JANUARY 1994

ECN-R--94-001

TRANSMUTATION OF FISSION PRODUCTS IN REACTORS AND ACCELERATOR-DRIVEN SYSTEMS

Some critical remarks

A.J. JANSSEN

ABSTRACT

A comparison is made between the use of nuclear reactors and of accelerator-driven systems for the transmutation of long-lived fission products. Energy flows and mass flows in several scenarios are considered. Economical and safety aspects of the transmutation scenarios are compared.

CONTENTS

1. INTRODUCTION

Transmutation of fission products by neutron capture should be done in a clean, safe and efficient way. It is sometimes argued that transmutation in a fission reactor is not a clean process [1]: the required neutrons are made available by the fission process which is accompanied by the production of exactly the same fission products that one wants to destroy (and, in addition, by the production of many other 'dirty' radio-active nuclides). Neutrons produced in a spallation source triggered by protons from an accelerator are said to be much 'cleaner'. Another argument put forward in favour of systems driven by a spallation source is the high neutron flux that can be established [2]; for a certain desired transmutation rate the inventory of fission products can therefore be kept relatively small, which would create an economical advantage. Finally, it is said [2] that accelerator-based systems do not experience criticality problems and are thus essentially much safer than nuclear reactors.

Such arguments in favour of accelerator-driven systems cannot be maintained if one has a closer look at the (optimized) transmutation process. This can be made clear by the following simple example.

Let us assume that a 1.5 GeV proton can create 50 neutrons and that all these neutrons transmute a long-lived fission product, e.g. Tc-99. For the production of this proton a certain amount of electrical energy is needed. One of the basic ideas about the solution of the waste problem is that the nuclear industry itself should take care of the nuclear waste that it created. This immediately leads us to the conclusion that the electrical energy for the production of that proton should be produced by a nuclear reactor and not, for instance, by a fossil-fired plant or a renewable energy system. With an assumed efficiency of 50% of the proton accelerator, a thermal efficiency of 33% of the reactor, and an energy yield of 200 MeV per fission, the production of this single 1.5 GeV proton required 1500x2x3/200=45 fissions (see Fig. 1). By these fissions (of, say, U-235) 2.44x45 = 110 neutrons were liberated; 45 of them were useful for the required energy production (but created also 90 'dirty' fission products), the remaining 65 neutrons produced other 'waste' like Pu-239 and U-236. The net result is thus that 50 Tc nuclei have been transmuted and that much more than 50 nuclei of other 'dirty' materials have been produced. An important point to observe too is the very poor energy efficiency of the process: 9000 MeV(th) were produced in the reactor, in the transmutation device perhaps 33% of the kinetic energy of the proton, i.e. 500 MeV, can be recovered as electrical energy.

Furthermore, the ability to establish a high neutron flux is not reserved for accelerator-driven systems. The only reason why one does not aim at increased neutron fluxes in power reactors is simply that this does not result in an economically optimal system.

The above example shows that it is not correct to consider accelerator-driven transmutation systems as 'clean and economical' in contrast to transmutation in reactors. In the sequel of this report we will try, with the help of a few simple scenarios, to place the various transmutation options in the right perspective by considering all relevant mass flows and energy flows. In the discussion of these scenarios some safety aspects of accelerator-driven systems will be considered as well.

