NEUTRON-INDUCED FISSION OF WEAPONS PLUTONIUM IN AN ACCELERATOR/TARGET/BLANKET SYSTEM

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ABSTRACT
A proliferation-proof method for the disposition of weapons-grade plutonium using an intermediate-energy proton accelerator is presented in this paper. The method makes use of a spallation neutron source and a subcritical plutonium-loaded blanket assembly surrounding it. The neutron source consists of a heavy water-cooled lead target, bombarded by a uniform-intensity 1 GeV, 100 mA proton beam. Plutonium is loaded in small graphite spheres (beads) which are enclosed in helium-cooled zircaloy pressure tubes. A subcritical configuration of these tubes blankets the spallation neutron source. Uniformly-distributed erbium is used to control reactivity. The system, operating at constant power, is capable of burning 300 kg of plutonium in a six-month period.

INTRODUCTION
Significant quantities of weapons-usable fissile materials, U235, U233 and Pu239 have been produced over the past decades in the U.S. and the former Soviet Union. In the aftermath of the Cold War, large quantities of these materials have become surplus to the national defense needs of the two Cold War superpowers. These excess fissile materials exist in various forms at various locations and pose a danger to the national and international security as they have a potential for nuclear weapons proliferation. Furthermore, because of the potential for safety, environmental and health consequences of these materials, it is important that they be properly managed. These concerns have led the U.S. and Russia to seek policies on the effective management of these materials aimed at their long-term storage and disposition. The principal goal in the management of these materials is to render them inaccessible or convert them into an irreversible form. Either option entails great technical challenges. To address these issues, a set of stringent criteria for the long-term storage and disposition of plutonium have been developed by the U.S. Department of Energy (DOE). These criteria reflect, among other factors, the analytical framework adopted by the National Academy of Sciences (NAS) in its study on the management and disposition of excess weapons plutonium (NAS, 1994). The focus in this paper is on Pu239.

A number of options for the long-term storage and disposition of weapons-grade plutonium have been under consideration by the DOE. These options encompass a broad range of methods. Deep borehole storage, vitrification and immobilization, burning in reactors and accelerators are some of the options initially considered for the long-term storage and disposition of plutonium. In both the reactor and accelerator options, the burning of plutonium can be accomplished through neutron-induced fission of the plutonium nuclei.

The reactor option provides for the mixing of plutonium in its oxide form (PuO2) with the uranium oxide (UO2) to form a mixed oxide (MOX) which is used as fuel in power reactors. In the accelerator option, plutonium is the sole fissile material and, owing to the much higher neutron densities obtained with an intermediate-energy proton accelerator, high burnup rates can be readily achieved. The study described in this paper examines the accelerator option. This option is also referred to as the Accelerator-Based Conversion (ABC) option.

The purpose of this work was to obtain preliminary estimates of selected basic parameters characterizing an accelerator-driven

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target/subcritical plutonium blanket system capable of high plutonium burnup.

SPALLATION NEUTRON SOURCE

The accelerator considered here is an intermediate-energy high-intensity proton beam linear accelerator (LINAC), about one kilometer long, similar to that used in the Accelerator Production of Tritium (APT) preconceptual design (Van Tuyl et al., 1995), capable of producing 1 GeV protons with a current of 100 mA. The narrow proton beam exiting the LINAC enters a section of the beam line known as the beam expander. Here, an array of quadrupole and octupole magnets is used to expand the beam uniformly in two dimensions, (x-y), creating a square "footprint" of 1.4 m x 1.4 m over the required area of the lead target system. The lead target system consists of an array of aluminum-clad, heavy-water cooled lead pins enclosed in aluminum pressure tubes. Fast neutrons are produced by the spallation/evaporation reactions resulting from the 1 GeV proton beam striking lead nuclei. These neutrons, having an energy distribution similar to that of a U-235 fission spectrum, but with a high-energy component, are moderated somewhat in the heavy water. A large fraction of these neutrons enters the plutonium blanket region, surrounding the lead target, causing fission of plutonium nuclei. Plutonium blanket assemblies surround laterally the neutron source as shown in Figure 1.

PLUTONIUM BLANKET

The design of the plutonium blanket assemblies considered in this analysis has been based on the fuel element design used in the Particle Bed Reactor (PBR) program (Ludewig, 1993). Figure 2 shows a cross section of the plutonium fuel assembly. While the fuel used in the PBR fuel element design was uranium carbide, the graphite spheres (beads), in the annular region of the plutonium blanket assemblies considered in this study, contain plutonium carbide. The active length of the cylindrical fuel element is 190 cm.

The annular fuel region contains randomly packed plutonium-bearing HTGR-type, coated graphite spheres (beads), 0.5 cm in diameter. This HTGR-type fuel is known for its fission product retention capability, high burnup potential and its performance at high temperatures. The annular fuel region is bounded by an inner hot frit and an outer cold frit. Helium gas, acting as coolant, is passed from the cold frit through the plutonium-loaded graphite beads to the hot frit. Small holes through the frits allow passage of helium from the inlet to the outlet flow regions. The entire cylindrical element is enclosed in a zircaloy pressure tube.

The subcritical plutonium fuel blanket configuration selected for this analysis consisted of 57 plutonium assemblies surrounding the neutron source, as shown in Figure 3. The initial plutonium loading in the graphite beads was taken as 0.1 g/cm². The total amount of plutonium in the blanket assemblies was 300 Kg.

