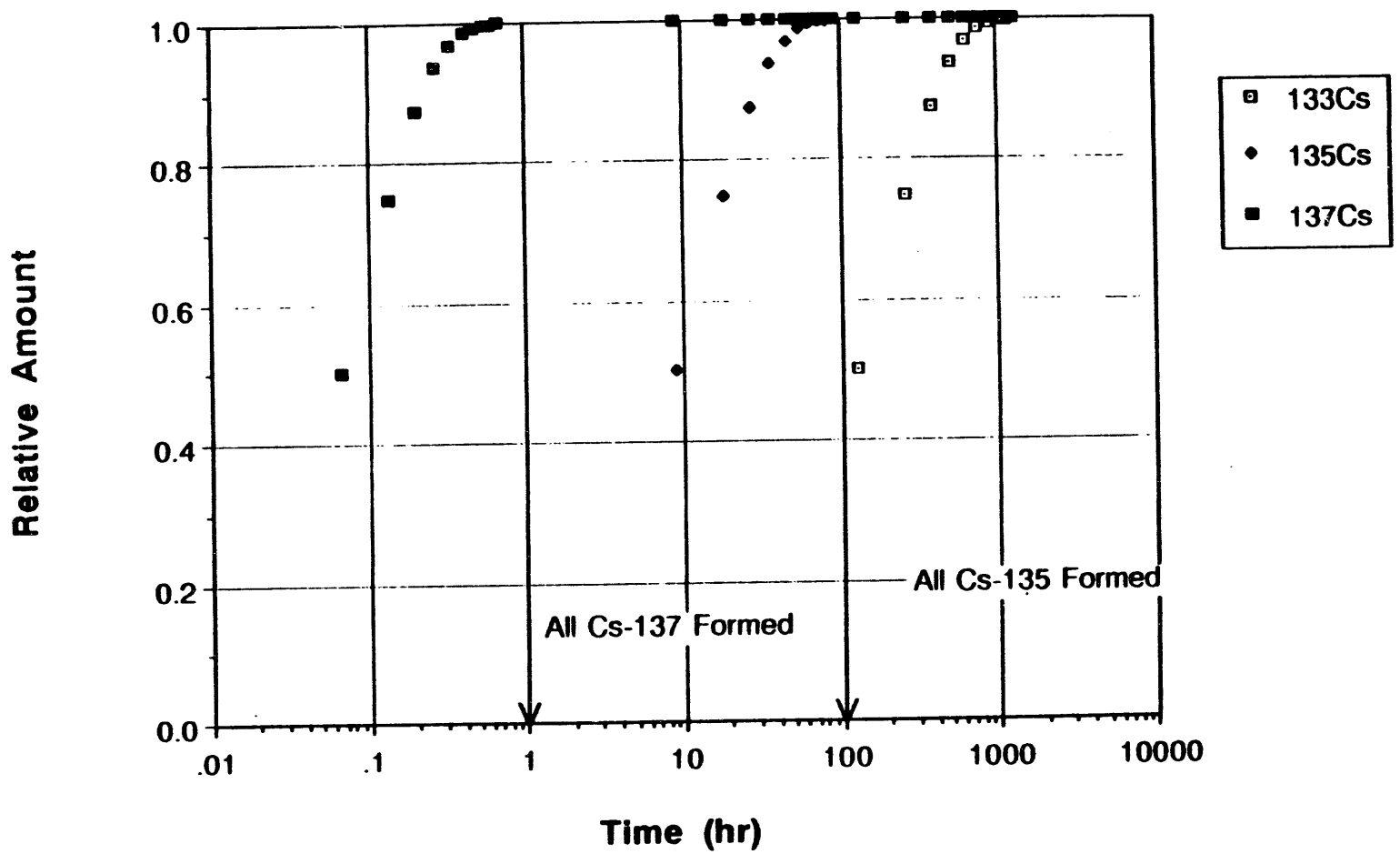
Features of the Molten Salt Fission Process Loop (Cont.)

- Sparging of gaseous fission products (Xe, Kr) with He
- Cryoseparation of Kr, Xe
- Graphite adsorption/storage of 10.7 yr ^{85}Kr
- Milking/separation of $^{133,137}Xe$ to separate cesium isotopes
- ^{135}Xe will transmute ($\sigma_\gamma = 2.6 \cdot 10^6$) to stable ^{136}Xe; only small amounts of ^{135}Cs will be present (from direct fission)

Growth of Cs Isotopes due to Xenon Decay

The differing half-lives of ^{133}Xe, ^{135}Xe, and ^{137}Xe offer a convenient separation scheme for the different Cs daughters, two of which must be transmuted. An automated process is possible whereby the xenon gas is pumped from one chamber to another after an hour interval to remove all the ^{137}Cs. The remaining gas, nearly 100% ^{133}Xe, will decay in 52 days to stable ^{133}Xe, which can be used or processed as low-level waste.

