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TITLE A NEW CONCEPT FOR ACCELERATOR DRIVEN TRANSMUTATION OF NUCLEAR WASTES

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ABSTRACT

A new concept for an accelerator-driven transmutation system is described. The central feature of the concept is generation of intense fluxes of thermal neutrons. In the system all long-lived radionuclides comprising high-level nuclear waste can be transmuted efficiently. Transmutation takes place in a unique, low material inventory environment. Presently two principal areas are being investigated for application of the concept. The first is associated with cleanup of defense high-level waste at DOE sites such as Hanford. The second, longer term area involves production of electric power using a coupled accelerator-multiplying blanket system. This system would utilize natural thorium or uranium and would transmute long-lived components of high-level waste concurrently during operation.

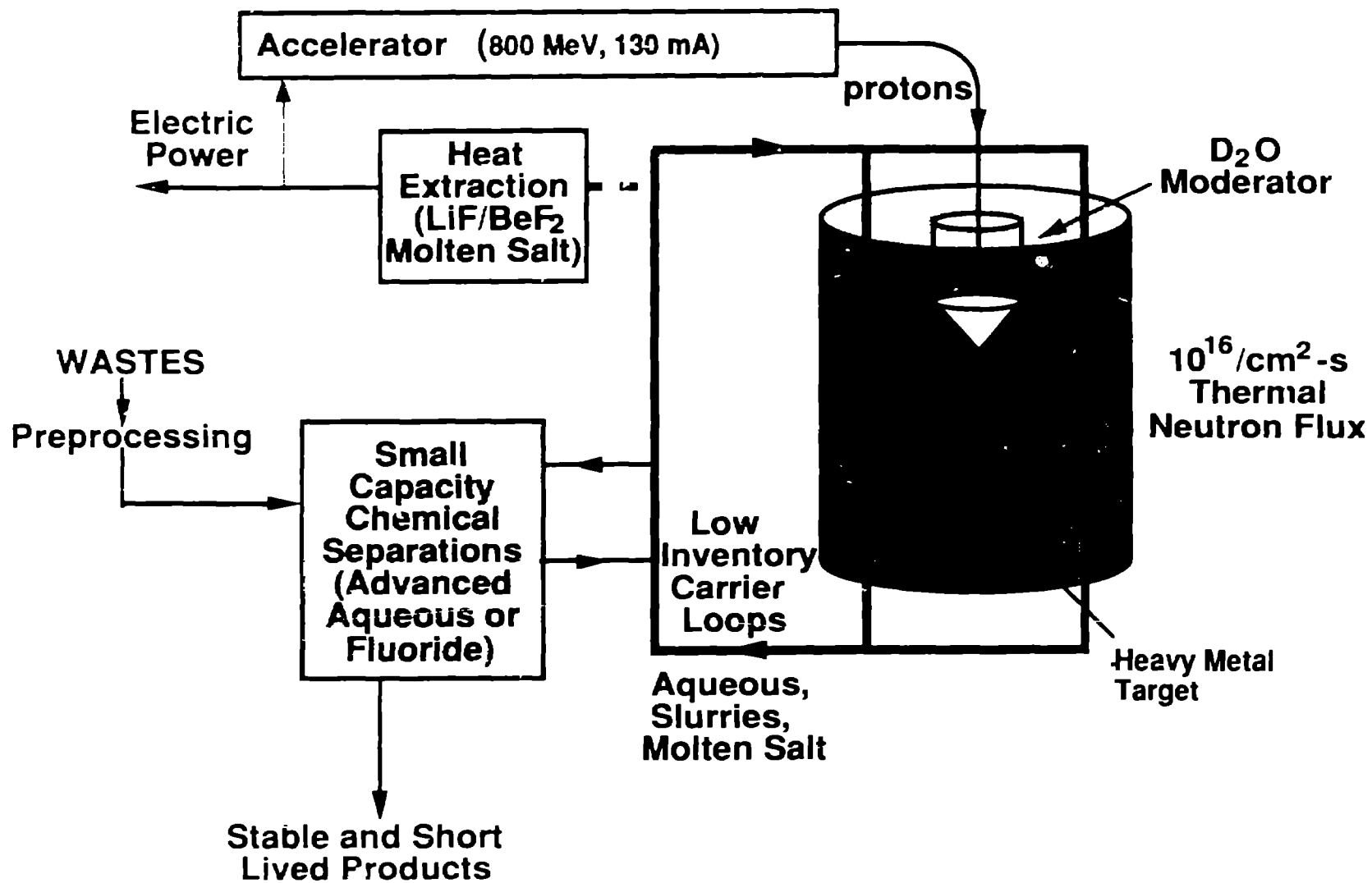
INTRODUCTION

A new concept for transmutation of long-lived components of high-level nuclear wastes (HLW) is under development at Los Alamos National Laboratory. The concept, called ATW, uses an accelerator-driven intense thermal neutron source to achieve several significant advances in system performance. Features of the concept include the ability to destroy all long-lived radioactive species (both fission products and actinides), operation in a high throughput, low-material inventory environment, small capacity, high decontamination factor chemical processing, and improved safety and environmental features resulting from accelerator driven and low material inventories.

This concept is a result of several technology advances. High-current linear accelerator technology under development for the Strategic Defense Initiative enables the design of medium energy (around 1 GeV) proton accelerators that can operate in the current regime required (approximately 100 mA) for an ATW system. Such an accelerator can be used to produce intense fluxes of neutrons (10^{16} n/cm²/sec) at largely thermal energies. This development led to the identification of a new transmutation performance region characterized by rapid burnup, dilute material loadings, and low material inventories present in the system. These features also imply continuous material flow and small capacity chemical processing systems. This last feature has led us to investigate new chemical processing options that will be described later.

COMPONENTS AND UNIQUE FEATURES