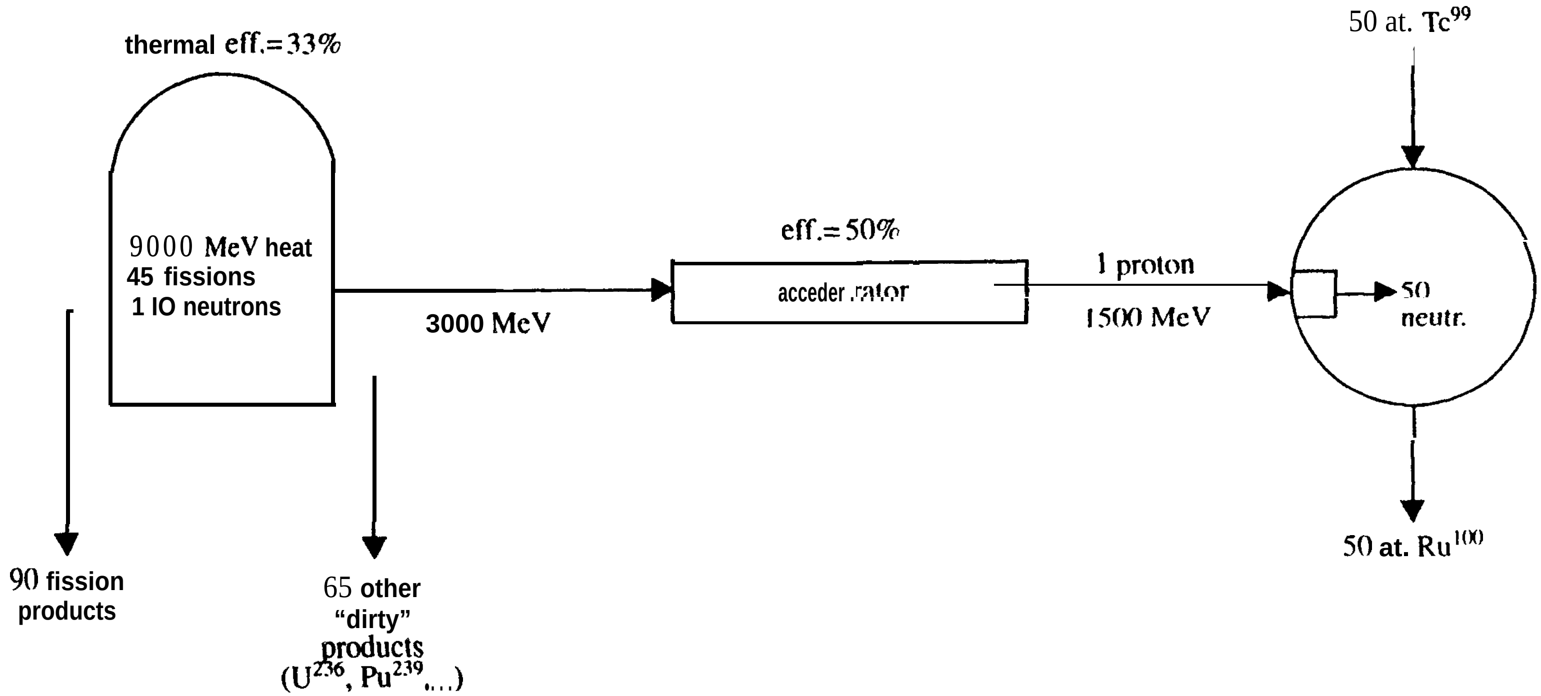


Fig. 1 *Energy flows and mass flows accompanying the transmutation of fission products in an accelerator-driven system.*

2. SCENARIOS FOR THE TRANSMUTATION OF FISSION PRODUCTS

In order to facilitate the comparison of the various transmutation options, we will make the accelerator-driven system resemble a nuclear reactor as close as possible. Both systems work with thermal neutrons. In our examples D_2O is the moderator; heavy-water moderated systems have the advantage of a high neutron economy. It should be pointed out, however, that the conclusions drawn from our example calculations are of a generic character and are equally valid for other types of reactors and moderators.

The reactor in our examples, a CANDU, is fed by slightly enriched uranium (SEU, 1.2% enrichment} continuous on-line refueling takes place, the average discharge burn-up is 21 GWd per Tome. The thermal efficiency of the reactor is 33%. The fuel costs account for 20% of the total electricity production costs (which, in our examples, are set equal to x $ per unit electrical energy).

The accelerator-driven system consists of a vessel filled with D_2O and fission products; it also contains a spallation source, triggered by 1.5 GeV protons from an accelerator. The heat liberated in the vessel is converted to electrical energy with an efficiency of 33%. The efficiency of the accelerator is 50%.

Energy flows are indicated in the figures by arrows ($\rightarrow$) and are expressed in MeV per unit of time; mass flows are indicated by double arrows ($\Rightarrow$) and expressed in particles per unit of time.

The fission product to be transmuted in our examples is Tc-99.

A. Transmutation in a reactor

Figure 2 shows, slightly schematized, k_∞ of the CANDU assembly as a function of burn-up [3]. To operate the reactor with sufficient margin, the assembly-averaged k_∞ should not be smaller than 1.05. This is the reason why the discharge burn-up is 21 GWd/T and the average burn-up of the assemblies resident in the reactor is 10.5 GWd/T.