NEUTRONIC ANALYSIS

As stated above, fast neutrons are produced by the 1 GeV protons striking the lead nuclei. The LAHET/MCNP (Prael, 1989) code system was used to calculate neutron transport from the lead target to the plutonium blanket. LAHET is a modified version of HETC (RSIC, 1977), the intranuclear cascade code, for evaluations above 20 MeV and MCNP, the widely used Monte Carlo transport code, for calculations from 20 MeV down to thermal energies. Both codes accept the same specification of the problem geometry, allowing an explicit description of the physical system under study. The 1 GeV proton beam with a current of 100 mA leads to a power in the target of ~1700 MW and a power density of ~0.2 MW/l. Extensive studies on the PBR, designed for much higher power densities, demonstrate the thermal/hydraulic feasibility of the system.

MCNP criticality calculations have also been performed for the 57-assembly configuration using ENDF/B-V data libraries and yielding an effective multiplication constant of 0.93. The subcritical condition of the blanket assembly constitutes an important safety feature of the system.

Assuming that the neutron source region and the blanket region are each homogeneous, and using a 30-group ENDF/B-VI cross section library, neutron spectra were determined in each of these regions with the ANISN code (Engel, 1967). These calculations showed that the spectrum in the plutonium blanket region is very similar to the spectrum in the core of the Fast Flux Test Facility (FFTF).

PLUTONIUM DEPLETION

Assuming a homogeneous medium in the plutonium blanket area, using one-group cross sections averaged over the FFTF spectrum and simple isotopic depletion, the burnup of Pu-239 in this blanket was estimated. Depending on the degree of purity of the plutonium, in addition to Pu-239, there could be other fissile isotopes in the fuel element, with the most common being Pu-238, Pu-240, Pu-241, Pu-242 and Am-241. For purposes of this analysis, it was assumed that the only fissile material present in the plutonium assemblies was Pu-239. A uniform distribution of erbium was assumed throughout the fuel region in order to accommodate the initial core loading of Pu-239, and yield a roughly constant reactivity with Pu-239 burnup.

Figure 4 shows the Pu-239 atom ratio, N(t) / N(t=0), as a function of time for three initial fluxes: 1.0x10¹⁴, 2.0x10¹⁴ and 3.0x10¹⁴ n/s/cm²/sec. It can be seen that starting with an initial flux of 3.0x10¹⁴ n/s/cm²/sec, and an assembly configuration shown in Figure 2, up to ~ 300 Kg of Pu-239 may be burned in approximately 180 days, assuming constant power during the burnup process. It should be pointed out, however, that since with an accelerator of the type described above, the proton beam current, and hence the resulting neutron flux, may only be increased by ~30%, the increase in the flux required to meet the constant-power assumption could be achieved by using alternate mechanisms such as, for example, removing poisons, reducing
neutron leakage and, in general, by any means aimed at increasing the subcritical \( k_{enr} \) while the latter does not exceed a value of \( \sim 0.95 \).

Independently from the above analysis, a constant power burnup calculation for the same subcritical blanket (300 Kg Pu-239) was carried out using the ORIGEN2.1 code (RSIC, 1991). The plutonium atom ratio during the depletion is also shown in Figure 3. The ORIGEN2.1 results, are also based on an initial flux of approximately \( 3.0 \times 10^{14} \) n/cm²/sec. These results show good agreement between the two calculations.

CONCLUSIONS

A proliferation-proof method of disposing of weapons-usable plutonium, using an accelerator-driven neutron source and a plutonium-loaded subcritical assembly, has been described. The accelerator considered in this method is based on the APT technology which is currently under development and which affords generation of intense proton beams and hence neutron fluxes of at least one order of magnitude higher than those achieved in the highest flux reactors available today. Preliminary analyses have shown that with a system, similar to that described above, it is possible, under various scenarios, to burn up to 300 Kg of weapons plutonium in a period of six months.

REFERENCES


BNL Accelerator Target for Pu Disposition

Neutron Source:
- Aluminum Pressure Tubes
- Lead Pins with Alum Cladding
- D2O Cooled and Moderated

Beam Expansion Chamber Head (Window)

Beam Expansion Chamber
(Vacuum = 10^{-5} torr)

LINAC Protons
100 mA at 1 GeV

Graphite Reflectors

Subcritical Blanket:
- Zircalloy Elements
  (Alternate is Inconel)
- Pu in Porous Graphite Pebble Bed
  Helium Cooled

2-Dimensional Beam Expander
Basis: BNL REF

Figure 1
PLUTONIUM FUEL ELEMENT

Top Spacer
Compression Spring
Top Spacer Positioning Spring
Flow Area

Section Z-Z

Discharge Port

Static Seal
Element Loading Port
Sliding Seal (Wiper Rings)
Top Spacers
30 cm DIA.

Cold Frit
Hot Frit
Pressure Tube
Bottom Spacers
Pressure Tube
Loading Port
Bottom Spacer
Lead Screw
Roller Nut
Lead Screw

Figure 2
BNL Accelerator Target for Pu Disposition

Graphite Reflectors

He-Cooled Pu in Graphite

D2O-Cooled Lead

1 GeV Protons

Figure 3
DEPLETION OF PLUTONIUM ATOMS
Constant Power Assumed in Pu Burning

![Plot showing depletion of plutonium atoms over time with different initial fluxes.](image)

- Init. Flux = 1.0E16
- Init. Flux = 2.0E16
- Init. Flux = 3.0E16
- ORIGEN2

Figure 4