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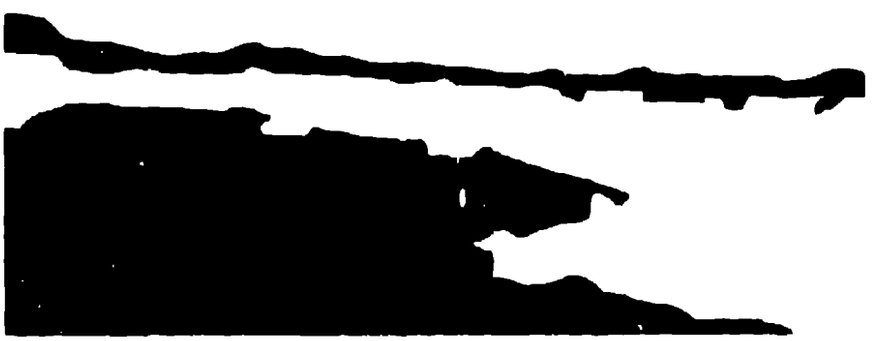

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HIGH-VALUE USE OF WEAPONS-PLUTONIUM BY BURNING IN MOLTEN SALT ACCELERATOR-DRIVEN SUBCRITICAL SYSTEMS OR REACTORS

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ABSTRACT

The application of thermal-spectrum molten-salt reactors and accelerator-driven subcritical systems to the destruction of weapons-return plutonium is considered from the perspective of deriving the maximum societal benefit. The enhancement of electric power production from burning the fertile fuel ^{232}Th with the plutonium is evaluated. Also the enhancement of destruction of the accumulated waste from commercial nuclear reactors is considered using the neutron-rich weapons plutonium. Most cases examined include the concurrent transmutation of the long-lived actinide and fission product waste (^{99}Tc, ^{129}I, ^{135}Cs, ^{126}Sn and ^{79}Se).

All of the fission systems considered are sized at 3000 MW thermal and are assumed to burn 1200 kg of actinide per year to produce this power. The annual waste actinide production from commercial power reactors is assumed to be 300 kg. The following options for burning 100 tonnes[1] of weapons plutonium in 30 years emerge from this study; (1) build ten reactors which derive 1/4 of their power from weapons plutonium and 3/4 from fission of thorium, (2) build four reactors which burn the weapons plutonium mixed with the actinide waste from six commercial nuclear power reactors, (3) build nine sub-critical accelerator-driven systems which burn the weapons plutonium and transmute the waste from 25 commercial nuclear power reactors, and (4) replace all existing LWRs at the end of their life with accelerator driven systems which destroy the plutonium and all long-lived LWR fission product waste. Options (1) and (3) were found to be the most attractive. Option (1) provides the greatest energy benefit from the plutonium. Option (3) makes use of the plutonium (and the high enrichment uranium) to destroy all the higher actinide and long lived fission product waste from U.S. commercial power reactors. The technology described here also can be used for production of "unlimited" nuclear energy from thorium in a subcritical system with the concurrent destruction of waste so that the objectives of fusion technology development, (1) "unlimited power", (2) subcritical system, and (3) no long-term high-level waste can be achieved with straightforward extensions of fission technology.

INTRODUCTION

The Los Alamos National Laboratory has been studying for several years[2] accelerator-driven subcritical systems for commercial nuclear waste transmutation and nuclear energy generation from thorium with concurrent transmutation of the long lived waste actinide and fission product. The accelerator performs the vital function of supplying the excess neutrons to the nuclear system necessary for transmutation of the waste. In reactors the neutron economy is not adequate for this unless enriched ^{235}U or ^{239}Pu are used to supplement the neutron economy. This is impossible for nuclear power over the long term because it leads to a rapid depletion of the ^{235}U resource or of the ^{239}Pu which is ultimately derived from the ^{235}U. In addition the accelerator allows the system to operate at high power in a subcritical mode which eliminates the nuclear run-away accident most feared by the public. A study of the design parameter space for practical implementation of these accelerator-driven systems has identified the molten salt system studied[3] at the Oak Ridge National Laboratory as the most practical means of deploying this technology.