Growth of Cs Isotopes due to Xenon Decay



Critical Technologies for Molten Salt Processing

For the molten salt fission loop to be successful, three critical processes must be in place. The purpose of these three processes is to remove the fission products from the molten salt to maintain as low a neutron poison ratio as possible. First, the ^{233}U fuel must be removed from the salt and recycled to the reactor as UF_4. In the MSR experiment, this was accomplished by fluorinating the uranium to UF_6 with elemental fluorine directly from the molten salt, a technology with great promise as well as serious technical challenges. In the future, we hope to examine this option in greater detail.

In the second step, the bulk of the fission products are separated from the molten salt in order to recycle the clean salt to the reactor. At Oak Ridge, reductive extraction into molten bismuth containing varying amounts of lithium metal was used to separate the rare earths and protactinium from the salt matrix. Another process which may have some applicability, and which we wish to evaluate further is fractional crystallization using a lower solubility material such as CeF_3.

Critical Technologies for Molten Salt Processing

1. Separation of U from salt at ~0.1% concentration
   - Allows minimal hold-up of fissionable material

   Technology: Oak Ridge MSRE employed fluorination

2. Separation of bulk fission products from salt
   - Return of salt to process stream; rapid isolation of fission products
   - Fission products can then be stored as low level waste, separated for transmutation, or held for future processing

   Technology: Extraction of fission products into liquid metal (Bi); fractional crystallization

Critical Technologies for Molten Salt Processing (cont.)

Finally, the third critical step in processing the molten salt is partitioning the fission products to isolate those materials requiring further transmutation from those materials which may be sent directly to low-level waste disposal. Many of the elements of interest are so-called noble metals, such as Mo, Nb, Te, Ru, and Tc, or semi-noble metals, such as Sn, which will not form fluorides in the molten salt in the absence of an oxidant. Presumably these elements can be separated by filtration or centrifugation from the salt. On the other hand, the iodine will likely be fluorinated from the salt with the uranium, and can easily be separated with appropriate chemistry or distillation.

Once the bulk of the fission products have been separated from the salt, aqueous processing of the elements in the third loop can be undertaken. For example, ^{137}Cs and ^{90}Sr will be separated, stored during the life of the reactor (30 yrs), and transmuted near the end of the reactor life to stable isotopes. Once dissolved, these materials may be separated from the remainder of the fission products by advanced extraction process which have been recently developed. A discussion of these available processes will follow on later slides.

Critical Technologies for Molten Salt Processing (Cont.)

3. Separation of selected fission products for further processing:

   Because of high flux levels, many longer-lived radioactive species will be burned to stable or short-lived products (e.g. ^{135}Xe)

   - ^{99}Tc, ^{129}I, ^{79}Se, ^{126}Sn, for further transmutation (as aqueous stream)

   Technology: Tc and Sn (perhaps also Se) will not dissolve in salt when formed. Bulk of I will fluorinate with UF_6

   - Separation and delayed processing for 137Cs, 90Sr. [30-yr retrievable storage followed by transmutation]

   Technology: Advanced extraction agents

Technical Issues for Molten Salt Fission Loop

Our choice of fluorination as a separations process for uranium is based on the extensive experience with this technology not only at ORNL and Los Alamos, but throughout the complex as well. An additional factor is the compatibility of the salt medium with elemental fluorine. In dialogues with at ORNL, we have recently become aware of the serious materials problems encountered in the process employing this technology. To address this problem, we intend to look not only at the methods developed at ORNL, but also at advanced materials such as nickel-based superalloys or ceramics developed over the last 20 years. An alternate technology we will examine is electrochemical fluorination, a widely used industrial technique. For example ZrB_2 or TiB_2 electrodes are routinely used to prepare metallic Al from molten Na_3AlF_6 at temperatures approaching 1000° C, and DuPont is exploring the electrochemical generation of F_2 directly from molten CaF_2.