Figure 1 illustrates the main features of the ATW system. A medium energy accelerator provides an intense proton beam to a central neutron production target. (Depending upon the application this accelerator would operate at energies around 1 GeV and at average current levels of 100 to 150 mA). Protons strike a heavy metal production target 50 centimeters in diameter and one meter in length. At an incident proton energy of 800 MeV each proton produces approximately 25 neutrons. This spallation target could be a flowing system based upon use of flowing lead. Solid target designs using pebbles of high-melting point metals such as tungsten or uranium oxide are also under investigation. Surrounding this central production target is a D₂O moderator approximately 3 meters in



diameter. Outside the wall of the central spallation target and extending over significant volumes (> 100 liters) are regions where the neutron flux is high (5×10^{15} n/cm²/sec to 1×10^{16} n/cm²/sec). Within this heavy water moderator materials to be transmuted would flow through a very thermalized neutron environment. A flowing material system is necessary because of high burnup rates that would make use of solid rods impractical. The intense flux characteristics of the system also lead to optimum performance for dilute material loadings (concentrations of less than one volume percent). Thus materials would be circulated in low-inventory carrier loops. Carrier material forms under investigation include oxide slurries, salts dissolved in water (or heavy water), and/or a molten salt such as LiF/BeF₂. Because of the low inventory feature of the system (to be described later), the capacities of the chemistry processing loops are also small (typically tens of kilograms of material). For this reason specialized chemical processing systems can be developed or methods such as ion exchange can be used that may not be practical for systems requiring larger processing capacities. Use of a fluoride molten salt as a carrier material enables two features. First this material can contain the heat generated during actinide fission in a multiplying blanket system operating at a k_{eff} of 0.8 to 0.9. This salt would flow through the system at temperatures of approximately 700^o C and could provide thermal to electric conversion efficiencies greater than forty percent. Under these conditions a coupled accelerator/multiplying blanket system would not only produce enough power to run the accelerator but could supply significant quantities of electricity to the commercial grid at overall system efficiencies around thirty percent. (This idea forms the basis for a new energy production concept reported at this Conference.) Use of molten salt could also enable advanced separations technology based upon fluoride chemistry.