In the meantime the disposition of weapons-return plutonium has become an issue of international concern as the dismantlement of warheads in the FSU and in the U.S. has begun to be seriously considered. Concerns about weapons plutonium have also led to the beginnings of a reevaluation of the danger of the plutonium in commercial spent fuel. The amount of plutonium in spent fuel in the world is much larger than the amount of weapons plutonium. If a dangerous nuclear weapon could be produced with 20 kg of commercial plutonium, the present world-wide inventory would allow the construction of 75,000 of these nuclear weapons. In the 15-20 years before repository storage or other means for handling the waste is developed, the number of possible weapons would grow to 150,000. The storage of this material in geologic repositories does not eliminate the possibility that this material might be removed for malicious purposes during the millions of years that this material or its daughters possess the features allowing the construction of nuclear weapons. A recent French study[4] has highlighted this concern and has urged the expenditure of as much effort on the study of means for destruction of the plutonium and other reactor spent fuel actinides higher actinides as exists now in studies in that country on the geologic storage of spent fuel.

If the actinide were destroyed, the need for geologic storage of this material would be eliminated were it not for the long-lived fission product which remains. Destruction of this component of the waste also, if done to the level of a 0.1% or smaller remnant, would further greatly reduce the danger from storage. A nation need no longer search out the single very best site in its geology, study it with great care, spend large sums to outfit it, and then haul this material perhaps thousands of miles to this single best site. With the reduction of the radioactive species by a factor of 1000 or more, the material might be made sufficiently benign that it can be placed in near surface storage in many safe places. Therefore, for example, in the U.S. a site could be located in almost every state. Interstate transfer of the nuclear waste would become unnecessary and the federal government might get out of the siting and development of geologic repositories and interstate waste transport, falling back to the more appropriate role of licensing. The nuclear power industry would receive an enormous boost from the transmutation of commercial waste to the point where low

level "local" rather than high-level "central" storage were required.

The aim of the transmutation program described here is to transmute the weapons plutonium, the commercial actinide waste including Np Pu, Am, Cm, etc., and the long-lived fission product sufficiently well to reach the above objective. The cost of transmutation is obviously of great concern in terms of practical deployment; the cost is greatly reduced if the energy produced by fission of the actinides can be captured and transformed to electric power with a high thermal-to-electric efficiency. The implementation of the accelerator-driven molten salt technology was chosen because it offers the prospect of a technology which is both practical and cost effective. The economics of the systems is greatly improved in most scenarios by the inclusion of thorium for the fission energy that can be derived from the Th-U cycle. A brief description of this system follows. The performance of this system is then summarized in a single figure for the full range of deployment options. Four of the most favorable deployment options are then identified and discussed.

TRANSMUTATION TECHNOLOGY DESCRIPTION

The transmutation concept described here referred to generically as accelerator transmutation of waste (ATW) operates by providing an intense thermal neutron flux to burn fissile material and to transmute both fission product and any derivative actinide materials rapidly with enhanced safety owing to subcriticality and a low inventory. When a medium energy intense beam (50-100 mA average at 500 to 1000 MeV) of protons or deuterons interacts with a spallation target, about 25 fast neutrons are produced for each proton. A surrounding graphite blanket slows the neutrons down to thermal energies and they induce fission in the "fuel" that is circulated through the blanket. The system design is required to have no possibility of a self-sustained chain reaction. Fluxes as high as 5×10^{15} n/cm^2-s can be achieved in the blanket volume with a neutron multiplication of 20 or equivalently with an effective reactivity factor of $k_{eff} = 0.95$. Because of the extra power to operate the accelerator, which might be as much as 20 % of the electrical power generated in these systems, the power conversion efficiencies must be made as large as possible. Molten salt fuel systems can operate at high temperatures without degradation of the flowing material using structural materials with demonstrated very low corrosion rates, and with the valuable safety and engineering feature of very low vapor pressure. As a consequence, an exceedingly valuable feature of the salt is the high thermal-to-electric conversion efficiency of about 40 % which allows the same net electric power output as a lower efficiency conventional LWR operating at the same fission power in spite of the power drain for operating the accelerator. The molten fluoride salt LiF-BeF_2 used in the Oak Ridge Molten Salt Reactor Experiment[3] (MSRE) that operated for five years in the late 1960's appears to be the best candidate carrier salt for the actinide and fission product fluoride salts.

