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Nuclear Physics Information Needed for Accelerator Driven Transmutation of Nuclear Waste

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Abstract: There is renewed interest in using accelerator driven neutron sources to address the problem of high-level long-lived nuclear waste. Several laboratories have developed systems that may have a significant impact on the future use of nuclear power, adding options for dealing with long-lived actinide wastes and fission products, and for power production. This paper describes a new Los Alamos concept using thermal neutrons and examines the nuclear data requirements.

Introduction

One of the most difficult problems associated with the worldwide use of fission energy is the question of management of the resulting high-level long-lived radioactive waste. That issue has been dealt with in most countries by considering some kind of long-term geologic repository, either by storing spent fuel material directly or with some combination of partitioning and recycling. In the United States, the repository solution has met with considerable public resistance, partly because of the difficulty of certifying container integrity for periods determined by regulatory agencies to be as long as 10,000 years and partly because of public concerns about storing high-level radioactive waste. In addition, the United States has the nearly unique problem of dealing with the accumulation of forty-five years of high-level waste associated with defense material production. At Los Alamos, Brookhaven National Laboratory (BNL), and the Japanese Atomic Energy Research Institute (JAERI), and elsewhere [1,2,3], studies are underway to investigate the role of spallation neutron sources in high-level nuclear waste disposal issues, either as a supplement to geologic storage or to produce energy from the waste. The work at Los Alamos began by considering defense applications; however, the great flexibility provided by an accelerator neutron source has led to the consideration of a highly efficient new energy production concept that has the potential to destroy its own high-level long-lived waste to the extent that surface disposal can be considered after managed storage for a period of time comparable with a human lifetime [4].

The concept of using a high power accelerator to produce fissile material or to transmute radioactive materials is not new [5]. It is the recent development of advanced high-power, high-efficiency accelerator technology which holds the key to the potential success of these systems. All three concepts discussed above use a medium-energy high current accelerator to produce neutrons through spallation. Both the JAERI and BNL designs use fast neutrons to fission actinides. Those systems may be an important component of the overall strategy needed to deal with high-level waste disposal of the high toxicity actinides that comprise the bulk of nuclear waste. They are inefficient when it comes to fission product destruction because there the principal means of transmutation is through neutron capture, a process which has a small cross section for fast neutrons. As shown in Fig. 1, because of their migratory nature, ^{99}Tc, ^{129}I, and ^{135}Cs dominate long-term risk scenarios [6] and need to be addressed as well. The Los Alamos concept discussed here provides a system based on thermal neutron interactions that can efficiently address both types of materials.

System Description

In the Los Alamos concept a high-power medium energy accelerator operating in the 600-1600 MeV range with beam currents from 50-140 mA is used to generate a high flux of neutrons through proton-induced spallation of a lead target. Those neutrons are moderated in surrounding D_2O, and interact with material circulating in piping within the D_2O. A schematic representation of the concept is presented in Fig. 2. The box labelled energy extraction is associated with extraction of energy resulting from fission of actinides or other fuel. To retain high efficiency and to remove stable or short-lived transmutation endproducts, it is necessary to perform a chemical separation either as part of the slipstream or by processing the material in batches. Calculations performed using the Los Alamos High Energy Transport (LAHET) code system show that thermal neutron intensities in the D_2O blanket can reach levels in the range of 10^{15} - 10^{16} n/cm2/s. Those intensities, typically one to two orders of magnitude larger that present in a PWR reactor and extending over a large volume, allow rapid transmutation of materials with small capture cross sections.

A key feature of the Los Alamos system is the generation of high thermal neutron fluxes to destroy actinide waste as well as fission products. Isotopes such as ^{237}Np and ^{241}Am are present in quantity in spent fuel or defense waste and are threshold fissioners; materials that have been traditionally believed impossible to destroy in a thermal system. Our calculations indicate that at sufficiently high thermal fluxes it is possible to efficiently burn those isotopes through a two-step process consisting of a neutron capture leading to a short-lived fissile isotope, followed by a second neutron interaction prior to the decay of the fissile isotope which usually leads to fission. This feature is illustrated in Fig. 3 for the case of ^{237}Np. The upper branch leads to fission of ^{239}Pu, the most probable outcome in a flux typical of that in a thermal reactor system, and requires 3 neutrons with a release of only about 2.7, indicating that the material acts as a net neutron absorber or poison. The lower branch dominates in a high thermal flux and leads directly to fission of ^{238}Np with the use of 2 neutrons but release of 2.7; in that case ^{237}Np is a fuel. To our knowledge this is the first time thermal neutrons have been considered as a viable option for destroying threshold fissioning actinides. These results need confirmation, either experimentally or calculationally after measured differential reaction data are available for the short-lived nuclei. The potential for destruction of both fission and actinide waste coupled with in-situ fissile material production is the feature that makes the Los Alamos thermal system a candidate both for waste transmutation applications and as an advanced energy source with the possibility of destroying its associated long-term high-level waste.