We could add some Tc to the assemblies and still keep the average k_∞ at 1.05 if we accept a lower discharge bum-up. Let us assume that the fuel is discharged at 7 GWd/T, which is reached after an irradiation time of about one year (this is the usual irradiation time if the CANDU is fed by natural uranium). The formula for k_∞ reads:

$$k_\infty = \nu F/(A_f + A_{par} + A_{Tc}) , \qquad (1)$$

where F is the fission rate and A is the absorption rate; the subscript f refers to fuel and par refers to other parasitic absorption. A_{Tc} is the transmutation rate of Tc. From Fig. 2 we see that Δk_∞ caused by the addition of Tc can be as large as -0.13 if a discharge burn-up of 7 GWd/T is accepted (at a bum-up of 3.5 GWd/T k_∞ is equal to 1.18 and 1.05 for an assembly without Tc and an assembly with Tc, respectively). With Eq. (1) it is easily derived that the

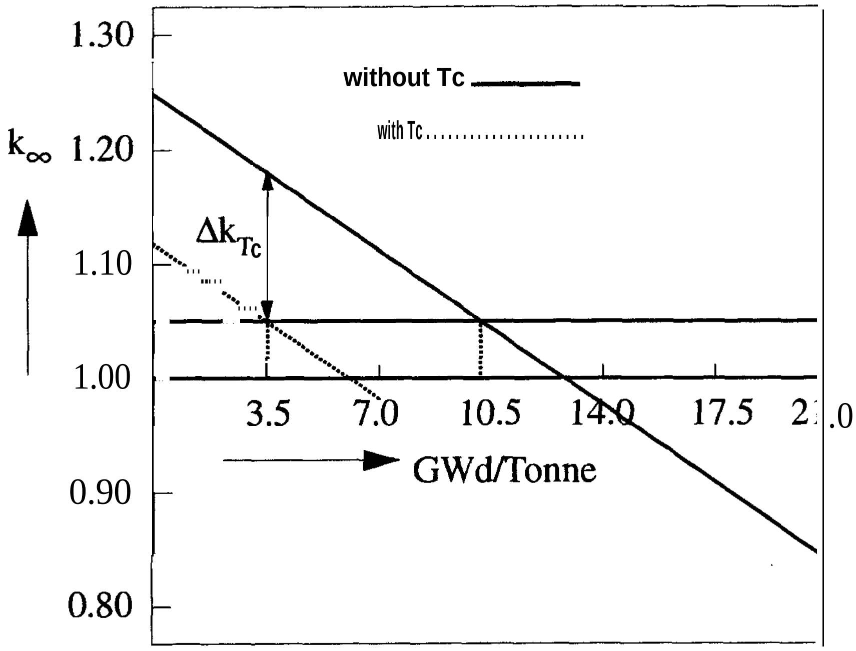


Fig. 2 *k_∞ of a CANDU assembly with Slightly Enriched Uranium (SEU), as a function of burn-up.*
Δk_{Tc} = 0.13, which means that the Tc absorption rate equals 0.105 × the neutron production rate or 0.27 × the fission rate (see text)

transmutation rate of Tc is then equal to 0.105 times the neutron production rate vF, or (with v of the fissile U/l% mixture ≈ 2.6) 0.27 times the fission rate F. In Fig. 3 the most important flows are shown.
The fission yield γ of Tc-99 is about 6% for both U-235 and Pu-239. So this single CANDU can transmute the Tc production of 4.5 CANDUs. The penal-