The most promising design concept uses a molten LiF-BeF_2 salt target surrounded by a uranium layer for conversion of neutron kinetic energy into additional neutrons. This neutron generation part of the assembly is surrounded by a graphite moderated multiplying blanket . The system is shown schematically in Figure 1. The salt-uranium target allows efficient neutron production and low parasitic neutron absorption because of its small size and the presence of the

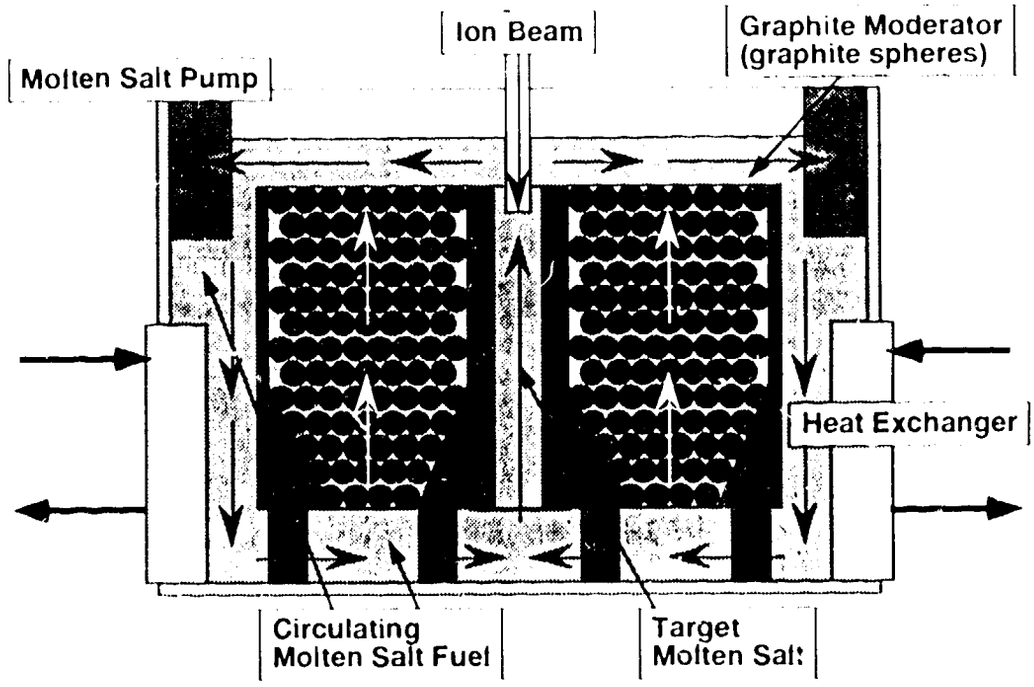

Figure 1. *Functional layout of the molten salt ATW blanket target assembly*

surrounding fission product transmutation region. Since the proton or deuteron beam interacts mainly with the LiF BeF$_2$ salt, the target produces few waste products.

The same molten LiF BeF$_2$ that constitutes the target salt also carries the plutonium and the thorium fuels as fluorides. The molten salt circulates through a bed of graphite spheres which constitute the neutron moderator and into intermediate (salt to salt) heat exchangers which are submerged in the tank containing the salt. Thorium breeding and the bulk of the power generation occur within this salt which is therefore at the same time target, fuel, and heat exchanging fluid. The fission products, isolated for transmutation through processing of a small slip stream, are placed in special high flux regions just outside the main portion of the target.

Once the fuel is in the blanket, the only process that has to be performed more or less continuously during power production is the separation of fission products from the fuel. At high neutron flux, which is associated with low inventory, fission products must be separated rapidly to avoid deterioration of the fuel's reactivity. Continuous on line processing of the liquid fuel makes this possible. Separation processes are desired that avoid external waste streams (solvents, catalysts, and reagents) which could become part of the radioactive waste stream. Physical chemistry processes are favored, which might perform these separations by fractional evaporation, precipitation, electrolysis, solubility and centrifugation[6]. Volatile fission product fluorides would be separated from the fuel using helium sparging

SYSTEM PERFORMANCE

A design of this system with realistic dimensions, power densities, liquid flow rates and heat transfer rates has been completed using materials compatible with the molten salt as established by the MSRE at Oak Ridge. The performance of this system has been calculated by a Monte Carlo system allowing arbitrary geometry and energy-differential cross sections and neutron flux so that resonance structure can be taken into account in detail. The code Los Alamos High Energy Transport (LAHET) has been used to transport the protons and to calculate conversion to neutrons. This code is coupled to the code MCNP which allows the detailed calculation of the neutronic performance of the system. An initial loading of material is followed by successive Monte Carlo calculations through changes in composition (as the higher actinides build-up) and through the accompanying changes in the neutron spectral shape until equilibrium is reached. The calculations take into account the flow, by neutron capture and by decay, though 33 nuclides between ^{232}Th and ^{252}Cf. Because the liquid fuel allows constant removal and replenishment of the salt constituents, a true equilibrium may be established in contrast to a the situation with solid fueled systems. The usual properties of the system are calculated including k_{eff}, temperature coefficients, etc.

