PROGRESS IN ACCELERATOR DRIVEN

TRANSMUTATION TECHNOLOGIES

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Abstract

For several years Los Alamos personnel have been developing applications of accelerator-driven spallation neutrons. These include the Accelerator Transmutation of Waste (ATW) and Accelerator Based Conversion of Plutonium (ABC) as well as the generation of electric power using Thorium as a fuel. In these applications, the spallation neutrons drive a subcritical blanket assembly containing actinides and fission products to be transmuted This paper is a report on the status of this work, and will describe political and technical motivation for the technology choices as well as high current accelerator, spallation target, and blanket development. The talk will include a description of the approach for separation chemistry and the nature of the final waste stream.

Introduction

The concept of using neutrons from accelerators to transmute nuclear waste is several decades old. However, the advances in accelerator and separations technology that could bring this concept into reality have occurred only recently. In particular, the Los Alamos Neutral Particle Beam Program under the Strategic Defense Initiative has developed low energy components for proton linear accelerators that make feasible medium energy (800 -1600 MeV) proton beams with currents of 200 mA and higher. Furthermore, experiment has shown that at 1000 MeV, each proton striking a high-Z target can produce 20 or more neutrons through spallations processes. A 200 mA beam, therefore, could produce 2.4 x 10^{19} neutrons/second, or more than 3 moles of neutrons/day. This fluence could be used, for example, to drive a subcritical blanket containing fissile material to a power level of over 6 GW (thermal) with a neutron multiplication of only 20. This multiplication corresponds to a k_{eff} of only 0.95, equal to the US standard for the storage of spent reactor fuel.

Clearly, numerous applications are possible for Accelerator Driven Transmutation Technologies, which is the program name for this effort at Los Alamos. This paper will focus on three: plutonium destruction, high level waste transmutation, and, with thorium fuel, electric power production through the next millennium. The first two applications are referred to as Accelerator Based Conversion (ABC) and Accelerator Transmutation of waste.

Plutonium Destruction

With the end of the Cold War has come growing interest in methods for dealing with weapons plutonium (W-Pu) in the US as well as in the former Soviet Union (FSU).

Terms like non-proliferation and proliferation resistance are used frequently. Near term effort must focus on issues such as dismantling weapons, securing the plutonium, and near term storage. In the long term, some appropriate method for ultimate disposition must be found. The fact that different countries view their W-PU differently adds political confusion. For example, at this time the US tends to view surplus W-PU as a troublesome waste, while Russia views her surplus W-PU as a national treasure.

However, W-PU represents only the tip of the iceberg. DOE Secretary O'Leary announced publicly that the US stockpile contains some 102 metric tons of Pu. If we assume that the FSU has a similar quantity, this total is still small compared to the plutonium in spent reactor fuel (SF), which is estimated globally to be 930 tons as of December, 1993, and is growing by 50-75 tons annually. Furthermore, it is known that, unlike uranium, plutonium in any isotopic mixture has a fairly small critical mass and can be used to make at least a rudimentary nuclear explosive. Plutonium that is useful in a weapon can therefore be obtained from reactor SF with only chemical separation, whose processes are widely known and straightforward. Uranium, because the isotope U-238 is not fissile, can be "denatured" so that isotopic separation and enrichment is required in order to obtain fuel for a nuclear explosive. Therefore, uranium can be easily put into a proliferation resistant form.

Given the relative ease with which plutonium can be obtained from spent fuel, effort should focus on rendering all plutonium proliferation resistant, and not focus only on W-Pu. This philosophy is reflected in recent reports from RAND and the National Academy of Sciences, who recommend that W-Pu, in the near term, need only be put in some form that makes it as hard to obtain as the Pu in SF. Their recommendation is to mix the W-PU with high level waste (HLW) and vitrify, or fabricate a mixed-oxide (MOX) fuel and bum through one cycle in a reactor. Indeed, although the ABC approach is capable of complete burnup of plutonium, we have recommended developing ABC only in the context of addressing a strategy to deal with all plutonium. This subject also is political in nature, as a number of nations are developing methods and plants for separation and use of Pu from commercial SF, while the US has a long standing policy against reprocessing commercial SF.