The system described here has several unique features that make it different from other concepts (reactor or accelerator-based) for transmutation. The first involves the feature of high performance with small radioactive material loadings. The transmutation rate of a system is given by

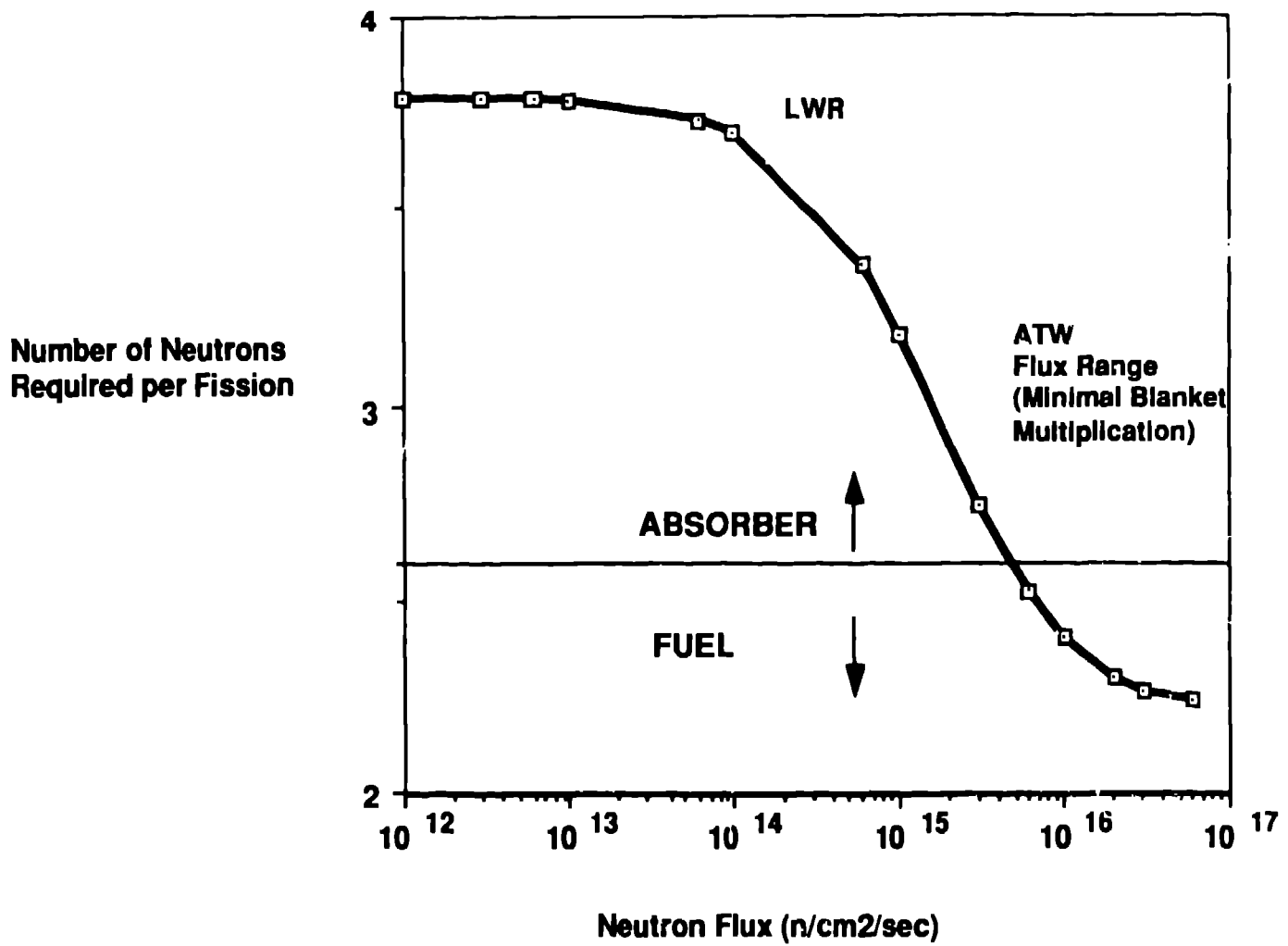
$$T_R = M \Phi \sigma_T$$

where T_R is the transmutation per unit time, Φ is the neutron flux, and σ_T is the cross section for the transmutation process. In the ATW intense thermal neutron flux system, the flux level is up to 50 times higher than thermal systems such as light water reactor. The peak flux level can also

be up to an order of magnitude higher than fast neutron systems. Cross sections for transmutation processes at the thermal neutron energies are often significantly more than an order of magnitude greater than for fast neutron systems. This cross section difference is true even for fission of threshold actinides such as neptunium, as will be explained later. The net impact of these factors is that the mass term in the above expression can be reduced by commensurate amounts. Specifically the high flux, large cross section features of the ATW concept allow material inventory reductions on the order of a factor of 100. The ATW system can achieve transmutation performance with a material inventory of less than 70 kilograms that is comparable to fast neutron systems (reactor or accelerator-driven) requiring inventories of up to 10,000 kilograms.