Technical Issues for Molten Salt Fission Loop

- Fluoride Volatility Recovery of Uranium: gaseous F_2 in LiF/BeF_2 at ~500°C

Advantages: Removes >99% U and some fission products
Large experience base in U, Pu volatility at national labs

Issues: Must be demonstrated in pilot-scale (molten salt)
Corrosion/materials problems in molten salts

Technologies: Advanced materials (e.g. NiAl, ceramics) resistant to F2 in molten salts

- Electrochemical Fluorinations (large industrial base)

Technical Issues for Molten Salt Fission Loop: $^{7}LiF/BeF_2$ Molten Salt

For the choice of salt media, we have relied heavily on the reasoning and experience developed at ORNL and published in the nuclear and chemical literature. For this process, LiF/BeF_2 has the advantages of transparency to thermal neutrons, an extensive experience base, good physical properties including melting point and low vapor pressure, as well as being an excellent solvent for many inorganic fluorides. Except for certain elements such as Cr, the salt is also nonoxidizing.

Technical Issues for Molten Salt Fission Loop

$^{7}LiF/BeF_2$ molten salt

Advantages: Essentially transparent to neutrons

- Extensive experience base from ORNL
- Good physical properties; low melting, low vapor pressure
- Excellent solvent for a wide range of fluorides
- Nonoxidizing/nonreducing for many fluorides

Technical Issues for Molten Salt Fission Loop: $^{7}LiF/BeF_2$ molten salt (cont.)

Some technical issues which must be addressed in this program are the relative lack of separations processes developed for molten salts. These are manifested in the absence of suitably developed ion exchange materials for LiF/BeF_2 and the limited numbers of solvent extraction systems appropriate for these salts. While fractional crystallization may be a good technique for separating a large number of materials from these salts, deleterious impurities may be introduced during the process.

Technical Issues for Molten Salt Fission Loop

$^{7}LiF/BeF_2$ molten salt (Cont.)

Issues: Separations processes in molten salts have not kept pace with aqueous systems

- Ion exchange for LiF/BeF_2 are not well developed
- Solvent extraction (liquid metal): corrosion, addition of high-σ impurities
- Excellent solvent properties complicate purification and recycle of LiF/BeF_2: Fractional crystallization/salting may add unacceptable levels of impurities

Technical Issues for Molten Salt Fission Loop

We have tentatively identified Hastelloy® N, a high nickel alloy developed for the MSRE program, as the material of choice for containing the salt in this loop. Except for a slight depletion of Cr from the surface of this metal, Hastelloy® N is nearly completely impervious to molten salts. However, under intense neutron fluxes, ^{58}Ni (68.3 % abundance) is transmuted to ^{59}Ni, having a relatively high cross section for (n,α) and (n,p) reactions which lead to gas embrittlement of the material. To solve this problem, which will exist only in the reactor and immediate surroundings, it will be necessary to prepare Ni depleted in ^{58}Ni. The plasma separation process, developed by TRW and stored at ORNL, is capable of separating multikilogram quantities of Ni isotopes. Another possibility which may also satisfy the need for materials compatible with handling elemental fluorine in molten salts is the use of coatings for low-nickel stainless steels.

Technical Issues for Molten Salt Fission Loop

Materials for handling $^{7}LiF/BeF_2$

- Hastelloy® N [71% Ni, 16% Mo, 7% Cr, trace Fe, Ti, C] for containing molten salts at elevated temperatures

  Advantage: low corrosion, ready availability

  Issues: High flux transmutes ^{58}Ni to ^{59}Ni with a high (n,α) and (n,p) cross-section ($\Rightarrow$embrittlement of metal)

  Technology: Separation of Ni isotopes for reactor tubes

  [Plasma Separation Process (PSP) developed by TRW available for ~100 kgs/yr separation of Ni isotopes]

- Coatings for stainless steel (e.g., ZrB_2)

Technical Issues for Molten Salt Fission Loop

An issue for partitioning the fission products once they are separated from the salt involves the difficulty with large scale separations of Cs and Sr from acid streams. Recently, processes have been developed at the University of South Carolina and Argonne National Laboratory involving crown ether extraction of Cs as well as Sr into organic solvents. Although good separations are achieved, crown ethers are inherently expensive reagents ($10/gram). Another process, developed in the Soviet Union, uses cobalt dicarbollide, a carborane reagent to achieve good separations of both elements from aqueous acid. Although carbollides are also expensive (due to relatively little demand), a Czechoslovakian firm has prepared approximately 100 kgs for use in Soviet pilot plant scale operations.