ABC

Given this philosophy, in the context of dealing with all plutonium, transmuting W-Pu is an excellent first step as it is technically more straightforward--no removal of cladding, uranium, or fission products is required for W-Pu. We have developed the ABC concept as an approach that provides a proliferation free product--all Pu is fissioned--as well as a proliferation resistant approach. In ABC, a “cauldron” or blanket containing graphite moderator is fueled with a lithium fluoride-beryllium fluoride molten salt carrier to which Pu is added. We have modeled a single module blanket producing 500 MW of thermal power, for which the initial Pu inventory is only 20 kg. The blanket assembly is driven with a 15-18 mA beam of 1 GeV protons that produce neutrons through spallation from a molten lead target inside the blanket. The graphite moderator slows both spallation and fission neutrons, providing a near-thermal spectrum. Neutron multiplication in the plutonium fuel is about a factor of 20 (k_{eff} = 0.96). This multiplication is well subcritical; the standard for storage of spent fuel is k_{eff} = 0.95.

As the Pu is burned out of the fuel, new Pu is added to replace it. In addition, the build up of fission products (FP) requires further addition of Pu to maintain reactivity. However, the fluid fuel allows the removal of more than half the fission products “naturally”. The noble gases are volatile, and the noble metals can be plated out on a cool surface or on an electrode. Yet, no plutonium is removed during a cycle. A cycle is concluded after about 10 years of operation as the concentration of Pu and FP approaches saturation in the molten salt. At this time, the Pu inventory has grown to about 230 kg, and a total of 2.3 metric tons of Pu has been fed into the blanket. All isotopes of Pu have been reduced by a factor of 10, and the Pu-239 isotope by a factor of 50. It appears that, with the addition of some highly enriched uranium (HEU), a second cycle can be initiated that will burn out the remaining Pu. With solid fuel there is no FP removal, limiting the maximum Pu reduction in a single cycle to about a factor of 3. Therefore, a solid fuel will require at least twice a many cycles to accomplish the same completeness of Pu destruction.

The rate of transmutation allows each 500 MWT module to keep up with the production of Pu and higher actinides in a 1 GWE light water reactor. A transmutation park is envisioned in which a single accelerator drives four to six blanket modules. Thermal energy would be used to power a steam plant for generation of electric power. Ten to *twenty per* cent of the electric power would be required to operate the accelerator, and the remainder would be available for commercial or other use.

Why is the accelerator important? The answer is safety and control. Plutonium offers a delayed neutron fraction that is only one-third that of uranium fuel, so that a reactor fueled by Pu only runs much closer to criticality from prompt neutrons than does a uranium fueled reactor. Furthermore, the addition of fertile U-238 to provide a negative temperature coefficient of reactivity results in breeding of Pu-239, which is counterproductive to the goal of complete Pu burnup. In addition, the growth of Pu and FP inventories greatly reduces the leverage or worth of control rods during a cycle, much like the addition of buffers to a solution reduces the impact on pH of the addition of a *small* amount of acid or base. For plutonium burning, the accelerator enables operation that is substantially subcritical, and provides a control mechanism that is independent of the changing inventories and the differences with delayed neutrons.

This molten salt blanket concept is based on extensive experience at Oak Ridge National Laboratory with the Molten Salt Reactor Experiment and with design effort toward the Molten Salt Breeder Reactor. Technical development will require bench chemistry to determine volubility limits in Pu fuel, and additional examination of material compatibility with the salt. Our modeling capability allows us to begin calculation with the interaction of the incident protons with the lead target, and to end with detailed values for the end-of-cycle inventories and activities of all isotopes.

High Level Waste Transmutation--ATW

Pigford has examined the relative importance of various long-lived isotopes in a long term storage system such as the geological repository for commercial spent fuel proposed for Yucca Mountain. He has developed a “relative dose index” based on the product of repository inventory, rate of dissolution, and dose conversion factor for each isotope. These indicate that of the seven fission products with very long lifetimes, Technetium-99, Iodine-129, and Cesium-135 have dose indices some 10,000 times greater than the remaining four long lived fission products (LLFP). (Plutonium-239 and 240 and Americium-241 and 243am the actinides with high dose indices, and these will be removed during ABC transmutation.)

