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TRANSMUTATION OF HIGH-LEVEL RADIOACTIVE WASTE BY A CHARGED PARTICLE ACCELERATOR*

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ABSTRACT

Transmutation of minor actinides and fission products using a proton accelerators has many advantages over a transmutor operated in a critical condition. The energy required for this transmutation can be reduced by multiplying the spallation neutrons in a subcritical assembly surrounding the spallation target. We have studied the relation between the energy requirements and the multiplication factor, k, of the subcritical assembly, while varying the range of several parameters in the spallation target. A slightly subcritical reactor is superior to a reactor with large subcriticality in the context of the energy requirement of a small proton accelerator, the extent of radiation damage, and other safety problems. To transmute the fission products, the transmutor reactor must have a good neutron economy, which can be provided by a transmutor operated by a proton accelerator. The paper discusses the use of minor actinides to improve neutronics characteristics, such as a long fuel burn-up rather than simply transmuting this valuable material.

1. INTRODUCTION

Accelerator technology has been developed extensively in the last few decades, and now has the potential to play an important role in the nuclear-fuel cycle.[1] We studied the concept of using an accelerator-breeder (fuel producer) in connection with a program of non-proliferation of nuclear material.

To deal with the problem of a disposal of long-lived, highly radioactive waste, plans have been made to permanently store it in a stable geologic formation, such as the Yucca Mountain in Death Valley. However, there is concern that such geologic formations might change over millions of years. Therefore, alternative approaches are needed to deal with the waste such as methods that would separate the long-lived nuclei from high-level waste by transmuting the former into short-lived or non-radioactive wastes. Here, we propose to use a proton accelerator to transmute the minor actinides (MA) and fission products (FP) in more economical and safe-ways.

2. THE ENERGY RREQUIREMENTS FOR TRANSMUTING THE MAs AND FPs[2].

2.1 The direct spallation of fission products and the direct use of spallation neutrons.

The original idea of exploiting the spallation process to transmute actinides and fission products directly soon had to be given up[3]. The proton beam currents required were much larger than the most optimistic theoretical design goals for the accelerator ,which are around 300 mA. Indeed, it was shown that the yearly transmutation rate of a 300 mA proton accelerator would correspond only to a fraction of the waste generated annually by one LWR of 1 GWe.

2.2 The direct use of Spallation Neutrons

To use only the spallation neutrons generated in a proton target, the fission products would have to be placed around the target. For the highest efficiency, depending on the material to be transmuted, the fast neutrons either would have to be used as they were emitted from the target, or they would have to be slowed down by a moderator to energy bands with higher transmutation cross-sections, as, for example, the resonance or the thermal regions[4,5].

Assuming that all the spallation neutrons could be made available for transmutation the following amount of energy is necessary to transmute the fraction q_{fp}, of radionuclei per fission process in a nuclear energy system:

$$E_{fp} = q_{fp} \frac{P_b}{n_{sp}} \frac{1}{\eta_b \eta_T} \quad [MW] \qquad (1)$$

where q_{fp} = fraction of fission products to be transmuted
P_b = proton energy
n_{sp} = neutrons yield from one proton
η_b = efficiency of converting electricity into proton beam energy (=0.5)
η_T = efficiency of converting thermal energy into electricity (=0.33)

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For a 1.5 GeV proton beam emitting about 50 neutrons per spallation in a lead target, the transmutation of ^{99}Tc, ^{129}I, ^{137}Cs, ^{90}Sr, ^{85}Kr, and ^{93}Zr (constituting 28% of all fission products, see Table I) would require 0.285*30/0.5/0.33 =51.3MeV. This amount of energy represents 51.3/200 =26% of the total power production. Because of the very optimistic assumptions made in this estimate, the actual percentage of the total energy production required would even be higher. Together with the cost for processing, this type of accelerator transmutation would become prohibitively expensive, at least in a commercial nuclear-energy system.

2.3. Accelerator-Driven Subcritical Assemblies

To improve neutron economy, however, there is the possibility of multiplying the spallation neutrons in a subcritical assembly[6,7]. In such a system, the bulk of the transmutation is accomplished by fission neutrons in a reactor-like facility. Technically, this scheme can be realized by surrounding a proton target region by fissionable material. Most designs have proposed using circulating liquid lead to remove the high specific heat released in the target. However, we note that the specific heat production per neutron is considerably lower than in a fission process (30 MeV compared with 80 MeV).

