The Energy Requirement for Transmuting Fission Products

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Abstract

Transmutation of minor actinides and fission products using spallation neutrons has been proposed. The energy requirement for this transmutation can be reduced by multiplying the spallation neutrons in a subcritical assembly which surounds the spallation target. We have studied the relation between the energy requirements and the multiplication factor k of the subcritical assembly in several parameter ranges of the spallation target.

1. Introduction

As an alternative to isolating high-level radioactive waste in a geologically stable storage formation, we investigated the possibility of transmuting the minor actinides and fission products using spallation and fission neutrons from a subcritical assembly(1,2). Using a small power accelerator with a slightly subcritical-fast neutron assembly, these minor actinides might be used to improve fuel economy as “well as to mitigate the problem of reactor safety.(3,4)

The original idea of exploiting the spallation process to transmute actinides and fission products directly soon had to be given up. 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 destruction rate of a 300 mA proton accelerator would correspond only to a fiction of the waste generated by one LWR of 1 GWe in the same period (5,6,7).

2. The direct use of Spallation Neutrons

To use only the spallation neutrons as they are generated in a proton target, the fission products would have to be placed around the target. For the best efficiency, depending on the material to be transmuted, either the fast neutrons 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(7).

Assuming that it is possible to make all the spallation neutrons available for transmutation, the following amount of energy is necessary to transmute the fraction, q_{fp}, of radio-nuclei per fission process in a nuclear energy system:

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

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)

In the case of a 1.5 GeV proton beam emitting 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) would require 0.28590/0.5/0.33 =51 .3MeV. This amountis 5 1.3/200 =26% of the total power production. Because of thevery optimisticassumptions made in this estimate, the actual percentage of energy required would even be higher. TogetherWith the cost forpmessing, this type of accelerator transmutation would become prohibitively expensive, at least in a commercial nuclear energy system.

3. Accelerator-Driven Subcritical Assemblies

To improve neutron economy, however, the possibility remainsof multiplying the spallation neutrons in a subcritical assembly. In such a system, the main part of transmutation is performed by fission neutrons in a reactor-like facility. Technically, this scheme is realized by surrounding a proton target region by fissionable material. In most designs, a circulating liquid lead is proposed to remove the high specific heat released in the target. However, we mention here that the specific heat production per neutron is considerably lower than in a fission process (30 MeV against 80 MeV).

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

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

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 2 shows the power production of an accelerator-driven facility is shown as function of 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 importance of a= 1. It leads to

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

Thus, it can be **seen that 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 transmutation process. 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 even a surplus of energy, while a negative sign indicates the need to add energy to the system from outside.

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

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 of in the [ ] of nominator is the energy consumption for transmutation by the spallation neutrons; 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 is the one due to the fission neutrons, including q_{fp} which is the increase in FP due to fission.

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

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

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 which n_{sp} is about 33.3 /per 1 Gev proton energy the break-even point requires a k_{br} value near 0.7.
But the number of nuclei transmuted depends on the power of the system, and therefore, on the proton current as shown by Equation 2.

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

Figure 2 shows the energy required to transmute 28% of the fission products (^{99}Tc,. ^{129}I, ^{137}Cs, ^{90}Sr, ^{85}Kr, and ^{93}Zr) assuming pb = 1 GeV , ν=2.5, n_{sp} =33.3, η_b=0.5 η_T=0.33, and

q_{fp}=0.285.

Figure 3 shows the energy required to transmute under the same conditions except when n^{sp} = 16.6, half the value of the last case.

To transmute the fission products of Cs^{137} and Sr^{90} by thermal neutrons, with an effective deacy time of 3 years, requires a very high neutron flux of 10^{16} -1017 n/cm^2/see, and it is difficult to achieve this high neutron flux with a fission reactor. Spallation neutrons with such a high flux might be generated without a subcritical assembly, but this would requires a very high power accelerator$^{(8,9)}$ and is not economical.