Technical Issues for Molten Salt Fission Loop

Available Aqueous Fission Product Separation Technologies

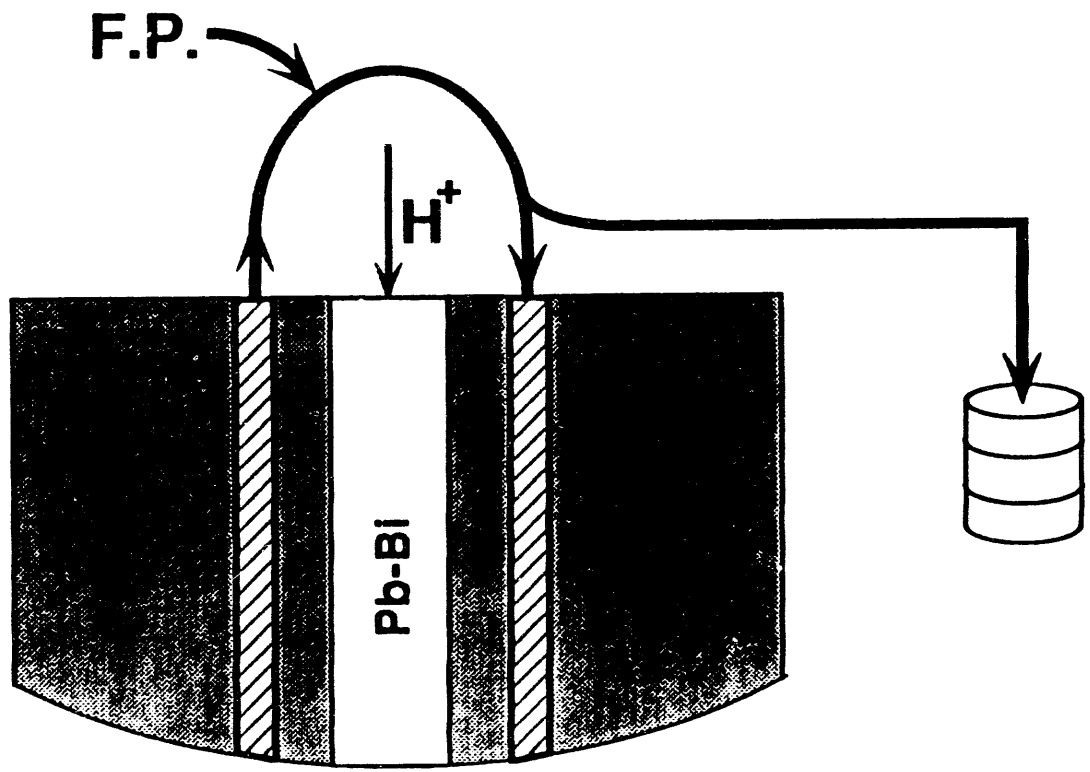
- Crown ether extraction of Cs, Sr from HNO_3: good selectivity and separation; expensive reagents

- Soviet/Czech process for Cs, Sr extraction (Co-dicarbollide): excellent selectivity and separation; expensive reagents, toxic solvent

Loop 3: Fission Product Transmuter

The third loop of the reactor concept exists to transmute the fission products which are of greatest concern long-term storage. In the inner region of the transmuter, substantially higher fluxes will transmute even transparent materials such as ^{137}Cs and ^{90}Sr.

Loop 3: Fission Product Transmuter



Fission Product Transmutation Loop

Because of the relatively small mass of fission products produced in the molten salt reactor/transmuter, we propose to accumulate the elements of interest and transmute them in separate "campaigns" for each element. By using this approach, we are able to select the best species for each element to separate the transmuted product from the fission product nuclide. By allowing the material to accumulate, we are able to concurrently optimize chemical separations methods for the fission products where such methods are not well-developed beyond the analytical scale.

Fission Product Transmutation Loop

- Small mass of fission products (approximately 3 kg daily) permits storage and separate campaigns to transmute

- Aqueous process allows flexibility in choosing species for each nuclide to transmute

- Delay processing of fission products

  Does not hold up energy generation; allows time for optimization of separation/transmutation scheme

Few Fission Products Require Transmutation

Because of the high flux in the reactor, many elements (e.g. ^{135}Xe, ^{151}Sm) will be transmuted directly and will thus not need to be separated. Other elements (e.g. Tc, Sn) will be easily separated because of their poor solubility in LiF/BeF_2. Of the eleven elements which comprise the bulk of the long-lived fission product activity, only six of these will require transmutation. Others will either be stored for decay (^{85}Kr, ^{107}Pd, and ^{93}Zr) or transmuted in the reactor.