Another ATW feature enabled by the high flux environment is the ability to efficiently fission higher actinides such as ²³⁷Np. In a neutron flux characteristic of thermal reactors, a neutron interacting with ²³⁷ is captured to produce ²³⁸Np which then decays after 2.1 days to ²³⁸Pu. Neutrons interacting with ²³⁸Pu are also most likely captured to produce ²³⁹Pu. Finally a neutron interacting with ²³⁹Pu will fission approximately 75 percent of the time. This fission produces about 2.7 neutrons. However at least three neutrons were required to produce fission so that in this instance a threshold fissioners such as ²³⁷Np behaves as a neutron absorber. Figure 2 illustrates this result for low flux conditions. There the number of neutrons required to produce a fission is plotted against neutron flux. This result was obtained from calculations that assumed a purely thermal spectrum and followed high actinide production chains up through californium.

Figure 2 also indicates the situation that occurs at higher flux levels. As the flux level increases there is an increasing probability that another neutron will interact with the ²³⁸Np produced during the first step of the process described above before decay can occur. ²³⁸Np has a large thermal fission cross section (> 2000 barns) and such interactions will most likely lead to fission. In this instance 2.7 neutrons are produced in the fission process and two neutrons were required to initiate it. Thus at higher fluxes (generally above about 4×10^{15} n/cm²/sec) neptunium will act as a net producer of neutrons. In addition the effective cross section for fission is a function of flux level and approaches values of about 50 barns for fluxes of 10^{16} n/cm²/sec. This value is significantly



larger than the corresponding fast neutron-induced fission cross section which is on the order of two barns.

Finally the feature of low material inventory has a very important impact on the processing chemistry required in the ATW system. Since inventories are on the order of less than 100 kilograms rather than thousands of kilograms required in more conventional system, small capacity chemical separation systems can be used. As will be illustrated later these have capacities of a few tens of kilograms which means that a working ATW chemical processing system is only a small scaleup from laboratory-sized systems. This feature also allows development of specialized separation processes that may be impractical for larger systems. Examples include use of ozonolysis in technetium/ruthenium separations loops, specialized ligand extractants for actinide/lanthanide separations, and ion exchange methods.

POTENTIAL APPLICATION AREAS

Two possible application areas for an ATW system are being pursued. The first is associated with cleanup of DOE defense high-level waste that has resulted from reprocessing of production reactor fuels. This area will be described in more detail below. The second area involves a concept for energy production using natural thorium or uranium fuels. This system offers the promise of power production at efficiencies comparable to present light water reactors and would have enough extra neutrons for concurrent destruction of long-lived high level waste generated by the system. Reference 1 describes this concept. Both of these applications offer the potential for development of ATW technologies that could significantly impact strategies for high-level waste management. They also provide opportunities for development of a system that is largely independent of policy decisions concerning reprocessing of commercial spent fuel.

A major thrust of ATW concept development has been in the definition and analysis of a point design applicable to transmutation of long-lived components of defense high-level waste. Such waste is stored at sites such as Hanford, Savannah River, and Idaho Falls. It is characterized by large volumes although current data indicate that long-lived fission product and higher actinide components total about 20 metric tons of material. A further emphasis of an ATW system analysis effort has been in the context of a possible application to HLW cleanup at the Hanford site. There about 2000 kilograms of technetium and iodine along with approximately 600 kilograms of actinides such as plutonium, neptunium, and americium exist in waste volumes totaling approximately 200,000 m³. A cleanup strategy based on chemical separations of radionuclides contained in these wastes followed by transmutation could dramatically reduce requirements for vitrification of large volumes of Hanford tank wastes. This in turn would reduce the load upon storage in a geologic repository such as Yucca Mountain. Transmutation of separated wastes could furthermore enable increased options for on-site storage, further reducing the dependence upon deep geologic storage in cleanup scenarios.