Figures 4 and 5 show the energy require to transmute 16.4 % of the fission products (^{99}Tc, ^{129}I, ^{85}Kr, and ^{93}Zr but not ^{137}Cs and ^{90}Sr) under the same conditions Figures 2 and 3, respectively.

Our study suggested that the energy required to transmute the fission products without multiplying the neutrons in a subcritical assembly is high, and that the multiplication factor of &which makes break even for energy requirement arerespectiveiy k_{br} = 0.7 and 0.82 for depend on the number of spallation neutrons produced from spallation target n_{sp} =33.3 and = 16.6. and this is not depend on so much on the efficiency of neutron capture by fission products η_{fp}.

Above the multiplication factor k_{br} , the energy generation by transmuting the fission product increase as neutron capture efficiency η_{fp} decreases. This is due to the large number of neutrons is required to transmute the fission product, and this condition requires larger proton currents than the case of efficient neutron capture by fission products.

As discussed in the above, the transmutation of ^{137}Cs and ^{90}Sr by thermal neutrons is very difficult. the use of epi-thermal neutron (or fast neutron) can be considered but the n-γ cross section is not well evaluated, it is required to have accurate measured value.

For transmutation of ^{137}Cs and ^{90}Sr by 14 Me neutron generated by fusion reaction was not discussed here, these studies will be find in reference (10).

4. Production power required to transmute Fiiion Products

When the fission products are transmuted by neutron generated in the **spallation** and fission processes generate heat. This heat has to be removed efficiently by coolant forgetting high neutron **fluxes, The** amount of **thermal** energy P_{fp} **generated by the spallation and fission processes is** expressed by

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

Figures 6-9 show the P_{fp} calculated the cases described in figures 2-5 as function of the multiplication factor k of the subcritical assembly and in the parameter of the neutron capture efficiency η_{fp} of **0.4-1.0.** As shown in **the** figures **6and** 7 for n_{sp} = 33.3, the generated heats **for transmuting the fission product are rather independent on the multiplication factor k of subcritical assembly except the case of capture efficiency of η_{fp} =0.4 in q_{fp} = 28.5%. For the case of n_{sp}** = **16.6, (Figures** 8 and 9) as the k **increse,** the generated heats decrease.

5. **Proton beam current**

Proton beam currents I_{fs} **which** required to **transmute the fraction** q_{fs} of **fission** product generated by 1 GWe power **LWR** by **spallation** neutron and the fission **neutrons is** expressed as

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

Where I = **56.7A.**

Figures 10-13 show the proton beam current required to transmute the fission product generated by 1 GW LWR in the parameter used in the ease of the figures 2-5. As the k becomes close to 1 the current is reduced as almost proportionally to. the sub-criticality.

The n_{sp} is defined as the neutron yields per 1 GeV proton injecting into the target. The power of accelerated 100 mA currents proton beam is 100 MW. As shown in the figure 11, the proton beam current to transmute the 16.48 % of FP (Tc + I + Kr + Zr) without multiplication of spallation neutrons (k =0) becomes 150 mA (150 MW) for η_{fp} = 0.8. When the spallation neutron multiplied with subcritical assembly of k= O.95, only 25 mA (25MW) beam current is required, the electric power of 50 MW is used for accelerating the proton beams.

6. Conclusion.

We investigated the energy required for transmutation of the fission products using the spallation neutrons and the fission neutron generated the subcritical assembly surround the spallation target. With the subcritical assembly which has a significant multiplication factor k the energy required to transmute fission product can be reduced, and above the break even point of k_{br}, the energy is gained from the fission reactions. The study indicated that the heat generated by spallation and fission reaction becomes somewhat independent on the k value. for the neutron yields of 33.3 per 1GeV proton to lead target. But when the yields decreases from 33.3 to 16.6 per proton, the generated heat decreases as k values increases.

The proton beam current to transmute the fission products which generated by 1 GWe LWR becomes small as the k value approaches to 1, and the large beam current is required when the no subcritical assembly which has large k , is provided.

Acknowlegements

We wish to thank Dr. A. D. Woodhead for her editorial work.