Once again, national policy and politics become ingredients in the development plan, as gaining access to these particular fission products requires their separation from spent fuel. However, several factors make transmutation of these three fission products very attractive. First, if we have embarked upon the path of destroying Pu in SF, the fission products are available for chemical separation. Second, the accelerator provides extra neutrons which can be used to transmute the important fission products through neutron capture followed by beta decay into stable or short-lived isotopes--and these three important fission products have high thermal neutron capture cross sections. Third, since in this scenario, the important actinides will have been destroyed, transmutation of these three fission products results in a major impact on the lifetime requirements of the repository: except for the four remaining isotopes, three of which are noble metals and all of which are only slightly radioactive, the remaining isotopes have half-lives of 30 years and less.

The ATW approach has not yet been worked out in detail. However, modeling has demonstrated that the neutron economy will allow transmutation of the three major FPs at the rates required to keep up with production. This is extremely difficult if not impossible to accomplish with a reactor. For FP transmutation, proliferation resistance of the process is not required, as no weapon can be constructed from these materials. Therefore, continuous processing is possible, and may well be necessary. This could be accomplished by running FP loops through a Pu burning blanket, keeping the fission products isolated from the Pu-bearing fuel. Alternatively, part of the accelerator beam could be used to drive a blanket containing only fission products, again using real time, on line separations technology. In this case, power from the Pu-burning blankets would be used to provide the necessary beam.

Both plutonium/actinide destruction and fission product transmutation would provide waste forms that are consistent with current standards and options. Noble metals would preferably be stored as ingots, perhaps along with ingots of the zirconium cladding from SF. Uranium from spent fuel can be recycled; its enrichment of 0.9% is slightly greater than that for natural uranium (0.72%), and mining is unnecessary. Cesium-135 transmutation will require that Cs be isotonically separated--the only element for which this is necessary. This is because stable Cs-133 will breed Cs- 135, and 30-year Cs- 137, with its high neutron capture cross section, would harm the neutron economy. The separated Cs- 137, a hard gamma ray emitter, can be contained very will as a silicotitanate, using a process developed by Sandia National Laboratories. Other fission products can be converted to oxides for vitrification.

Energy Generation with Thorium Fuel

Natural thorium is 100% Th-232, which will breed U-233, through neutron capture followed by two beta decays in a thermal neutron environment. U-233 itself is fissile, and can make an excellent fuel. The advantages to this approach are that thoriumis plentiful in the earth's crust; any unburned U-233 can be "denatured" with U-238 so that it cannot be used for a nuclear weapon, and no U-238 is present in the neutron flux to breed Pu. Furthermore, using thorium in an ABC/ATW-like system can result in a relatively clean waste stream that contains few, if any, long lived fission products. Surplus W-PU or HEU could be used to supply neutrons to initiate the U-233 breeding, therefore providing a value added for these Cold War relics that exceeds the value to be obtained by burning them only for their energy.

Although this was one of the initial ADTT concepts, it for many reasons will probably be the last to be adapted. However, given the vastly increasing global energy needs forecast for the next half century, mostly due to emerging nations, energy generation could well be the ADTT application that has the most significant impact on society and the biosphere.

Conclusion

Accelerator Driven Transmutation Technologies include a number of applications that can play significant roles overcoming decades. First, the ABC approach can destroy plutonium with a completeness and proliferation resistance well exceeding those of other proposed methods. Second, ATW transmutation of fission products in commercial spent fuel, coupled with SF Pu and actinide destruction with ABC, can have a major impact on waste storage scenarios. The volume and mass to be stored is greatly reduced, the proliferation attractiveness is nonexistent, and the storage period requirement becomes several human lifetimes, rather than geological times. Finally, energy production with thorium fuel could become the environmentally clean source necessary to meet growing global requirements.