Few Fission Products Require Transmutation

Nuclide	$T_{1/2}$ (yrs)	σ_γ (barns)	^{233}U (kg/yr)	^{239}Pu (kg/yr)	PWR (kg/yr)
^{79}Se	$6.5 \cdot 10^4$	10 ?	0.48	0.15	0.19
^{90}Sr	28.8	0.9	24.8	7.5	13.5
^{99}Tc	$2.1 \cdot 10^5$	22	19.5	24.6	25.1
^{126}Sn	$1 \cdot 10^5$	0.14	1.27	1.28	0.91
^{129}I	$1.6 \cdot 10^7$	19	7.70	6.60	5.87
^{137}Cs	30.	0.11	37	37	31
^{85}Kr*	10.7	1.7	1.44	0.31	0.40
^{93}Zr*	$1.5 \cdot 10^6$	1	26.2	14.4	23.3
^{107}Pd*	$6.5 \cdot 10^6$	1.8	0.49	14.1	7.4
^{135}Cs†	$3 \cdot 10^6$	8.7	33.6	41.4	9.5
^{151}Sm†	90	15,000	1.89	4.60	0.38

†^{151}Sm and ^{135}Xe parent of ^{135}Cs will be transmuted in the reactor.

*^{85}Kr, ^{93}Zr, ^{107}Pd will be stored, not transmuted.

Fission Product Transmutation Chemistry

The separations chemistry of the fission products will be vastly simplified by processing each element separately. In addition for many of the elements, the transmutation products will have vastly different chemistry than the fission product elements. Expected products are shown on the following two pages. For most of these separations (e.g. Sr/Zr; Sn/I) we will take advantage of existing radiochemical or other facile separations chemistry. The final destination of the transmuted products, as well as the vast bulk of the fission products which do not require transmutation will be grouting and above ground storage or disposal.

Fission Product Transmutation Chemistry

- Tramsmutation Products have vastly different chemistry from starting elements (e.g., Cs/Ba; I/Xe)
- Chemistry needs to be developed on a element-by-element basis
- Grouting/storage of low-level and stable fission products
- Grouting/storage of secondary transmutation products (i.e., Ru from ^{99}Tc, etc.)

Fission Product Transmutations Lead to Stable Nuclides

Fission Product Transmutations Lead to Stable Nuclides

^{79}Se $\qquad$ $^{79}Se \xrightarrow{n,\gamma} {}^{80}Se$

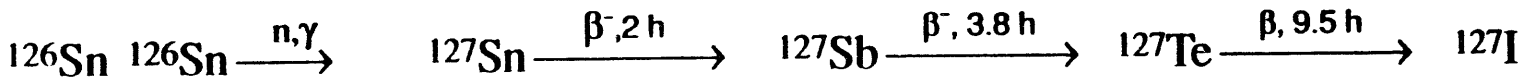
^{90}Sr $\qquad$ $^{90}Sr \xrightarrow[(\sigma=0.8)]{n,\gamma} {}^{91}Sr \xrightarrow{\beta^-,9.5\ h} {}^{91}Y \xrightarrow{\beta^-,58d} {}^{91}Zr$

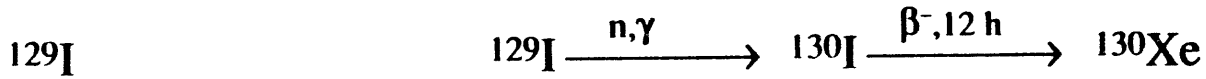
$^{91}Y \xrightarrow[(\sigma_\gamma=1.4)]{n,\gamma} {}^{92}Y \longrightarrow {}^{92}Zr$

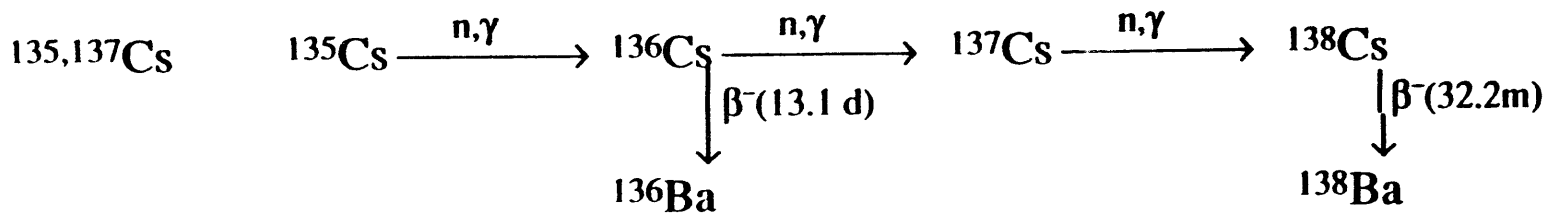
^{99}Tc $\qquad$ $^{99}Tc \xrightarrow{n,\gamma} {}^{100}Tc \xrightarrow{\beta^-,\ 16\ s} {}^{100}Ru$