	90-Sr	137-Cs	99-Tc	129-1	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)			0.2	0.2			
ϕ_{th}** ~3y HL_{eff}*	>1017	$>4.10^{16}$	$4.0*10^{14}$	$2.5*10^{14}$			
ϕ_{fast}** ~10 y HL_{eff}*			>1016	>1016			

* Effective half life, ** unit in $n/cm^2/sec.$,

Table I. Fission products yield,half life times,thermal and fast (n,gamma) cross section, transmutaion fluxes.

1. H. TAKAHASHI, “The Role of Accelerator in the Nuclear Fuel Cycle" Proc. of 2nd Int. Symp. in Advanced Nucl. Energy Research. p. .77, Mito, JAERI, Jan. 24-26, (1990).

2-a. “H. TAKAHASHI, "Actinide Transmutation by the Spallation Process,” presented at Workshop on the Feasibility of Research Program in Actinide Transmutation by Spallation Process, Euratom, Ispra, Varese, Italy, June 18-21, (1985).

2-b. P. BONNAUE, H. RIEF, P. MANDRILLON, and H. TAKAHASHI; "Actinide Transmutation by Spallation in the Light of Recent Cyclotron Development”; NEACRP-A-9 10, Session B. 1.2, (European American) Reactor Physics Committee Report ,(1987).

3. H. TAKAHASHI, “A Fast Breeder and Incinerator Assisted by a Proton Accelerator, ” Inter. Conf. on Emerging Nucl. System (ICENES-91), Monterey, CA, June 16-21, 1991 [2P-23], Transactions of Fusion Technology, 20, 657, (1991).

4. H. TAKAHASHI, ‘The Use of Minor Actinides and a Small Power Proton Accelerator for Fast Reactor with a High Breeding Gain,” (Alternative Ways to Dispose of High Level Waste: The Merits of Antactic Icefield, the Moon, and Outer” Space), the Symposium on Accelerator-Based Transmutation held at Paul Scherrer Institute, Switzerland, March 23-26, 1992.

5. H.TAKAHASHI and N.MIZOO, “ Transmutation of Cs-137 by Using 10 GeV proton spallation reaction" J. Nucl.Sci.and Tech. 16 p.613 (1979).

6. E.M.KRENCIGLOWA, A.A.HARM, Nucl. Instr. and Meth. 185, p393, 1981.

7. H.U. WENGER, P.WYDLER, F. ATCHISON," The influence of Difference Recycling Scheme on Toxicity Reduction for Transmuattion System Using High-Energy Spallation Reactions: OECD-NEA Specilist’s Meeting on Accelerator based Transmutation. PSI Wurenlingen/ Villigen, March 24-26,1992.

8. H. TAKAHASHI, N. MIZOO, and M. STEINBERG, “Use of the Linear Accelerator for Incinerating the Fission Product of Cs^{137} and Sr^{90}, " International Conference on Nuclear Waste Transmutation, July 22-24, 1980, University of Texas at Austin.

9. C. BOWMAN, ‘Data Needs for Construction and Application of Accelerator-Based Intense Nuetron Sources”, Proc. of 2nd Int. Symp. in Advanced Nucl. Energy Research, p. 149, Mite, JAERI, Jan. 24-26, (1990).

10. H. HARADA,.H.TAKAHASHI, A. ARONSON,H. TAKASHITA,K.KONASHI,T. KASE, AND N.SASAO. “Transmutation of Fission Products and Transmutation by 14 MeV Neutrons, ANS/ENS International Meeting Nov. 15-20, 1992 Chicago, Illinois.