First, the power production, P_{fi}, of a subcritical assembly fed by spallation neutrons is quantified:

$$P_{fi} = n_{sp} \frac{a.k}{\nu(1-k)} \frac{i}{C} E_f \qquad (2)$$

where;

k = multiplication factor
a = importance of the target position
ν = mean number of neutrons in a fission process
E_f = energy release per fission ($=3.1*10^{-11}$ J, ~200 MeV)
n_{sp} = neutron yield from one proton
i = proton current
C = proton charge ($1.6*10^{-19}$ A sec)

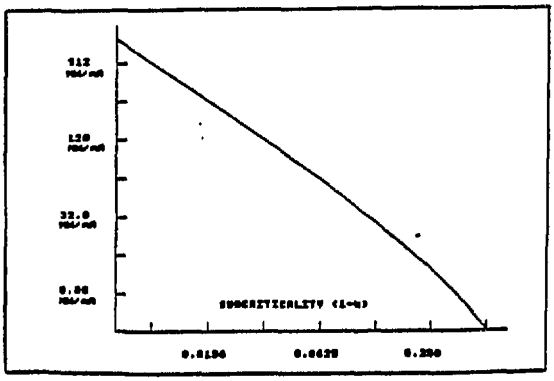

Figure 1: Power production of an accelerator driven booster as a function of subcriticality (1- k) assuming a proton beam of 1 mA at 1.5 GeV enetering a lead target leading to a release of 50 neutrons per proton.

Figure 1 shows the power production of an accelerator-driven facility as a function of the multiplication factor, k. It is assumed that a proton beam of 1.0 GeV impinging on a Pb target releases 33.3 neutrons per spallation, with an importance of a=1. This assumption leads to:

$$P_{fi}(1\ mA) = 0.796\ n_{sp} \frac{k}{(1-k)}\ [MW] \qquad (3)$$

Thus, near criticality, a 1 mA current already generates relatively high fission-power. For n_{sp} = 33.3 and k=0.974, more than 100 MW can be achieved.

The additional neutrons from the subcritical system, as well as its fission power which can be transformed into electricity, are now exploited to run the process of transmutation. Expression 4 quantifies the thermal energy required to transmute a fraction, q_{fp}, of fission products in such a system. A positive sign of E_{fp} means that there is a surplus of energy, while a negative sign indicates the need to add energy to the system from outside:

$$E_{fp} = \frac{\left[n_{sp} \frac{k}{\nu(1-k)} E_f - \frac{P_b}{\eta_b \eta_T} \right] q_{fp}}{n_{sp}\left[(1-\frac{k}{\nu}) \eta_{fp} + \frac{k}{1-k} \left((1-\frac{k}{\nu}) \eta_{fp} - \frac{q_{fp}}{\nu} \right) \right]} \qquad (4)$$

where q_{fp} = the fraction of fission products which will be transmuted by neutron gamma reaction, and:

$$\eta_{fp} = \frac{\Sigma_c(FP)}{\Sigma_c(FP+Fuel+Struct.Mat.)} \qquad (5)$$

In Eq.4, the second term in the square brackets of the numerator is the energy consumed by the spallation neutrons for transmutation; the first term is the energy gain due to the fission reaction in a subcritical assembly. The first term of denominator is the number of fission products transmuted by spallation neutrons; the second term represents the fission neutrons, including q_{fp} which is the increase in FPs (which also are to be incinerated) due to fission.

The condition for break-even or a positive energy balance is given by:

$$k \geq \frac{1}{1 + \frac{n_{sp} E_f \eta_b \eta_T}{P_b . \nu}} \qquad (6)$$

We note that this expression is independent of the proton current, and also, to a large extent, of the type of system considered. For a lead target and proton beam of 1 to 2 Gev in which n_{sp} is about 33.3 /per 1 Gev proton energy, the break-even point requires a k_{br} value near 0.7.

Table I shows the main yields of fission products generated by a LWR , their half life, and the thermal and fast (resonance) neutron- capture (n-γ) cross-sections.