It is well known that the Th-U cycle is superior to the U-Pu cycle for thermal spectrum systems and this is confirmed by these calculations. Moreover study of this and related designs shows that their neutronic and overall performance can be enhanced with the inclusion of thorium in the system and the generation of the major portion of energy from the Th-U cycle rather than from the fission of plutonium or other higher actinide wastes. The performance of these systems is summarized succinctly in Figure 2.

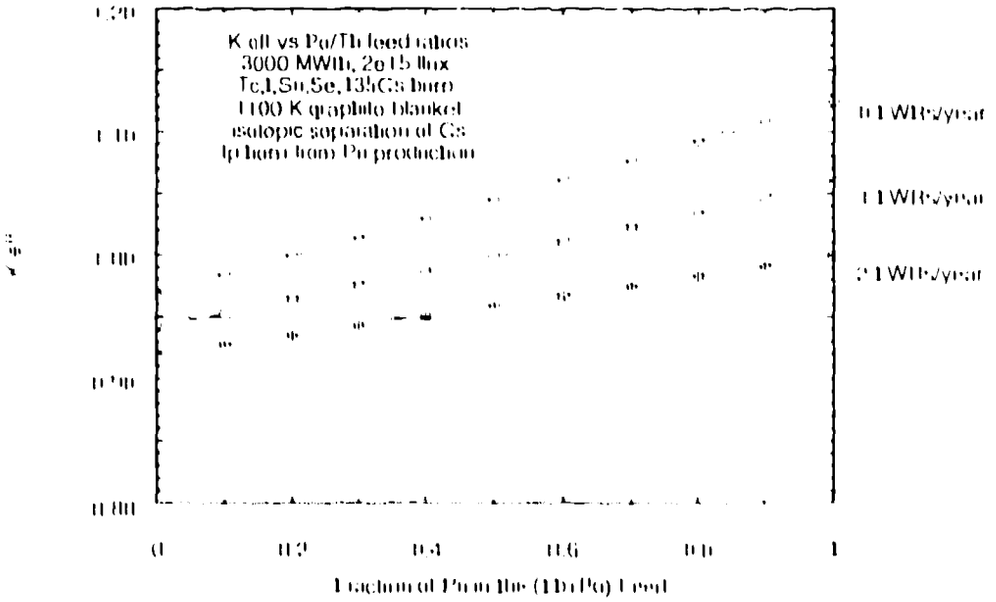

Figure 2 *Performance of various accelerator driven plutonium burners*

Figure 2 shows k_{eff} vs the fraction of Pu in a (Pu+Th) feed to a system operating at a power of 3000 MWt which may be burning commercial waste. All of the actinide fed to the system from any source is fissioned and the fission products Tc, I, Sn, Se, and ^{135}Cs are transmuted so that engineered near-surface storage of the remnant waste is practical. The three curves with data points show the performance under the conditions of burning, along with weapons-return plutonium, the plutonium, higher actinide and fission product waste from commercial spent fuel. The curves refer to the transmutation of the waste in this system operating at 3000 MWt from two, one, or zero LWRs which also operate at 3000 MWt. It is assumed that a 3000-MWt system of any type fissions 1200 kg of actinide per year. It is further assumed that a 3000-MWt LWR produces a total of 300 kg per year of Np, Pu, Am, Cm, etc. The abscissa is the fraction of weapons plutonium in the thorium plus weapons plutonium feed. The ordinate is k_{eff}; a horizontal line is drawn at $k_{eff} = 0.95$ which is probably a practical operating value for the ATW system. It allows a high neutron multiplication and yet is far enough from unity to guarantee that the system never can become critical. We illustrate the use of this figure with two examples.