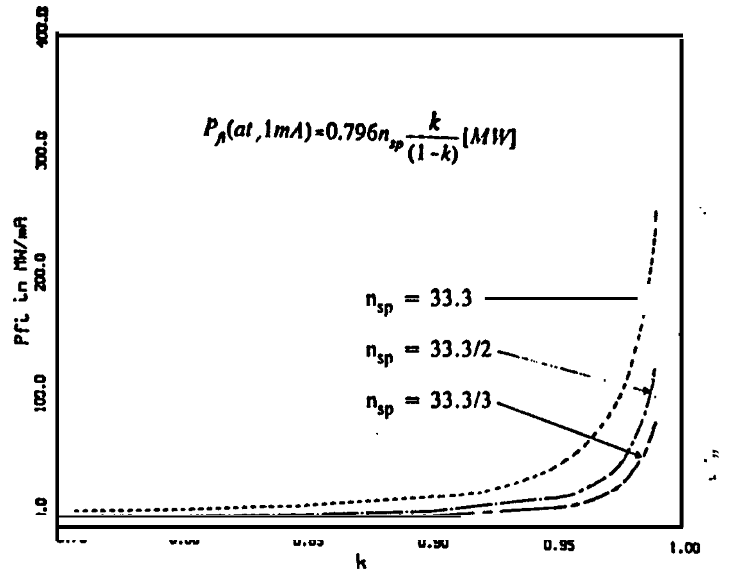

Figure 1. Power production of accelerator-driven booster as function of multiplication factor, k, assuming a proton beam of 1 mA at 1 GeV entering a lead target generating of 33.3 per

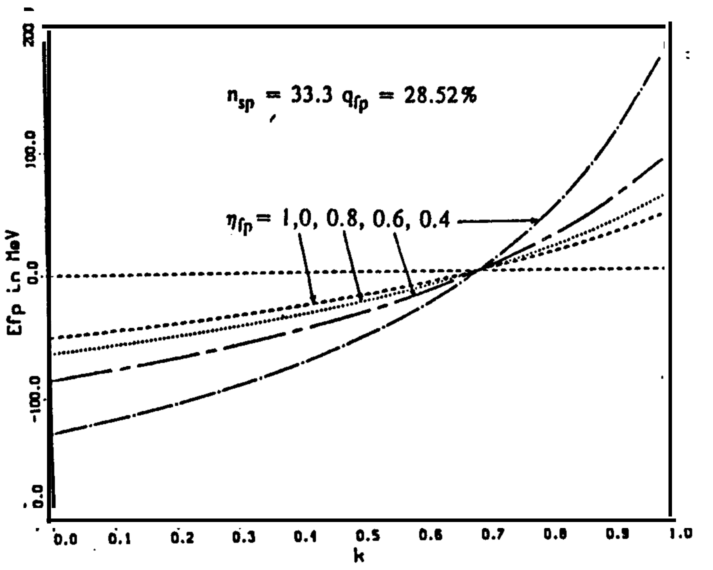

Figure 2. Thermal energy required to transmute FP (fraction of q_{fq}) produced by one fission. (q_{fp} = 28.52% Cs + Sr + Tc + I + Kr + Zr , n_{sp}=33.3)

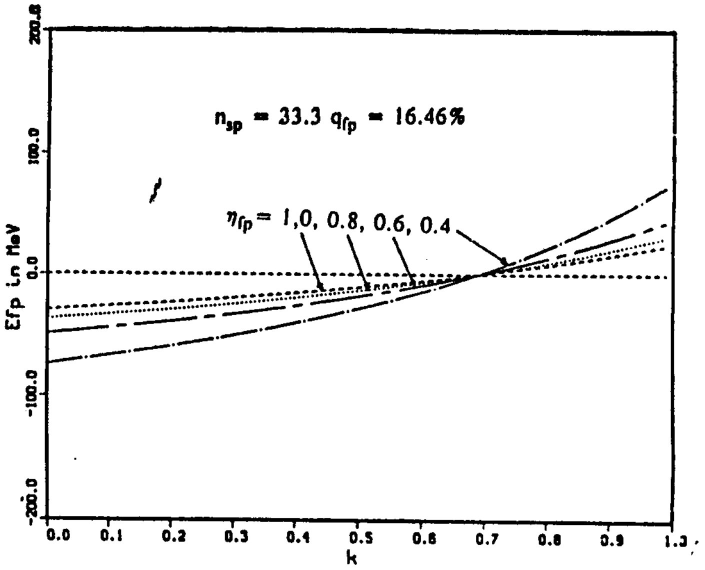

Figure 3 Thermal energy required to transmute FP (fraction of q_{fq}) produced by one fission. (q_{fp} = 16.46% Tc + I + Kr + Zr, n_{sp}=33.3)