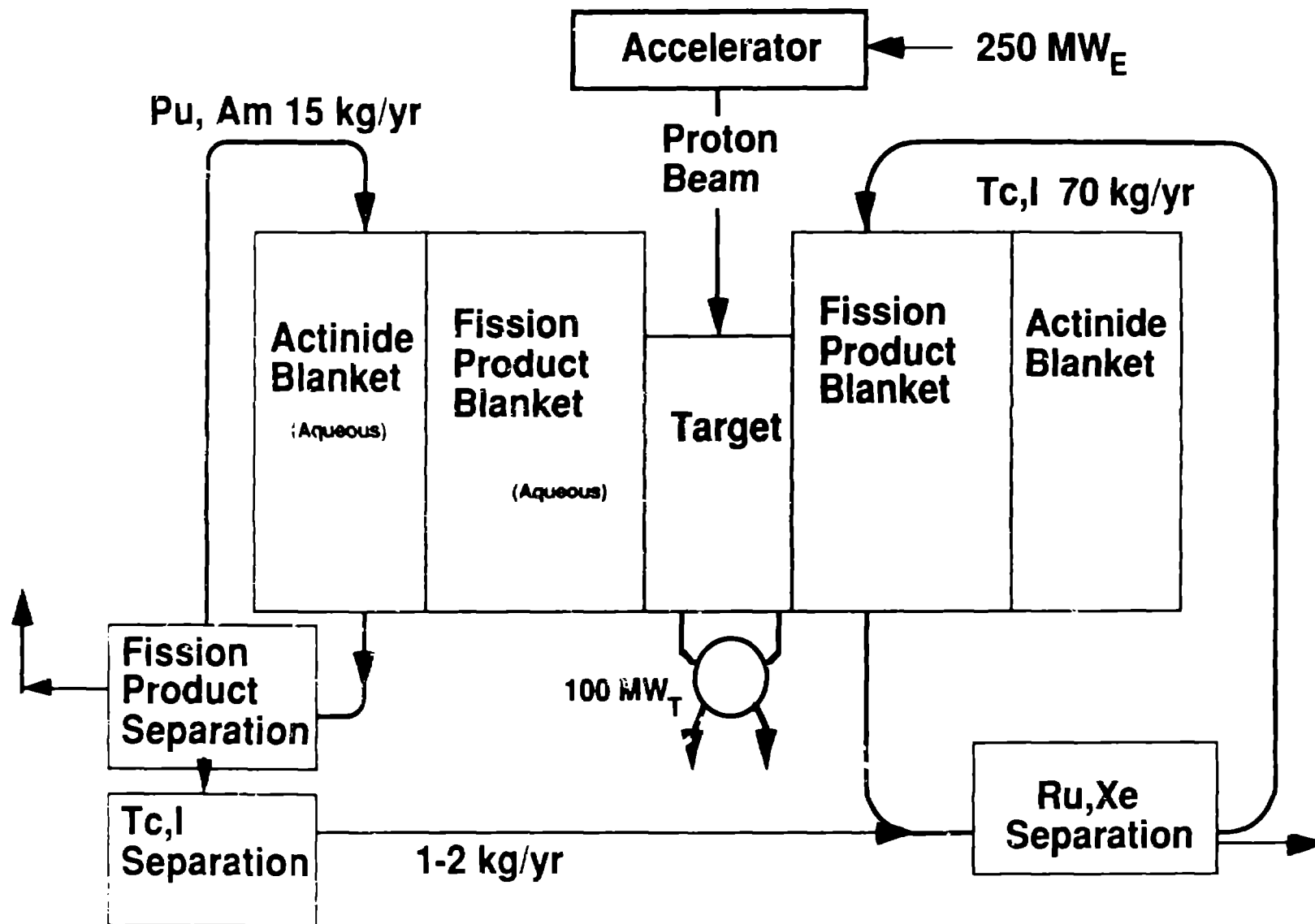
An ATW system capable of handling the Hanford long-lived fission product and higher actinide waste described above is illustrated in Figure 3. An operations scenario of 30 years is postulated resulting in the requirement to transmute approximately 100 kilograms of long-lived radionuclides per year. This requires an accelerator having a beam power of approximately 100 MW for a blanket system that has minimal multiplication. As described in Reference 2 the most efficient beam and current parameters for operation are a beam energy of 800 MeV and an average current of approximately 140 mA. Such an accelerator would require 250 MW_e for operation. A central neutron production target is surrounded by the heavy water blanket described earlier. Neutronics performance of an initial design of a target blanket system are reported in two companion papers presented at this conference.^{3,4} Within the blanket, two aqueous processing loops would be required. The first would be located near the spallation target and would contain technetium or iodine at a concentration of approximately 60 grams/liter. Separations required for this loop are technetium from the stable byproduct ruthenium and stable xenon produced during transmutation of ¹²⁹I. The outer region of the blanket would contain an actinide mixture of plutonium, neptunium, and americium. Material concentrations would be less than 10 grams/liter. Separations required for