To transmute the fission products of ^{137}Cs and ^{90}Sr by thermal neutrons, with an effective decay time of 3 years, requires a very high neutron flux of 10^{16} - 10^{17} n/cm^2/sec; this high neutron flux is difficult to achive with a fission reactor. Spallation neutrons with such a high flux might be generated without a subcritical assembly, but this would require a very high powered accelerator^{4,5} and would not be economical. Thus, we explored the case when only 16.46 % of the fission products would be transmuted (^{99}Tc, ^{129}I, ^{85}Kr, and ^{93}Zr but not ^{137}Cs and ^{90}Sr).

Figure 2 shows the energy required that then would be assuming p_b =1 GeV , ν=2.75, n_{sp} = 33.3 η_b = 0.5, and η_T=0.33. This figure suggest that the a great deal of energy would be required to transmute the fission products without multiplying the neutrons in a subcritical assembly, and also that the multiplication factors for k_{br} which is the break-even point for energy requirement are, respectively, k_{br} = 0.7 and 0.82 depending on the number of spallation neutrons produced from spallation targets of n_{sp} =33.3 and =16.6; further, these values do not depend on the efficiency of neutron capture by fission products η_{fp}.

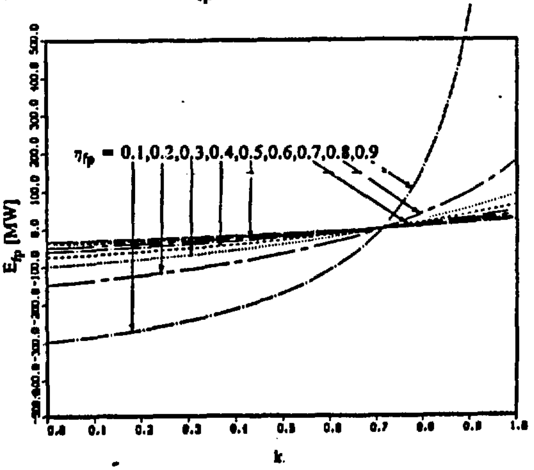

Figure 2:Energy required to transmute 16.5 % fission products (^{99}Tc, $_{129}$I, ^{93}Zr, and ^{85}Kr) assuming P_b = 1000Mev , ν =2.75 , n_{sp} = 33.3, η_b =0.5, η_t = 0.33, q_{fp} = 0.165

Above the multiplication factor k_{br}, energy generation by transmuting the fission product increases as the neutron capture efficiency η_{fp} decreases: this is due to the large number of neutrons that are required to transmute the fission products. Consequently, larger proton currents are needed than in the case where neutron capture by fission products is efficient.

We note that when q_{fp} is larger than (ν-k) η_{fp}, more fission products are created than are transmuted by fission neutrons; therefore, when k approaches to unity the E_{fp} becomes negative.

As discussed earlier, the transmutation of ^{137}Cs and ^{90}Sr by thermal neutrons is very difficult. The use of epithermal neutrons (or fast neutrons) might be considered, but presently the n-γ cross section is not well evaluated, and an accurate measurement is needed.

	90-Sr	137-Cs	99-Tc	129-I	85-Kr	93-Zr	135Cs
Yield (%)	5.91	6.15	6.12	3.56	1.33	5.45	
sub sum		12.06				16.46	
tot sum						28.52	
Half life (year)	29.	30.2	2.1* 10^5	1.6* 10^7			3.*10^6
σ_{th}(b)	0.01	0.25	20	31			
σ_{fast}(b)	0.4#	0.4#	0.2	0.2			
ϕ_{th}** ~3y HL_{eff}*	>10^{17}	>4.10^{16}	4.0 *10^{14}	2.5 *10^{14}			
ϕ_{fast}** ~10 y	>10^{16}	>10^{16}	>10^{16}	>10^{16}			

2.4. Thermal energy generated when fission products are transmuted.

Heat is evolved when fission products are transmuted by neutrons generated in the spallation and fission processes. This heat has to be removed efficiently by a coolant to get the high neutron fluxes which are desirable to attain a high transmutation rate. The amount of thermal energy, P_{fp}, generated by spallation and fission is expressed by:

$$P_{fp} = \frac{\left[\, n_{sp} \dfrac{k}{\nu(1-k)} E_f + \dfrac{P_b}{\eta_b \eta_T} \,\right] q_{fp}}{n_{sp}\left[(1-\frac{k}{\nu})\,\eta_{fp} + \frac{k}{1-k}\left((1-\frac{k}{\nu})\,\eta_{fp} - \frac{q_{fp}}{\nu}\right)\right]} \qquad (7)$$