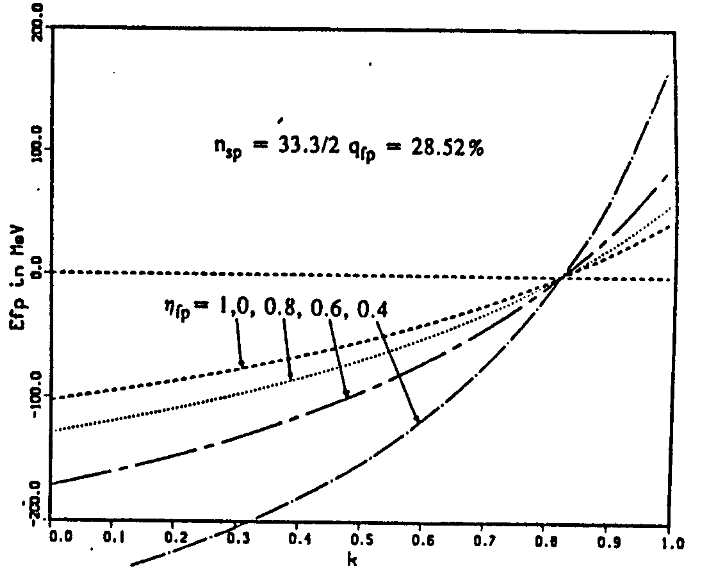

Figure 4. Thermal energy required to transmute FP (fraction of q_{fq}) produced by one fission. (q_{fp} = 28.52% Cs + Sr + Tc + I + Kr + Zr , n_{sp}=16.6)

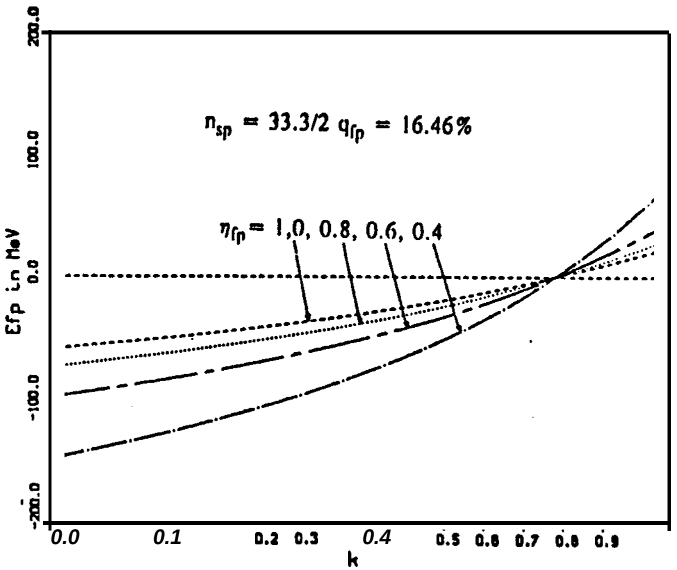

Figure 5. Thermal energy required to transmute FP (fraction of q_{fq}) produced by one fiss
(q_{fp} = 16.46% Tc + 1 + Kr + Zr, n_{sp}=16.6)