Example 1

If we wish to operate at a $k_{eff} = 0.95$, and to burn the weapons-return plutonium along with the waste produced per year from two LWRs, we identify the $k_{eff} = 0.95$ line and move across until it intersects the two LWRs/year curve. We find there will be about 40% plutonium in the (Th + Pu) feed. For a 3000-MWt system, 600 kg of the 1200 kg of actinide fissioned will come from the two LWRs (300 kg X 2 = 600 kg). Therefore the remaining fission will be apportioned between the weapons plutonium, which we hereafter may refer to alternatively as ^{239}Pu, and thorium. Of course the thorium is actually burned primarily as ^{233}U. Therefore the amount of ^{239}Pu burned is 600 Kg x 0.4 = 240 kg/year. If we wished to burn 100 tonnes of ^{239}Pu in 30 years, the number of ATW systems required would be 100/ (0.24 x 30) = 14. During the destruction of this 100 tonnes of ^{239}Pu, the plutonium, higher actinide and fission product from 14 x 2 = 28 LWRs will have been destroyed as well. If one calculates the enrichment of the plutonium in the system when the spent fuel plutonium is mixed with the ^{239}Pu from weapons, it is found to be 76 %. If one takes the best current estimate of the cost of these ATW systems at $2.5 billion each, the total capital invested would be $35 billion. The value of the electric power generated in the destruction of this 100 tonnes of ^{239}Pu would be $147 billion assuming 50 mils per kilowatt hour.

Example 2

The curves include situations where k_{eff} is greater than one and these can be used to calculate the performance as a reactor. A horizontal line at 1.025 would be appropriate for the reactor because about 2.5 % of the reactivity would have to be spent in neutron absorption in the control rods required by the reactor but not by the subcritical system. This example therefore illustrates the use of these curves to estimate the performance of this molten salt system as a reactor. Suppose we wish to destroy ^{239}Pu while concurrently destroying the waste from one LWR per year. Entering the ordinate at 1.025 and moving across to the one LWRs/year curve, we find that the fraction of ^{239}Pu in the (Th+^{239}Pu) feed must be about 70 %. The amount of

actinide burned from one LWR per year is 300 kg so that 1200 - 300 = 900 kg of the Th + ^{239}Pu mixture must be fissioned per year. Since the fraction of ^{239}Pu in this 900 kg mixture is 70 %, the amount of ^{239}Pu burned per year in the reactor is 900 X 0.7 = 630 kg. The fissioning of 100 tonnes of ^{239}Pu in 30 years would require 100/ (0.63 X 30) = 5 of these molten salt reactor systems. The reactor would concurrently destroy the waste from 5 X 1 = 5 LWRs. The enrichment of the plutonium in the reactor taking the isotopic content of the LWR plutonium into account is 90 %.

The performance, as derived from Fig. 2, of the molten salt system as an accelerator-driven system (ATW) and as a reactor are shown in Table 1 for burning up 100 tonnes of weapons-return plutonium in 30 years. The reactor system might not be directly comparable to other reactor systems for plutonium burning because this system burns not only the ^{239}Pu and the higher actinide resulting but also the associated long-range fission product. The general feature of the reactor version of the ATW technology is that fewer transmutation systems are required but that the impact on commercial spent fuel waste is small. Therefore the use of ^{239}Pu in the reactor to burn significant waste from the LWRs concurrently is of little impact. However the accelerator-driven systems are quite effective in destroying both the weapons plutonium and the LWR waste. For example nine of these systems operating at 77 % effective enrichment can burn the waste from 25 LWRs while destroying 100 tonnes of weapons plutonium. The figure of 25 LWRs at 3000 MWt thermal is not far from the amount of deployed nuclear energy in the Former Soviet Union (FSU). A system to destroy the U.S. spent fuel would be four times larger, or would take four times as long. It would also require the use of 300 tonnes of highly enriched yranium (HEU) in addition to the 100 tonnes of ^{239}Pu.

Accelerator-driven Systems (Th+Pu Molten Salt, 3000 MWth, Keff = 0.95)

Plutonium enrichment (%)	Systems required	LWRs handled	Capital invested (G$)	Power value (G$)
74	37	37	92	390
76	14	28	35	147
77	9	25	23	95

Reactor-based Systems (Th+Pu Molten Salt, 3000 MWth, Keff = 1.025)

Plutonium enrichment (%)	Systems required	LWRs handled	Capital invested (G$)	Power value (G$)
88	4	6 [>6 is subcritical]	10	42
90	5	5	13	53
93	6	3	16	64
100	10	0 [burns its own actinides and fp]	25	106

Table 1. The performance of ATW systems and reactors as plutonium burners

Upon examining this Table, four options worth consideration emerge; two associated with the ATW-like reactor and two with the accelerator.