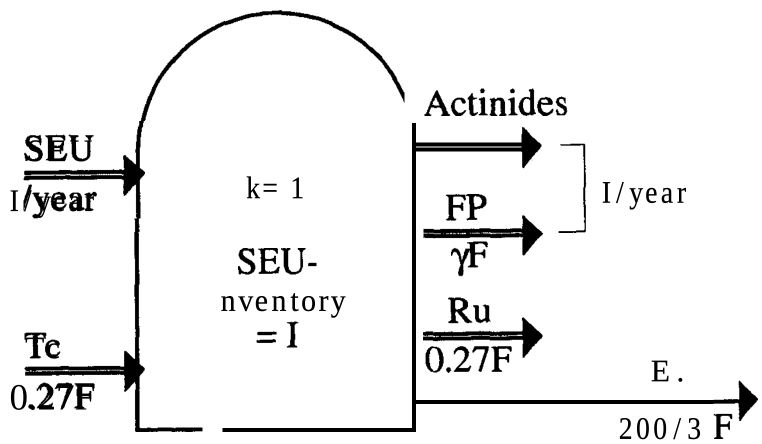
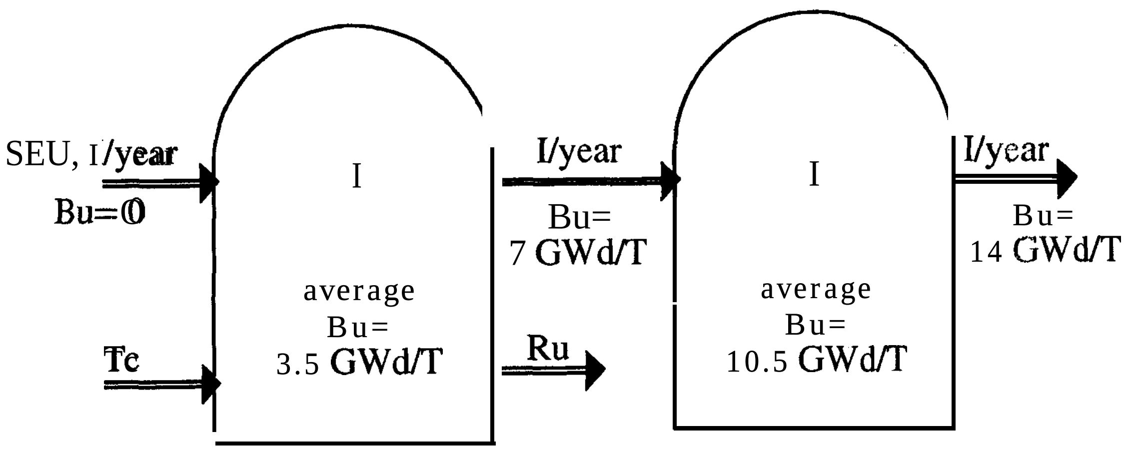


Fig. *3 Energy flows and mass flows in a CANDU with Tc transmutation.*
A = absorption rate; F = fission rate; E_0 = energy output;
FP = fission products; γ = fission yield;
200 MeV per fission; thermal efficiency = 113;
A_{Tc} = 027F (see Fig. 2) = 0.27(3/200)E_0 = 0.004E_0

ty to be paid for these transmutations is the lower fuel burn-up. However, it should be noticed that the discharged fuel, with a bum-up of only 7 **GWd/T,** can be used very well in a **CANDU** without Tc transmutation, In such a **CANDU** it can be burned up to 14 **GWd/T,** which results in an **assembly-**averaged burn-up of 10.5 **GWd/T** with the desired average k_∞ of 1.05 (see Fig. 2). Such a symbiosis of two **CANDUs** is pictured in Fig. 4, where also a cost evaluation is made. Transmutation of the Tc production of 4.5 **CANDUs** turns out to increase the electricity production costs of the two **CANDUs-in-**symbiosis by 10%; otherwise stated, the 4.5 **CANDUs** that burn their own Tc production will experience a 4.4% higher electricity production cost. (Of course, other costs connected to recycling are involved as well, but we will not consider these here since they are the same in each transmutation scenario.)



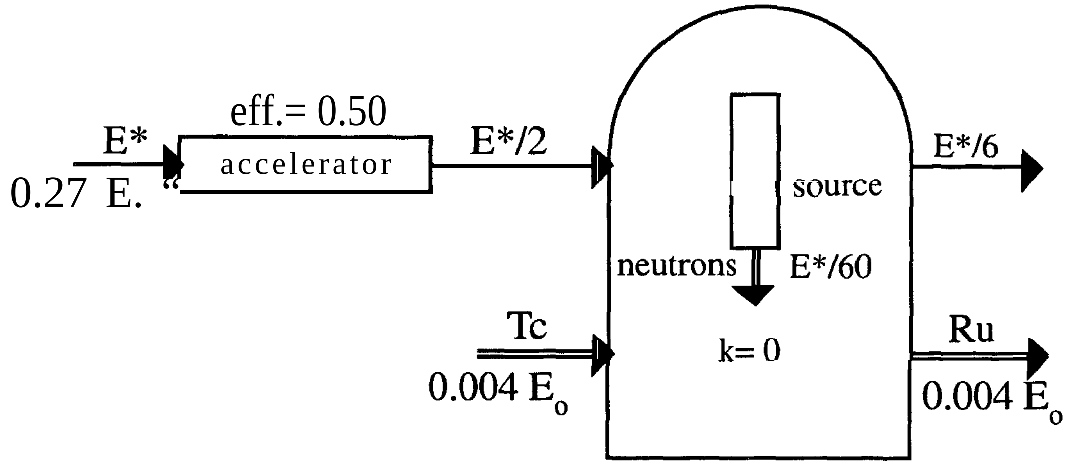
	lectric ty ts				
	freed costs		fuel costs		
One **CANDU,** burn-up O → 7 **GWd/T**	0.8x		0.6x		
One **CANDU,** burn-up 7 + 14 **GWd/T**	0.8x		-		+
	1.6x	+	0.6x	= 2.2x	
Two **CANDUs,** burn-up O → **21 GWd/T**	1.6x	+	0.4X	= **2.0x**	—
Cost of Tc transmutation				0.2X	