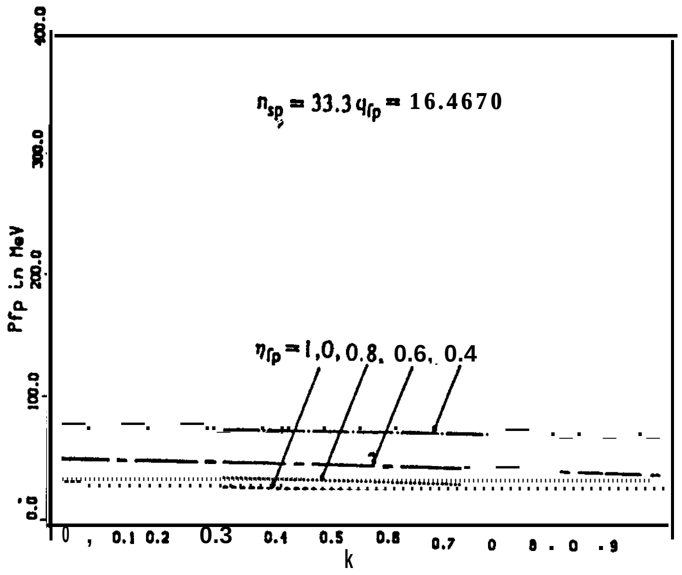

Figure 7. Heat generated to transmute FP (fraction of $q_{f\alpha}$) produced by one fission
(q_{fp} = 16.46% Tc + I + Kr + Zr, n_{sp}=33.3)

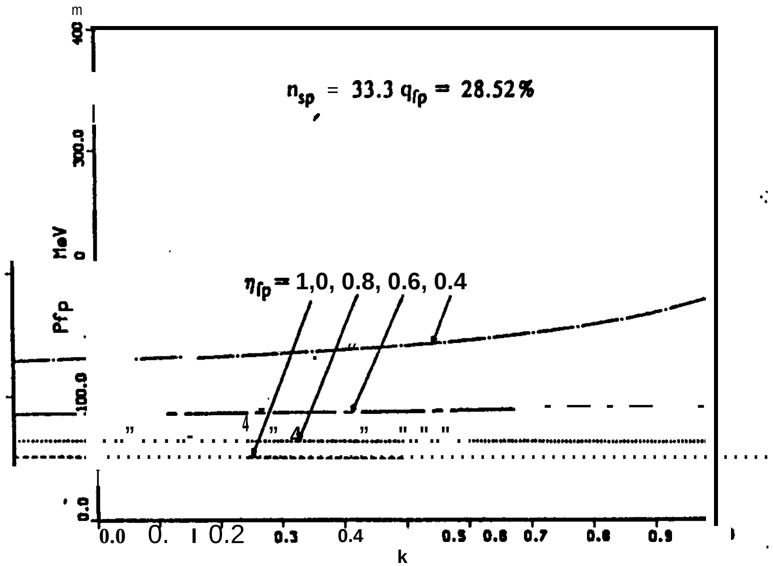

Figure 6. Heat generated to transmute FP (fraction of $q_{f\alpha}$) produced by one fission
(q_{fp} = 28.52% Cs + Sr + Tc + 1 + Kr + Zr, n_{sp}=33.3)

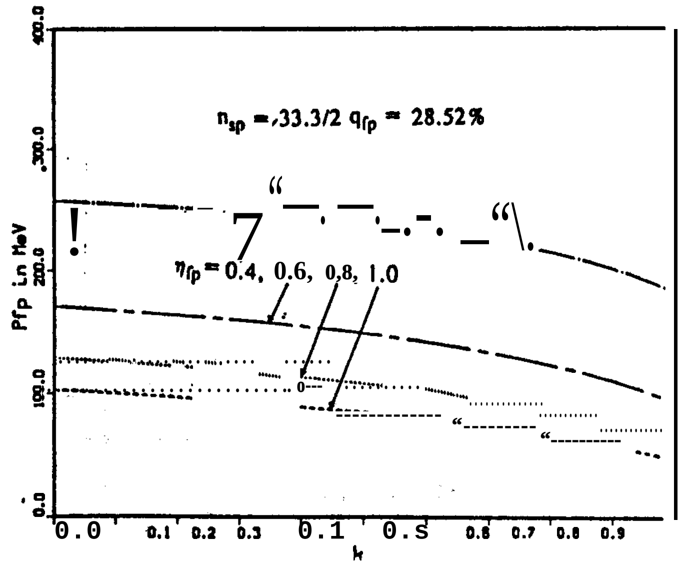

Figure 8. Heat generated to transmute FP (fraction of $q_{f\alpha}$) produced by one fission
(q_{fp} = 28.52% Cs + Sr + Tc + I + Kr + Zr, n_{sp}=16.6)