Fission Product Transmutations

Fission Product Transmutations



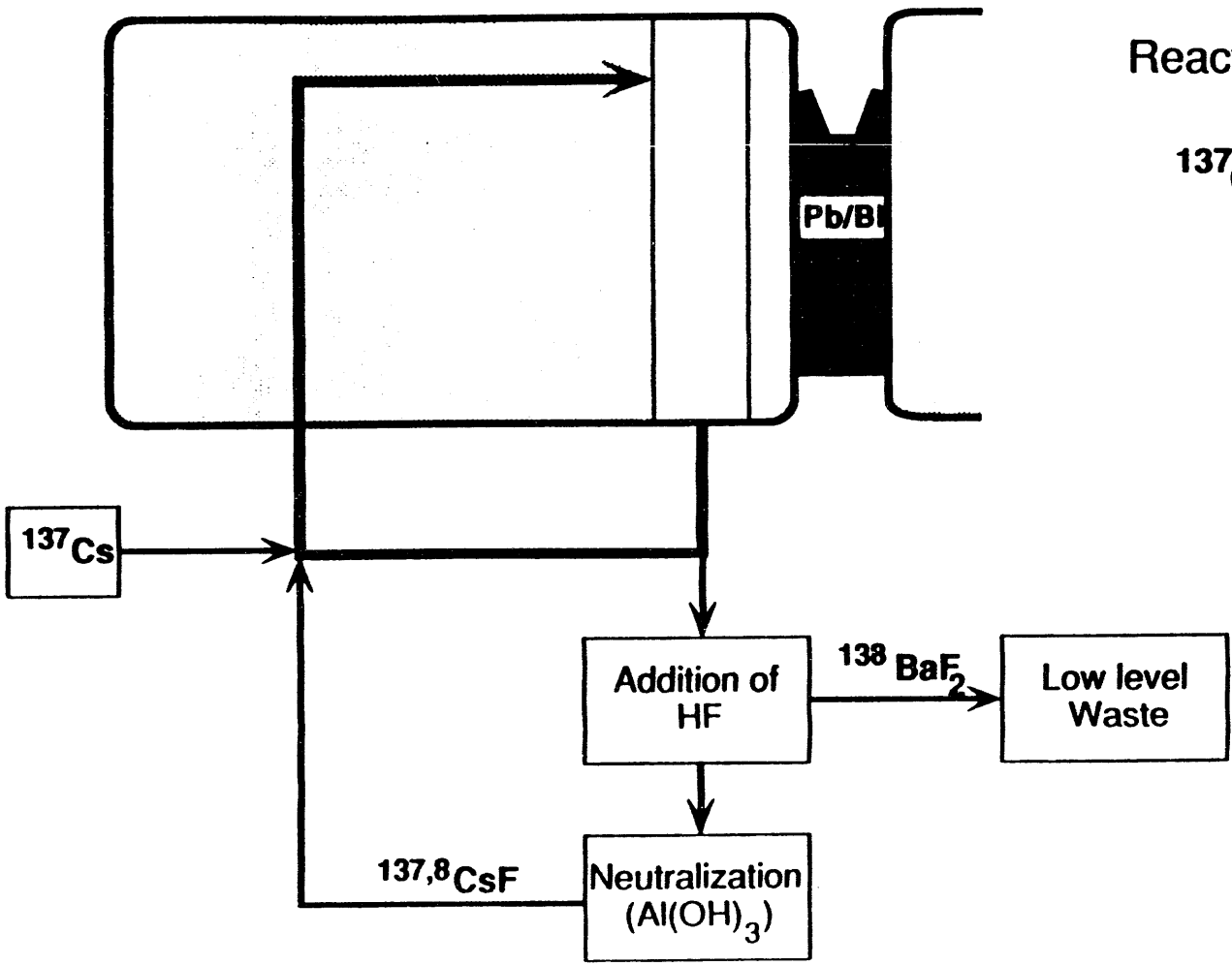




ATW Fission Product Transmutation Loop

One possible schematic for separating fission product cesium from its barium transmutation product takes advantage of the poor solubility of BaF_2 relative to CsF in acid media. Aluminum is added afterwards as needed to complex the excess fluoride.