Fig. 4 ***Symbiosis of two CANDUs, one with and one without Tc transmutation.***

A few words should be said about the amount of Tc **in** the transmuting reactor. By considering the capture cross-section of Tc and the fission **cross-**section of the fuel, one can show that for each **fuel** bundle (with 37 fuel pins) the reactor should contain about one pin of the same dimensions as the fuel pins and completely filled with Tc. This Tc pin could be placed in the **centre** of the moderator where the thermal neutron flux is maximal. On the other hand, putting this pin in the centre of the fuel bundle would serve another desired **modification** of the reactor in that it would make the coolant void coefficient less positive.

B. Non-multiplying transmute

A 1.5 GeV proton can produce 50 neutrons; 90% of these neutrons are assumed to transmute Tc, the remaining 10% is lost by parasitic absorption and leakage. For a straightforward comparison with scenario A we choose the dimensions of the system such that it can transmute the same Tc production of 4.5 CANDUs: $A_{Tc} = 0.27F = 0.004E_0$ (see Fig. 3: F is the number of fissions per unit of time in a CANDU, E_0 is the electricity production of that CANDU expressed in MeV per unit of time).



$$\frac{Tc}{0.9} = \text{neutron source} = \frac{E^*/2 \times 50}{1500} = \frac{0.004\, E_0}{0.9}$$

$$\therefore E^* = 0.27\ E_0$$

Fig. 5 ***Energy flows and mass flows in a Tc transmute fed by 1500 MeV protons. Same transmutation rate as in the CANDU, viz. $0.004E_0$ (see Fig. 3). 50 neutrons 'per proton; neutronic efficiency of transmutation is 0.90 (10% losses due to leakage and parasitic absorption)***

In Fig. ***5 the*** energy flows and mass flows are shown, together with a derivation of the required energy input to the system, which turns out to be equal to $0.27E_0$. One might argue that the transmutation of Tc in this scenario B is accompanied by a number of fissions which is only 27% of the number of fissions involved in scenario A. However, far more important is the fact that in scenario A E_0 units of electricity are produced which can be sold, whereas in scenario B (5/6)x0 .27E_0 units must be purchased. From a comparison with the cost estimates of scenario A given in Fig. 4 we can deduce that - even if we neglect all investment costs of accelerator, spallation source and transmute - transmutation according to scenario B is more expensive than transmutation according to scenario A. Including the - very large - investment costs would more than double the transmutation costs of scenario B.

The main problem of the accelerator-driven system is its bad energy balance. We can try to improve its energy efficiency by adding some fissile material and taking advantage of the neutron multiplication, which brings us to scenario C.

C. Multiplying transmute

In order to stay as close as possible to scenarios A and B, some SEU is added to the transmute. Just like with the transmuting CANDU, the fuel will be discharged at a bum-up of 7 GWd/T; the average burn-up of the fuel in the transmute is 3.5 GWd/T. The effective multiplication factor of the transmute k will depend on the amount of added fuel, but will have a value between O and 1. The following relation holds fork

$$k = \nu F/(A_f + A_{Tc}/0.9)\,, \qquad (2)$$

where - just like in scenario B - a ratio of 9:1 is assumed of the Tc transmutation rate A_{Tc} and the neutron loss rate by parasitic absorption and leakage. For the neutron balance in the transmute we have the relation (see Fig. 6):

$$VF + E^*/60 = A_f + A_{Tc}/0.9\,. \qquad (3)$$

The dimensions of the transmute are chosen such that the transmutation rate A_{Tc} is again equal to the Tc production rate of 4.5 CANDUs: $A_{Tc} = 0.004E_0$ (see Fig. 5). The values of F and A_f will of course depend on the amount of added fuel. From Eqs. (2) and (3) we find that the source multiplication M is equal to

$$M = \text{(fission source)/(spallation source)} = \nu F/(E^*/60) = W\text{-}k). \qquad (4)$$