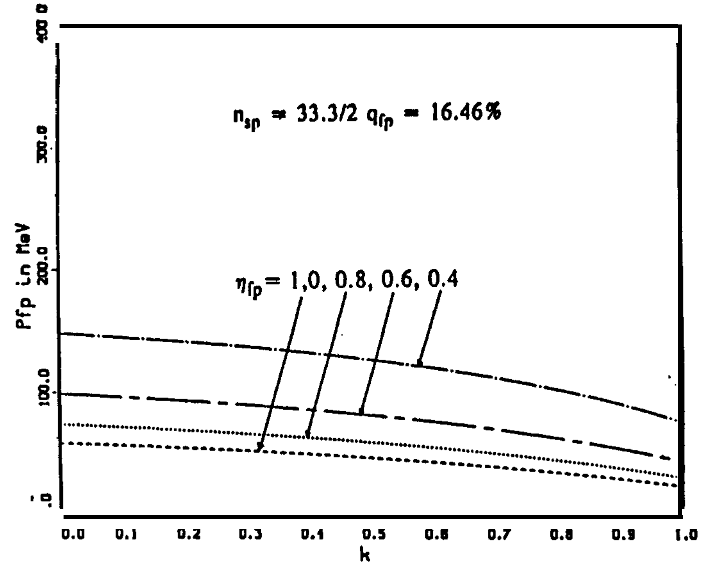

Figure 9. Heat generated to transmute FP (fraction of q_{fq}) produced by one fission

(q_{fp} = 16.46%   Tc + I + Kr + Zr, n_{sp}=16.6)

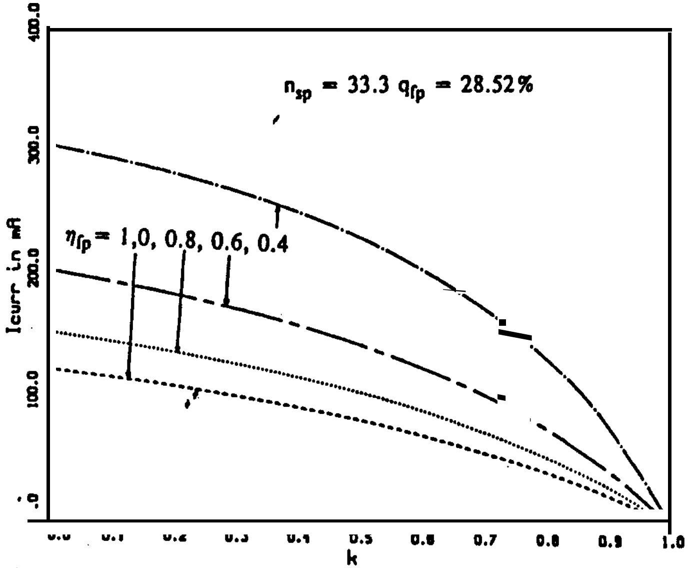

Figure 10. Proton beam current I_{curr} (in mA) required to transmute FP (fraction of q_{fq}) generated by 1 GWe LWR.

(q_{fp} = 28.52%   Cs + Sr + Tc + I + Kr + Zr , n_{sp}=33.3)