this loop are more complex than for the fission product loops described earlier. Here the untransmuted actinides must be separated from fission products produced during actinide burnup. Most of these fission products are short-lived and would be stored on-site for cooling. The long-lived technetium and iodine-129 would be separated from this lump and would be reintroduced into the fission product aqueous portion of the blanket.

Chemical separations play a major role in the overall efficiency of a transmutation system. We have been defining flowsheets for possible separations that could be utilized in an ATW system. Figure 4 illustrates one such example for separation of technetium from ruthenium when an oxide slurry is utilized as the carrier medium in the blanket. Technetium oxide (TcO₂) is introduced into the blanket. After transmutation the flow stream contains a mixture of technetium and ruthenium oxide. A slipstream is taken off from the main loop where this mixture goes into a calciner. There the mixture is heated to about 400^o C producing the volatile Tc₂O₇. Ruthenium dioxide does not volatilize but instead precipitates out and is sent to grout. The Tc₂O₇ is then reduced back to TcO₂ using hydroxylamine and is then reintroduced back into the transmuter. This separations process appears to provide capabilities for high chemical separations (> 0.9999) and produces small waste streams that can be cleaned up and recycled readily.

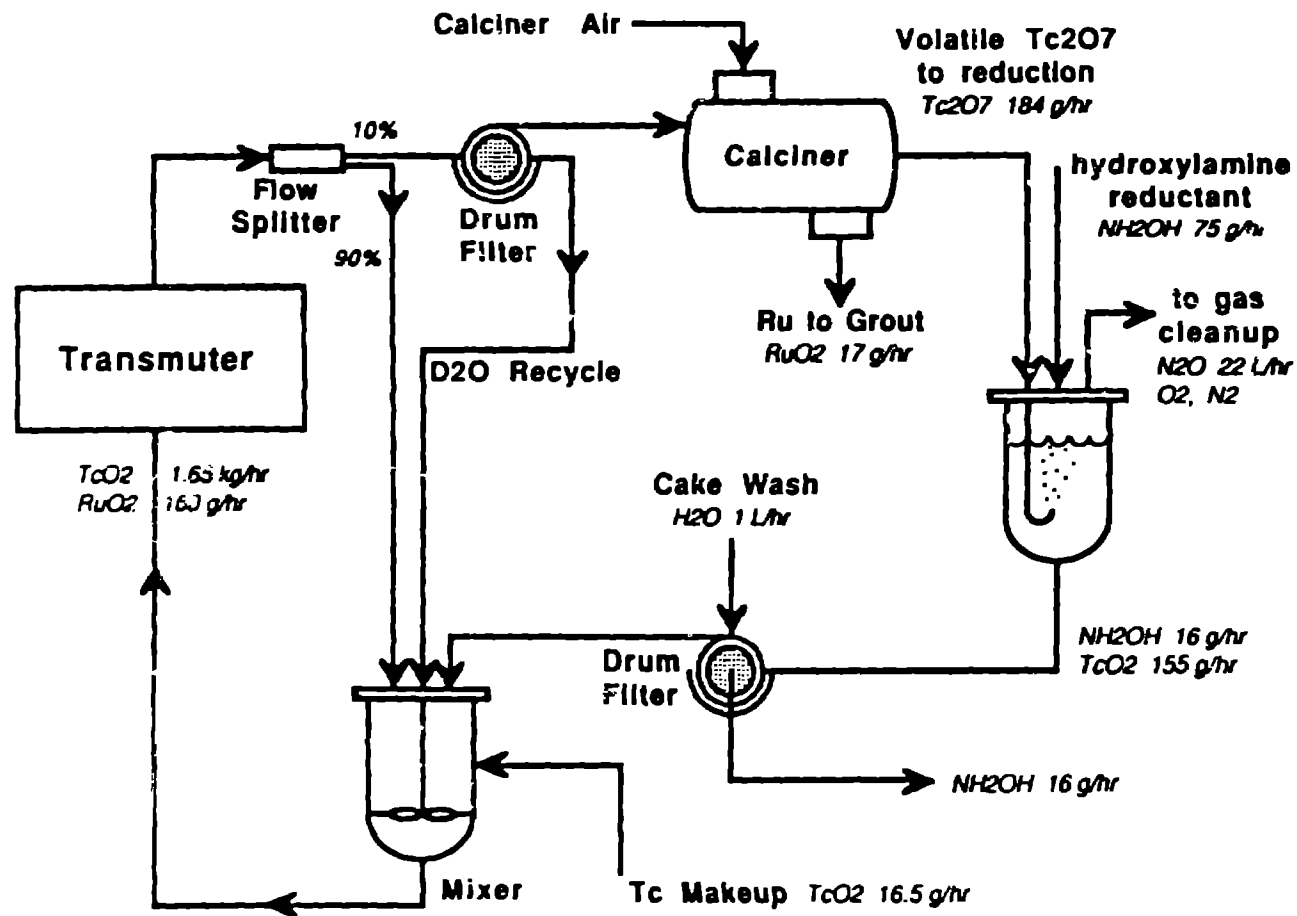
The defense HLW application described here provides a realistic basis for overall concept design and assessment of a transmutation system. Efforts are now underway to define a point design and then assess it in terms of an overall mass balance -- mass of material transmuted during the operational lifetime, mass of long-lived radionuclides produced during operation, untransmuted residues left at end of life, and waste streams resulting from chemical separations. Proof-of-principle experiments are also underway to measure separation factors for processes such as the slurry-based system described above. This technology development and systems analysis obviously have direct impact and extrapolation to possible applications involving transmutation of long-lived radionuclide obtained from reprocessed commercial reactor spent fuel