Option 1.

Build ten ATW-like reactors for the primary purpose of destroying 100 tonnes of weapons plutonium.

These systems would involve no commercial reactor spent fuel. They would however destroy all of the higher actinide and long-lived fission product waste produced in destroying the weapons plutonium and in deriving energy from thorium. The fission energy in the ^{239}Pu is in effect multiplied by a factor of four since 1/4 of the power comes from ^{239}Pu and 3/4 from thorium. This increases the societal benefit from the weapons plutonium from the perspective of optimal energy gain by enhancing the energy output, but does not address the commercial spent fuel problem.

Option 2.

Build four ATW-like reactors which burn the waste from 6 LWRs concurrently while destroying 100 tonnes of weapons plutonium.

The burning of the commercial waste adds an additional requirement for neutrons which requires the burning of more ^{239}Pu and therefore fewer systems are required to destroy the ^{239}Pu. However, the impact on commercial waste is negligible and not worth the additional complexity of dealing with commercial spent fuel. If getting rid of the weapons plutonium as fast as possible is the main objective, ^{10}B could be substituted for the commercial waste. However the use of ^{10}B would not optimize the societal benefit of enhanced energy production which comes in a reactor with Option 1. Therefore this option is not attractive either from the perspective of optimal energy output or impact on the commercial waste problem.

Option 3.

Build nine subcritical accelerator-driven systems for burning 100 tonnes of weapons plutonium and LWR waste concurrently.

This option allows not only an attractive multiplication of the energy equivalent to Option 1, but it also allows the concurrent destruction of the waste from 25 LWRs. This option works well for the FSU since by increasing somewhat the amount of weapons plutonium burned or substituting enriched ^{235}U on an almost 1:1 basis for weapons plutonium, all of the LWR waste in that nation could be transmuted to innocuous material. A practical means of deployment would be replace the energy supplied by ten or so older reactors with these ATW systems located in several energy parks.

Option 4.

Replace LWRs as they go out of service with ATWs on a 1:1 basis with the ATWs burning the waste from the retired LWRs and the weapons plutonium and its derivative waste. In this option, 37 ATWs would replace 37 LWRs in burning the 100

tonnes of plutonium. The drawback here is that too many reactors are required for the energy park concept so that these new ATW systems would have to be sited more or less as reactors are presently sited and the plutonium transported to them. If no weapons plutonium were employed in the ATWs, these systems still could burn the existing LWR waste on a 1:1 basis by simply increasing the accelerator current somewhat to make up for the absence of the weapons plutonium. The inclusion of weapons plutonium appears to have little impact on these systems and the complications with it appear not to make it worthwhile.

Therefore of the four options only 1 and 3 deserve further consideration. Both involve the deployment of a relatively small number of systems and they are therefore both compatible with an energy park concept. The reactor of option 1 is the simplest to deploy since no accelerator or LWR spent fuel is involved. It achieves the release of substantially more fission energy than is contained in the weapons plutonium itself, but it does not address the commercial waste problem. The most attractive option to the authors is Option 3 which addresses both the weapons plutonium and the LWR waste effectively. The weapons plutonium substantially reduces the size of the accelerator and the fraction of the power which must be spent in the accelerator apparently providing a maximum societal benefit from the weapons plutonium.

The curves from Fig. 2 also can be used to estimate the commercial value for the plutonium. The slope of these curves is the change in k_{eff} per change in the Pu feed fraction. We know that it takes 55 mA of 800-MeV proton beam to drive a $k_{eff} = 0.95$ system at 3000 MWt, and that it requires half as much to drive it for keff = 0.975. We may then convert the change in k_{eff} into a change in the current requirement. We know from our systems studies that the capital cost of the accelerator is about $10 million per mA at 800 MeV and 50 mA current. If this investment is paid off at a 10 % interest rate over a 30-year lifetime for the accelerator, the total debt retirement cost is actually three times higher at $30 million per mA per 30 years or 1$ million/year—mA for the accelerator. The power cost for the accelerator operating at 45% efficiency is about $ 0.6 million per mA. Neglecting the operating costs, the cost of the beam therefore is $1.6 million/mA-year. Converting the feed fraction to kilograms, we find a value for the plutonium for this system of about $0.25 million/kg. All three of the curves give about the same value because the abscissa is different terms of absolute kilograms of weapons plutonium burned for each case. Bloom et. al. estimate that burning weapons-return plutonium in LWRs or LMRs would generate a beneficial value of $9000 and $56,000 per kilogram respectively.