Using the concepts discussed above, together with a system for in-situ fissile material production and utilization, we have developed a system to burn ^{232}Th. Applying methods implemented in the Molten Salt Breeder Reactor to the accelerator system, it appears that one could develop a practical energy producing system which operates far from criticality, has a fissile loading during operation of less than 100 kg, and has a cost and efficiency competitive with reactor systems [4].

Data Requirements

Much of the nuclear data currently needed for the Los Alamos system is available in sufficient accuracy, either through measurement, modelling, or nuclear systematics, to allow an initial positive assessment of the concept. As the design becomes more sophisticated, a considerable amount of new more accurate data will be required. Because of the vast range in both proton and neutron energy encountered in the accelerator and the target-blanket shield, it appears impractical at the present time to develop a comprehensive data library based on evaluated nuclear data measurements such as was done for reactor systems. It may be necessary to rely on an improved suite of nuclear models benchmarked by measurements to provide the necessary information in all but a few critical areas.

It appears that the accelerator can be designed and operated at currents of up to 250 mA with beam losses small enough to allow hands-on maintenance for most of the major components. For regions where beam transitions take place, for accident analysis, and for regions near the neutron production target, it will be important to determine yield data for thick target (p,xn) and (p,xγ) reactions and to carefully chose materials to be sure that induced activities are not unexpectedly high. The issue of shielding data needs associated with accelerators was recently addressed, [7] we note however that at minimum accurate total cross section measurements extending to the maximum neutron energy expected, together with enhanced modeling, will be required to provide needed transport information.

It appears that for the near term, the most important nuclear data needs for confirmation of the Los Alamos concept are for neutron production target yield, for spallation product mass yield, and for capture, fission, and total cross sections of short-lived actinides and fission products.

Verification of the calculated neutron yield, spectrum, and thermal neutron flux in the D_2O blanket resulting from medium energy proton interactions with a massive lead target is needed both to answer questions about overall efficiency, and to be able to accurately address radiation damage problems. We note that neutron leakage from thick targets depends heavily on effects associated with transport. Neutron capture, multiplication in the lead, and absorption in the spallation products which will build up as a poison over time will all be important to investigate as details of the system evolve.

Good resolution measurements of the fission and total cross sections of 16-hr ^{242}Am, 2.1-day ^{238}Np, and 1.3 d-^{232}Pa (produced in the conversion of ^{232}Th to ^{233}U in the energy concept) over the range from thermal to 1000 eV are important to verification of effective burning of higher actinides using low-energy neutrons and to transport calculations. For the fast neutron concepts of JAERI and Brookhaven, fission and capture data need to be extended to the MeV range. At present there are no direct measurements available, although some fission cross section information has been obtained from fission probabilities calculated from charged particle measurements and from integral tests.

Finally, better information is needed for differential neutron total and capture cross sections of fission products with half lives in excess of 11 years. The list of fission products under consideration are given in Table 1. Of those, ^{99}Tc and ^{129}I are most important from a risk standpoint, and each need better information over the range of interest.

Conclusions

Because accelerators provide neutrons in addition to those present when fissionable material is destroyed, it appears possible to achieve the goal of energy production and concurrent waste destruction that can not be reached in a system based on fission neutron economy alone. It will be important, both for a system designed to destroy waste through transmutation alone, and for one using actinide fuel, to improve the existing nuclear data in order to show that such systems are scientifically practical and economically feasible.

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Table 1. Long-Lived Fission Product Transmutation Candidates

Nuclide	Half-Life Yrs	Thermal Cross Section (barns)
^{79}Se	6.5×10^{4}	?(10)
^{90}Sr	2.9×10^{1}	0.9
^{93}Zr	1.5×10^{6}	2.5
^{99}Tc	2.1×10^{5}	20
^{107}Pd	6.5×10^{6}	1.8
^{126}Sn	1.0×10^{5}	0.14
^{129}I	1.6×10^{7}	27
^{135}Cs	3.0×10^{6}	8.7
^{137}Cs	3.1×10^{1}	0.25
^{151}Sm	9.0×10^{1}	152000