The second focus area for ATW is described in detail in Reference 1. However a brief summary is provided here for completeness. Zero dimensional analyses of a coupled accelerator multiplying



Slurry-Based Tc Blanket Flowsheet

(10% Slip Stream, hydroxylamine reduction)



blanket system have been made using ATW performance parameters. From such analyses a concept for producing power using natural thorium or uranium has been devised which is illustrated schematically in Figure 5. For such a system the ATW blanket would be divided into three regions. The outermost region would be an aqueous one where, in the case of a thorium-based system, thorium would be converted into protactinium via neutron capture and ^{233}Th decay. The ^{233}Pa would be continuously removed from the system where it would decay to ^{233}U . The features of continuous material flow and processing, coupled with the highly thermal spectrum of the system, could be used to minimize production of ^{232}U . The ^{233}U would be introduced into a molten salt portion of the blanket which would operate at a k_{eff} of approximately 0.9 and would produce 3000 MW_T. Heat extracted from this molten salt loop would be converted to electric power at an efficiency of greater than 40 percent. Of the power produced approximately 20 percent would be required to power the accelerator. The accelerator is described in another contribution to this Conference.⁵ It would consume about 200 MW of electric power, although this figure could be reduced through use of superconducting technology for the accelerating structure. Analyses of this system indicate that enough extra neutrons would be available within the system to transmute the portion of fission products created that have half lives greater than 20 years. Eight such fission products need to be considered for transmutation. These would be introduced into an aqueous inner loop for transmutation. Aqueous chemical processing similar to that described above would be used for the required separations. The waste stream arising from these transmutations could meet surface disposal requirements assuming decontamination factors in the range of 0.999 to 0.9999. In such a thorium-based system higher actinide production would be inconsequential. Thus this system could produce electricity using a long-term fuel source at efficiencies comparable to current LWRs. It would also destroy all components of long-lived, HLW produced during system operation.

SUMMARY

Technological advances provide the basis for a new concept for transmutation of long-lived

radionuclides using an accelerator-driven intense source of thermal neutrons. This source enables conceptual design of a system that can efficiently transmute all long-lived components of HLW and which can do so in the context of a unique low-inventory operating environment. This system is now being analyzed in the context of possible application to transmutation of defense HLW as well as development of an advanced concept for energy production that creates little or no long-term high-level waste stream.

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