For the FSU one can imagine that the energy parks, where the weapons plutonium is burned, also should become the sites where the LWR waste is transmuted. These sites, which might be former weapons materials production sites, should also become the sites for placement of the commercial spent fuel. One hundred tonnes of weapons plutonium matches fairly well the amount of LWR spent fuel in the FSU. If both of these materials are to be brought to the same site, the weapons plutonium could be diluted immediately for safe intermediate storage by mixing to achieve a Pu enrichment of the 77 % required by option 3. The material would be decidedly unattractive for clandestine removal until burning were completed about 50 years from now.

The amount of spent fuel is the U.S. is three to four times larger than that in the FSU. While the weapons plutonium could be destroyed in U. S. energy parks

along with about 1/4 of the spent fuel, the remainder would have to be destroyed elsewhere. One option is to replace aged reactors with ATWs on the same site which burn the waste from the retired reactor but not to use weapons plutonium at these sites. We see from Fig. 2 that this is possible if the waste is burned in 30 years by using a system with $k_{eff} = 0.94$. (This value is derived from the intercept of the 1 LWRs/year curve. After the waste from the retired LWR has been destroyed the system can convert over to the derivation of all of its energy from thorium. From the intercept of the 0 LWRs/year curve we find a value of $k_{eff} = 0.97$ during this period. It is worth noting that this latter mode requires the smallest accelerator of any of the systems considered, no front-end chemistry, a simpler blanket configuration, and little higher actinide from thorium burning to contend with. This concept referred to presently as the ATW energy producer is therefore technically the simplest to develop and the easiest to make economically competitive.

With the number of different ^{239}Pu burning scenarios in hand, it is useful to consider other factors in deployment. Since weapons plutonium is perceived to be more dangerous than spent fuel, these systems should be built close to the weapons-return plutonium storage sites rather than the commercial waste generation sites. Clustering them into an energy park would allow the front-end chemistry facilities to serve several systems and thereby reduce costs. However, if there is only one site, probably too much power (9 GWe) would be dumped into one point on the commercial grid. Presumably there should be a few sites with a contingent of two or three plutonium burners for each site totaling nine burners altogether.

SUMMARY

The ATW technology can be deployed for weapons plutonium burning either as an ATW-like reactor which burns only the plutonium with substantial energy multiplication, or it can be deployed for the purpose of concurrent weapons plutonium destruction along with LWR waste destruction and large energy multiplication. Both systems amplify the societal benefit from plutonium over simply fissioning it away in a pure plutonium system. Using the ^{239}Pu for destroying LWR waste would work especially well for the FSU. The existence in the U.S. of a larger amount of LWR waste relative to weapons plutonium may be addressed by using, in addition to the system of Option 3, an original version of the accelerator-driven LWR waste burner which did not incorporate weapons plutonium burning. Or the LWR waste could be destroyed in the same ATW systems by continuing their operation using 300 tonnes of HEU.

Denaturing the weapons-return plutonium by dilution to about 77 % fissile material "enrichment" with spent fuel can be done quickly with a minimum of technology development so that storage of the plutonium at an energy park in an unattractive form for surreptitious removal appears practical. In this form with the fission product and other higher actinide present, it is unusable fuel for LMRs, or LWRs, or even for ATWs operating as reactors. However it is highly viable fuel for the non-aqueous ATW system in which the accelerator not only allows sub-critical operation, but also serves the vital function of supplementing the neutron economy. Nine accelerator-driven systems operating at a power level of 3000 MWt could destroy 100 tonnes of ^{239}Pu in 30 years by this means along with the waste from 25

LWRs. The value of the weapons return plutonium is about five times higher for this system than for other plutonium burning systems. These systems might be clustered into energy parks which are constructed at each weapons storage facility. A very favorable neutron economy and a high thermal-to-electric conversion efficiency are required from any competitive system to reach the performance reported here. The technology also could be deployed as ATW-like reactors which do not deal with LWR waste. This is less desirable but it still produces greater societal benefit than any of the other reactor systems proposed.

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