Figure 3 shows the P_{fp} calculated for the cases described in figure 2 as function of the multiplication factor, k, of the subcritical assembly, with a neutron capture efficiency, η_{fp}, of 0.1-0.9. As shown in figure 3 for n_{sp} = 33.3, the heat generated for transmuting the fission products is rather independent of the multiplication factor, k, of the subcritical assembly except for a capture efficiency of η_{fp} =0.1 and 0.2 in q_{fp} = 16.46%. In the case of n_{sp} =16.6, as the k increases, the heat generated decreases.

2.5. Proton beam current

The proton beam current, I_{f}, which is required to transmute the fraction q_f, of fission products generated in a 1 GWe power LWR by spallation and fission neutrons is expressed as:

$$I_{fp} = \frac{q_{fp} \cdot I \quad \text{Amp}}{n_{sp}\left[(1-\frac{k}{\nu})\,\eta_{fp} + \frac{k}{1-k}\left((1-\frac{k}{\nu})\,\eta_{fp} - \frac{q_{fp}}{\nu}\right)\right]} \qquad (8)$$

where I = 56.7 A.

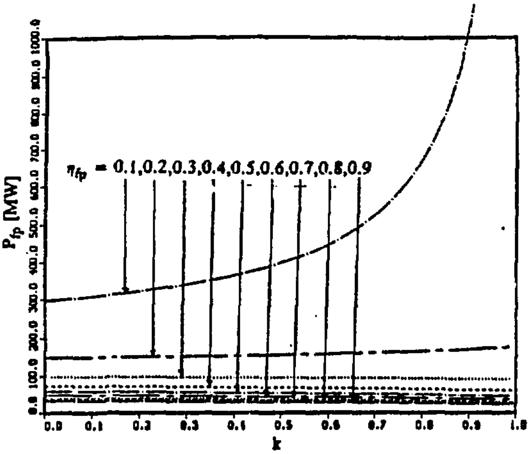

Figure.3: Thermal energy generated to transmute 16.5 % fission products (^{99}Tc, $_{129}$I, ^{93}Zr, and ^{85}Kr) assuming P_b = 1000Mev , ν =2.75 , n_{sp} = 33.3, η_b =0.5, η_t = 0.33, q_{fp} = 0.165

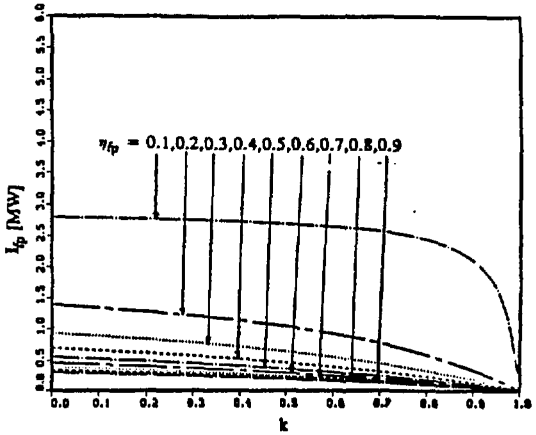

Figure.4: Proton current required to transmute 16.5 % fission products (^{99}Tc, $_{129}$I, ^{93}Zr, and ^{85}Kr) assuming P_b = 1000Mev , ν =2.75 , n_{sp} = 33.3, η_b =0.5, η_t = 0.33, q_{fp} = 0.165

Figure 4 shows the proton beam current required to transmute the fission products generated by a 1 GW LWR with the parameters used in figure 2. As k approaches 1, the current is reduced almost proportionally to the sub-criticality when the η_{fp} is larger than $Q_{fp} = q_{fp}/(\nu\text{-}k)$. However, when η_{fp} is less than Q_{fp}, such as η_{fp}=0.1 in the case of ν=2.75, the current increases indefinitely as k approaches 1, because the number of FPs transmuted is smaller than the number created by fission.