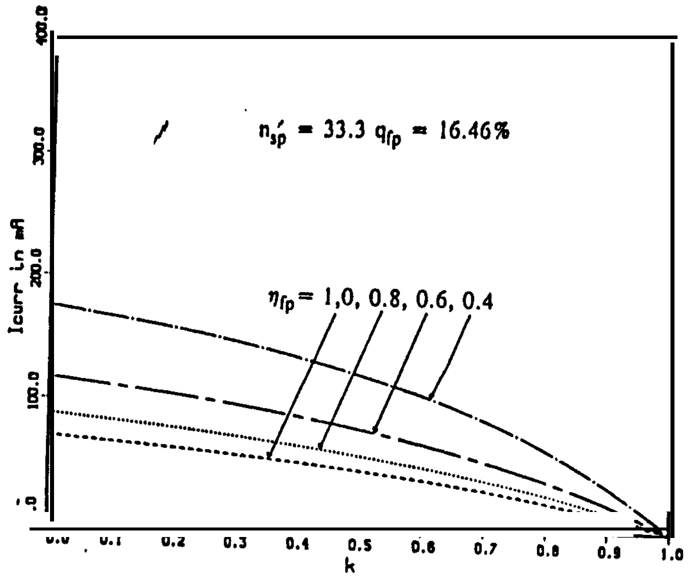

Figure 11. Proton beam current I_{curr} (in mA) required to transmute FP (fraction of q_{fq}) generated by 1 GWe LWR.

(q_{fp} = 16.46%   Tc + I + Kr + Zr, n_{sp}=33.3)

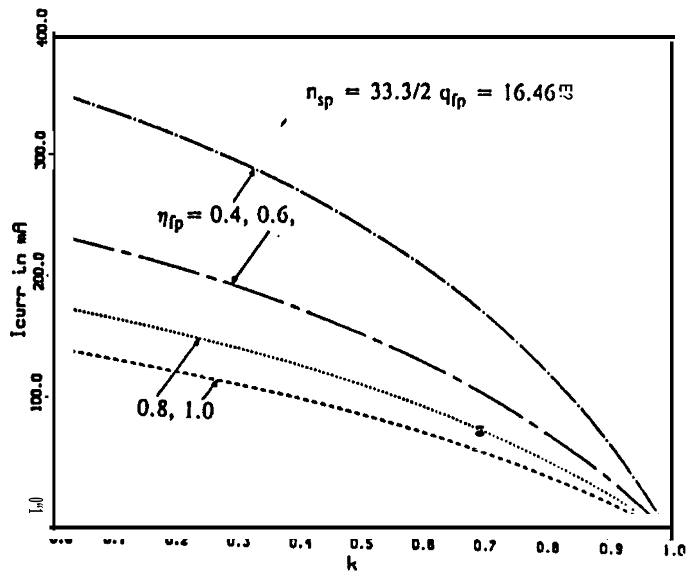

Figure 12. Proton beam current I_{curr} (in mA) required to transmute FP (fraction of q_{fq}) generated by 1 GWe LWR.

(q_{fp} = 16.46%   Tc + I + Kr + Zr, n_{sp} =16.6)

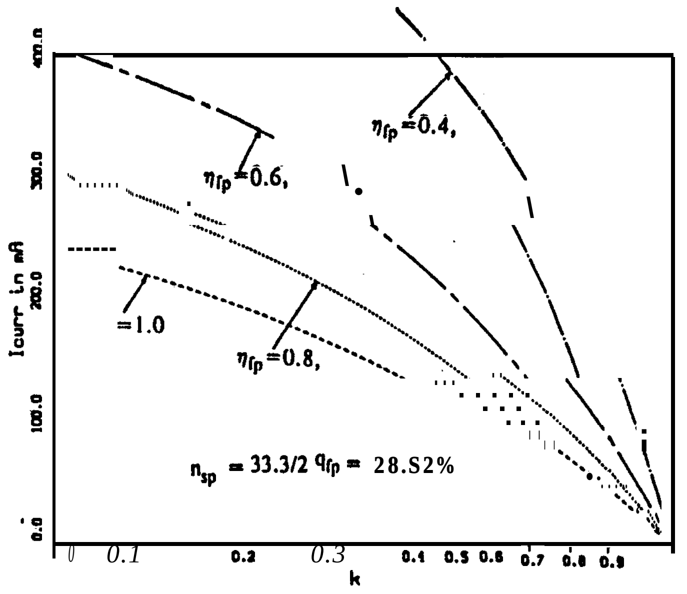

Figure 13. Proton beam current I_{curr} (in mA) required to transmute FP (fraction of q_{fq}) generated by 1 GWe LWR.

(q_{fp} = 28.52% Cs + Sr + Tc + I + Kr + Zr, n_{sp} = 16.6)