ATW Fission Product Transmutation Loop



Reactions:

$$^{137}Cs \xrightarrow[(\sigma = 0.11)]{n,\gamma} {}^{138}Cs \xrightarrow[(t_{1/2} = 32.2\ m)]{\beta^-} {}^{138}Ba$$

Immediate Research Needs

Immediate Research Needs

232Th/233U Breeder Loop

- System definition for Th, U/Pa loadings; expected radiation effects on solvents and process materials

233U/Molten Salt Fission Loop

- Definition of system variables: salt volume, flow rates, processing quantities
- Examination of alternate materials for fluorination process
- Survey of alternate processes for salt purification/fission product removal

Near-Term Research Needs

Near-Term Research Needs

$^{232}Th/^{233}U$ Breeder Loop

- Radiation effects of ^{233}Pa must be defined and studied

^{233}U/Molten Salt Fission Loop

- Materials for fluoride volatility in 4-500°C $^{7}LiF/BeF_2$
- Separations techniques for fission products, U from molten salt
- Isotope separation for low-^{58}Ni Hastelloy® N/ alternate materials

Fission Product Transmutation Loop

- Materials for high-flux transmutation of ^{137}Cs, ^{90}Sr

Molten Salt Chemistry: Summary

Molten Salt Chemistry: Summary

$^{232}Th/^{233}U$ Breeder Loop

- Separations Chemistry of Th from U, Pa, and U from Pa is well documented

^{233}U Molten Salt Fission Loop

- Large body of experience with $^{7}LiF/BeF_2$ at Oak Ridge
- Fluorination of UF_4 to UF_6 will remove >99% of U from salt
- Fission products separation/salt purification techniques are issues

Fission Product Transmutation Loop

- High flux reduces number of fission products that need to be transmuted
- Techniques are available to separate Cs, Sr from bulk fission products
- Most transmutation products will be easily separated from their precursors

SUMMARY

Summary and Research Issues

Edward Arthur
Theoretical Division

Summary

A New Los Alamos Concept for Accelerator Transmutation of Waste and Fission Energy Production

Edward D. Arthur

December 11, 1990

Summary

- ATW is a new approach to waste transmutation
- Its intense thermal neutron flux enables
  - New regime for actinide burn → net n production
  - Efficient transmutation of fission products
  - New low inventory operating regime
  - New options for process chemistry

ATW Has Two Focus Areas

- Near Term - Defense Wastes
- Advanced - Fission Energy with a Minimal Long-Lived High-Level Waste Stream
- Both areas utilize accelerator system advantages to
  - Transmute all components of HLW
  - Transmute in a low-inventory environment
  - Utilize advanced chemistry processing methods

Near-Term Application
Hanford Defense Wastes

- Reasonable extrapolation of several technology areas
  - Linac design
  - Neutron spallation source design
  - Aqueous chemistry
- No apparent "show-stoppers"
- Production plant could be developed in approximately 10 years

Advanced Concept

- Energy production potential
  - Uses thorium (thousands of years' supply)
  - In situ breeding
  - Minimal long-term HLW stream

- Enablers
  - Spallation physics impact on fission systems
  - Low inventory, continuous flows → new approach to required separations

- Direct extrapolation of near-term development areas
  - -Accelerator
  - Target/blanket
  - Aqueous processing loops

Issues

Near-Term Application

- High-current CW accelerator operation
- n/p production, fundamental σ's, and blanket optimization
- PbBi containment and materials compatibility
- Radiation damage
- Aqueous chemistry process loop definition
- Waste stream identification and minimization
- Overall mass balances
- Costs and safety