In our figures, we chose the value of k independently of the value of η_{fp}; however, these values are related because the total number of neutrons, which are used for fission and for transmuting the FP, is limited to a value less than the ν value. When the value of η_{fp} is more than 0.5, it is difficult to reach a k value higher than 0.7-0.9; consequently that the current required to transmute FP is not small unless k is very close to unity.

This finding suggested to us that FP should be transmuted by fission neutrons created in a reactor which has a good neutron economy. Further, when the FPs are transmuted by spallation neutrons created by injecting medium energy protons into a lead target, a rather large powered proton accelerator is needed, and the spallation reaction should not create long-lived isotopes.

We assumed for our calculations a 1 GeV proton, 100 mA current proton beam corresponding to 100 MW. As shown in figure 4, the proton beam current needed to transmute about 16.48 % of FP (^{99}Tc + ^{129}I + ^{85}Kr + ^{93}Zr) without multipling the spallation neutrons (k =0) is 300 mA (300 MW) for η_{fp} = 0.8. When the spallation neutrons are multiplied in a subcritical assembly of k=0.99, only a 25 mA (25MW) beam current is required for η_{fp} = 0.2, with an electric power of 50 MW to accelerate the proton beams. However, it is difficult to obtain this high k value when the η_{fp} is large, because a substantial number of neutrons are captured by fission products and then the multiplication factor k fall considerably unless transmutor has superior neutron economy. Thus, to transmute the fission products, it is vital to have transmutor which has good neutron economy; and this kind transmutor can be obtained by using the spallation neutrons created by injecting medium-energy protons into the reactor without having to be concerned about the safety problems associated with criticality.

3. DEGREE OF SUBCRITICALITY

3.1. The safety advantage of a subcritical reactor.

When we proposed using a subcritical transmutor operated by an accelerator[6,7], subcritically of 0.9 -0.95 was chosen rather arbitarily to prevent recriticality occuring, which could happen if there was a loss of coolant. Since the reactor's power can be controlled by the beam current, a control rod is not required. This large sub-criticality has been adopted in other subcritical transmutors[5,8]. However, the subcriticality which is chosen for the reactor is very important for its safety. If a loss of coolant occured, it would cause the fuel assembly to melt. Recriticality might occur due to condensation of the melted core. The choice of a subcriticality of 0.9-0.95 does not guarantee that recriticality would not occur, and the choice has not been validated further. Melting of the core is a very serious accident that should be prevented from beginning. If a large subcriticality is adopted, many difficulties will be encountered including the need for large proton-accelerator power, the generation of local heat by local proton injection, and considerable radiation damage. When we use a small subcriticality, these difficulties disappear. As shown below, the kinetic behavior of the reactor's power slows down and becomes manageable; other control mechanisms that might be used include a fuse type of liquid neutron capture material that can prevent criticality from occuring. The control rods then might be used to maintain this small subcriticality.

In contrast to the critical reactor, the operation of slightly subcritical reactor is tremendously stable. Figure 5 shows the relative change of power due to step-wise insertion of reactivity in the critical reactor, calculated with a point reactor kinetic model. Even with only a small reactivity insertion of $0.6\text{-}0.8\,\beta$, the power increases as much as 10 times and reaches 10^3 times normal within 5 sec after insertion: this calculation does not include the negative doppler coefficient of the fuel element, thus it is too pesmistic. Due to the positive sodium density coefficient, short life time, and the small delaved neutron fraction the reactivity of the fast reactor, especially one with MA fuel, has to be controlled very carefully in critical operation.

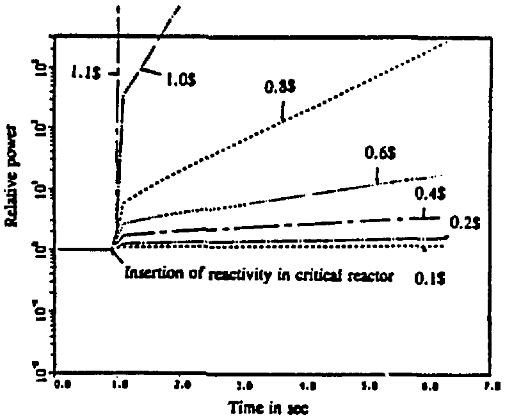

Figure 5: Relative power change in a critical reactor.