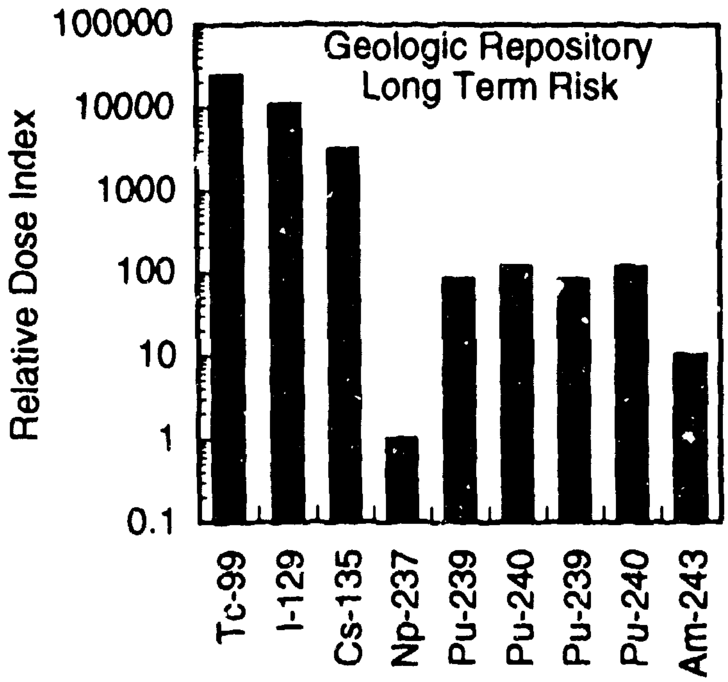

Fig. 1. Relative Radiation Dose Index for Some Fission Products and Actinides during geologic storage. From Ref. [6].

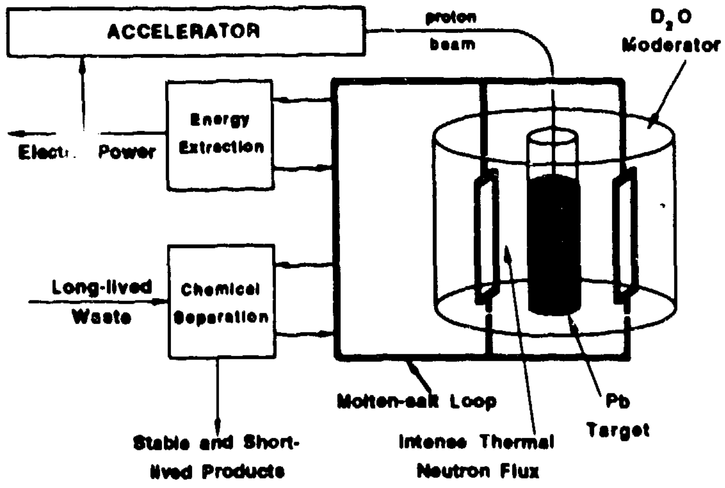

Fig. 2. Schematic diagram illustrating the Los Alamos concept for energy production and waste transmutation. The proton beam generates neutrons from a lead target which are moderated in the heavy water blanket. Molten salt circulating through the blanket carries fissile material for heat generation and power production. Nuclear waste is also circulated through the blanket and transmuted to stable and short-lived nuclides which are then chemically extracted.

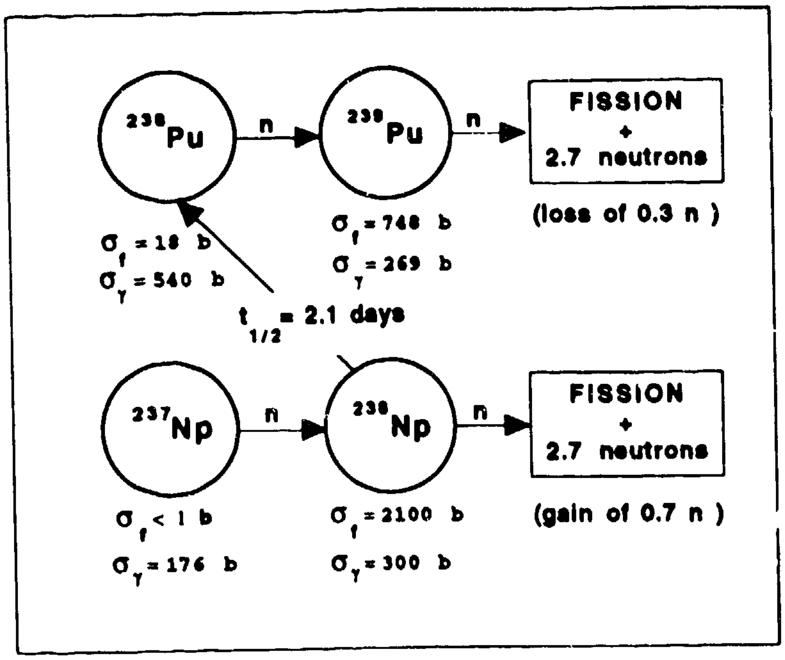

Fig. 3. Diagram illustrating two-step process for thermal burning of threshold fissioning actinides. The upper branch dc:ninates at low fluxes.