Advanced Application

- Molten salt process definition
- Materials compatibility
- Costs

Relevant Capabilities

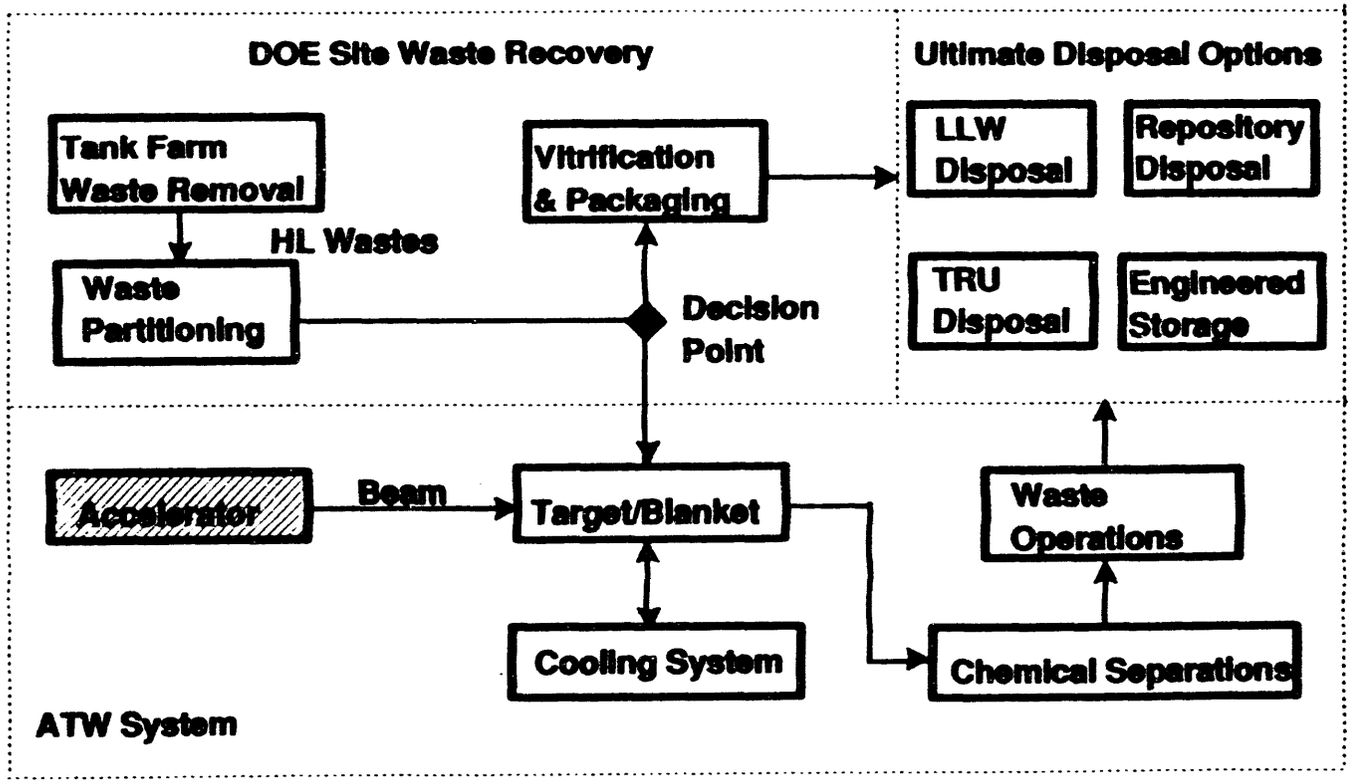
- Demonstrated high-current accelerator component operation (low duty cycle) (LAMPF, ATS, GTA)
- High-intensity neutron sources (LANSCE, WNR)
- Advanced calculational tools for target design and optimization
- Processing chemistry
  - Tc experience
  - Plutonium Facility (TA55) actinide processing
- Materials
  - ceramics, material development
  - radiation damage research

Immediate Tasks for Near-Term Application Development

- Accelerator injector experiment → demo current, emittence properties
- Optimize spallation target choices (materials, geometry)
- Review yield experiment database → assess spallation product calculation accuracy
- Optimize blanket design (materials choices, neutron leakage, absorption)
- Assess critical nuclear data needs (σ's, n spectra)
- Systems analysis of near-term concept - tradeoffs and impact

A Complex-Wide Systems Model will be Developed to Assess System Trades

Trades: Integrated Dose, Waste Volumes, Technology Payoffs, Costs



Immediate Tasks
Near-Term Application

- Initial experiments to verify target fluid dynamics
- Materials compatibility - review database to define future experiments
- Molten PbBi - product compatibility mix experiment
- Develop flowsheets for FP and actinide aqueous loops
- Demo scale technetium/ruthenium separations loop
- Demo scale actinide aqueous loops (ATLAS Facility)
- Identify waste streams - develop waste processing flowsheets
- Safety issues, conceptual design details, costs

Immediate Tasks
Advanced Application

- Full 2D or 3D neutronics analysis of proposed system
- System definition for Th/U/Pa loading, radiation levels, effects on processing
- Definition of U233, molten salt processing loops
- Initial definition of fission product extraction techniques
- Examine database from Molten Salt Reactor Experiment
- Develop alternatives for fluorination process

Next Level of R&D
Proof of Principle

- Couple DTL to RFQ - demonstrate high current operation
- Beam load simulation of CCL
- High efficiency RF source development
- Experimentally verify n/p
- Thermal, epithermal neutron σ measurement
- Spallation product yield experiment
- Improved medium calculational models
- PbBi test loop

Next Level R&D (Cont.)

- Demonstration of Am/La separations
- Begin development of on-line analytical techniques
- Demonstrate waste polishing techniques
- Facility design for waste processing
- Identify, verify radiation resistant material choices
- Demonstrate fission product extraction in aqueous loop
- LAMPF experiments with aqueous systems
- Continue systems optimization study
- Safety analysis, costing, conceptual design details

Next Level R&D
Advanced Concept

- Extraction of Pa from Th
- Begin development process for U233 removal, salt purification, fission product removal
- Extraction of selected products from fission product mix
- Materials compatibility investigations
- Accelerator efficiency optimization

END

DATE FILMED

1/12/94