When the reactor is operated subcritically, the insertion of small reactivity does not cause a power excursion. Figure 6 shows the power change of subcritical reactors operated with spallation neutrons where the initial subcriticalities are -3, -6, -12, and -24 $, a reactivity of 1.1 $ is inserted step-wise at 1 sec, and the proton beam is shut down at 2 sec.

The power increase is, at most, less than 44 % when the initial subcriticality is -3 $. Thus, a subcritical reactor can be operated in a more relaxed condition than a critical reactor. The safety of the reactor associated with criticality can be greatly improved, and also, its operation is more economical because safety equipment is not needed to reduce this risk. Figure 6 shows the level of decay heat which is not included in the calculation.

The cause of recent trip that occurred in a phoenix reactor is still under study[10]; one of the speculations was that the trip was caused by the sudden expansion of the core. The kinetic behavior of the power suggests that about one dollar negative reactivity was inserted and the increase in the power after this dip means that an increase in positive reactivity followed. This kind of reactivity is very undesirable from the safety point of view. Many fast reactors rely on the expansion of the core-supporting plate to get a negative reactivity insertion which compensates for the change of positive reactivity due to the reduction of in the density of the Na coolant . When operation rely upon this mechanism for reducing reactivity, the sudden inflow of cold coolant into the core might cause a positive reactivity insertion, which could result in sudden increase of power. The operation of slightly subcritical reactor can avoid this kind of situation.

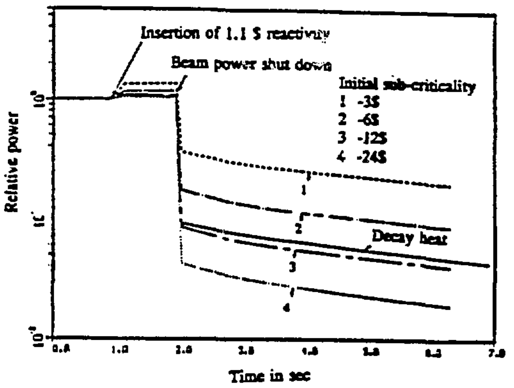

Figure 6: Relative power change in sub-critical reactor.

3.2. Neutron economy and Core geometry (flatness)

To reduce the positive reactivity caused by a decrease in Na density in the regular, sodium-cooled fast reactor, the reactor has flattened core in which D/H = (Diameter of core / Height of core) = 2. For the metal-fueled MA transmutor, which has harder energy spectrum [11], a ratio of D/H of 5-6 is adopted to maintain its inherently safe characteristics. This leaky reactor requires a large inventory of fissile material to compensate for the large neutron leakage; this requirement, in turn, makes the linear power rate very high, so that it becomes difficult to remove heat from the flattened core. Also, the neutron economy worsens. Due to the latter, this type of reactor is not suitable for transmuting fission products, although some improvement in neutron usage can be achieved by installing moderator materials in the upper and lower regions of core. However, the hard energy spectrum which is invaluable for efficiently transmuting MA, becomes softened by this moderator material which enhances the thermal neutron capture by the FPs.

The insertion of small reactivity into the subcritical reactor does not cause a sudden excursion of power; thus a positive Na-coolant coefficient is not as serious a problem as in the case of the critical reactor. Therefor, we can adopt a solid-core geometry of D/H $\approx$ 1, which reduces neutron leakage. Then, the inventory of fissile material can be reduced, the internal conversion from fissile or MA material to fissile material increases, and the fuel can be burned for a longer time without reprocessing, provided that the fuel can withstand radiation damage, or that the accumulation of fission products does not affect the metallurgic properties of the fuel. Consequently the neutron economy can be improved and FP can be transmuted very efficiently in the subcritical reactor..

3.3. Beam expansion and radiation damage

A small beam current has another advantage, that of shortening the beam expansion section. To reduce radiation damage to the target material, a proton beam with low luminosity must be expanded a using combination of dipole and quadrupole magnets. The beam expansion section for a high current beam must be lengthened when a conventional magnet is used; this causes trouble with shielding in the beam expansion section. When beam is inserted vertically, the reactor must be situated deep underground, increasing the cost of construction; furthermore, many neutrons will leak out from the front surface of core, and the neutron economy will deteriorate.

Another disadvantage of running a large subcritical assembly is the sharp gradient of flux distribution, which is of more concern from the safety point view from problems with criticality. To avoid this sharp distribution of power, the proton beam must be spread, requiring a long beam-expansion section.

Another problem in using a beam of high current is the radiation damage to the target and surrounding materials. The energy of spallation neutrons is high, and the highly energetic charged particles which are created damage the target material. When we use the high current beam, these component must be replaced frequently and, hence, plant efficiency is reduced substantially.

3.4. Use of MA to increase the fuel burn up.

The metal-fuel fast reactor has a small reactivity swing so that it does not require MA to achive a long burn-up time[11]. However, due to the presence of Na as coolant and covering material of the core, refueling takes more time and is more complex than in the LWR. Therefore, infrequent fuel exchanges are desirable. Because the addition of MA can change the reactivity swing to a positive one, we can lengthen the time for fuel burn-up, provided that the metallurgic properties of the fuel are not greatly changed by the accumulation of fission products.

A study has shown that by adding MA to the Pu fuel to a concentration of more than 10%, the burn-up reactivity swing of the oxide fuel becomes positive[12], and the oxide fuel can be burned for much longer. Due to its material properties, oxide fuel can accommodate more fission products than metal fuel.

Since MA is a very useful material for increasing fuel burn-up, MA should be used in the subcritical reactor instead of simply incinerating it.

3.5. Accelerator

Because the target assembly of the fast breeder and incinerator is slightly subcritical, k=0.99, a proton beam power only of about 15-20 MW can run the reactor at about 3.3 GWT. Small beam powered protons can be accelerated using the so-called "multistage-parallel" cyclotron arrangement[7] instead of the linear accelerator. The linear accelerator can convert electric power to beam power with higher efficiency than the cyclotron accelerator. A high current beam of 250-300 mA, accelerated by a linac, can be split by a laser, gas jet (foil), or time-wise as a pulse, and many fast reactors and transmutors can be operated in a nuclear park with only the small additional cost of an accelerator.

4. CONCLUSION.

We investigated the energy required for transmuting fission products using spallation neutrons and fission neutrons generated in a subcritical assembly surrounding the spallation target. Because subcritical assembly has a significant multiplication factor, k, the energy required to transmute fission products can be reduced; above the break-even point of k_{br}, energy is gained from fission reactions. Our study indicated that the heat generated by spallation and fission reactions is partially independent of the k value for neutron yields of 33.3 per 1 GeV proton to a lead target. However, when the yields decrease from 33.3 to 16.6 per proton, the heat generated decreases as k values increase.

Although neutron economy usually is not very important for transmuting minor actinides because neutrons are produced by fission, the neutron economy is very vital for transmuting of FPs, because neutrons are consumed in the (n,γ) process. As was shown during the development of fast breeder, it is not an easy task to get a breeding gain of more than 1.2. The fast neutron reactor, which has a hard energy spectrum, has an advantage over the thermal or regular fast reactor in generating extra neutron's which can be used to transmute the FP. However, its safety is more questionable. From this view point, the operation of a subcritical assembly is more desirable because it increases both the safety margin and the neutron economy by reducing neutron leakage.

We propose to use a proton accelerator to run a slightly subcritical fast breeder and incinerator of minor actinides. By injecting medium-energy protons into the subcritical assembly, and by providing external neutrons produced by spallation and by high-energy fission reactions, the reactor can be operated more safely than a reactor operated at criticality. The safety problems associated with supercriticality, which might be created by factors such as a positive Na-coolant coefficient and a sudden recovery of fuel bowing, can be alleviated. Further, the extra subcriticality can allow a flexible choice of structural material and more flexible operation.

The metal-fueled fast reactor shows a small decrement in reactivity of power and burn-up; by mixing the MA of ^{237}Np with the oxide-fueled reactor, the decrement of reactivity can be reduced substantially. Thus, these reactors can be operated at a subcriticality of k=0.99 with a small beam proton power of 15-20 mA and 1 GeV energy (15-20 MW). This slightly subcritical condition produces a power distribution that is more or less flat, which is important for reactor safety.

^{237}Np is very valuable fertile material for improving the neutronic characteristics of the oxide-fueled reactor, such as decrements in power and burn-up reactivity. MA material should be used efficiently in this way, rather than simply incinerating it.

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