A similar relation can be derived for the 'energy multiplication'. If we denote the ratio $A_f/\nu F$ by α, the input energy flow E“ can be written as a function of k (see Fig. 6):

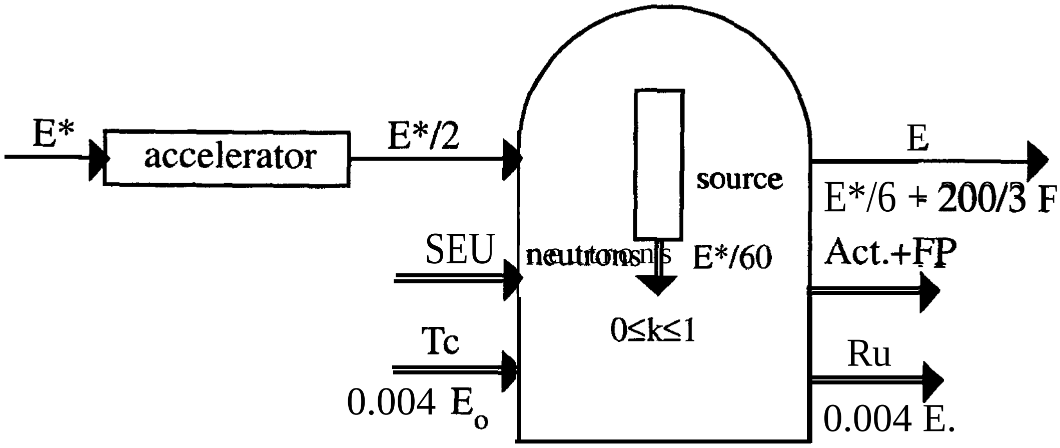
$$E^* = (0.8/3)E_0(1\text{-}k)/(1\ \text{-}\alpha k)\,. \qquad (5)$$

The output energy flow E rdso follows from Fig. 6

$$E = E^*/6 + (200/3)F = (0.4/9)E_0\{1 + k(20/3\nu - 1)\}/\{1 - \alpha k\}\,, \qquad (6)$$

and the energy multiplication is

$$E/E^* = 1/6 + (1/0.9\nu)k/(1\text{-}k)\,. \qquad (7)$$



$$\alpha = \frac{A_f}{\nu F}$$

$$k = \frac{\nu F}{\alpha \nu F + A_{Tc}/0.9}$$

$$A_{Tc} = 0.004\ E_o \qquad \Bigg\} \quad F = \frac{0.004\ E_o}{0.9\ \nu(1/k - \alpha)}$$

$$\Rightarrow E^*$$

$$\text{neutron balance : } \nu F + E^*/60 = \alpha \nu F + A_{Tc}/0.9 \ \}$$

Fig. ***6 Energy flows and mass flows in a subcritical system with the same Tc transmutation rate as in the CANDU***

Equations (5) and (6) can be **quantified** further. For ν we again take the value 2.6 (see scenario A). The value of α of the fuel is independent of the multiplication factor k of the system. For k = 1 we can make use of Fig. 2 for **the** critical **CANDU** with Tc transmutation, where we **find** (at the average burn-up of 3.5 **GWd/T):**

$$k = \nu F/(A_f + A_{Tc}/0.9) = 1 = 1/(\alpha + 0.105/0.9)\ , \qquad (8)$$

from which it follows that α = 0.883. With these numerical values the input and output flows have been calculated. They have been plotted in Fig. 7, both normalized to the electrical output E_0 of a single **CANDU.** These flows E* and E are also a measure for the power and dimensions of the accelerator and transmute, respectively. Input and output energy flows are equal if k is equal to

$$k = 1/(1 + 4/3\nu) = 0.66. \qquad (9)$$

It appears that nothing really new has been found in this scenario. In the limit of k = O we have again the **(low-efficiency)** scenario B, in the limit of k = 1 we have the (high-efficiency) scenario A, and in between these limits **all** relevant parameters vary in a very smooth way.

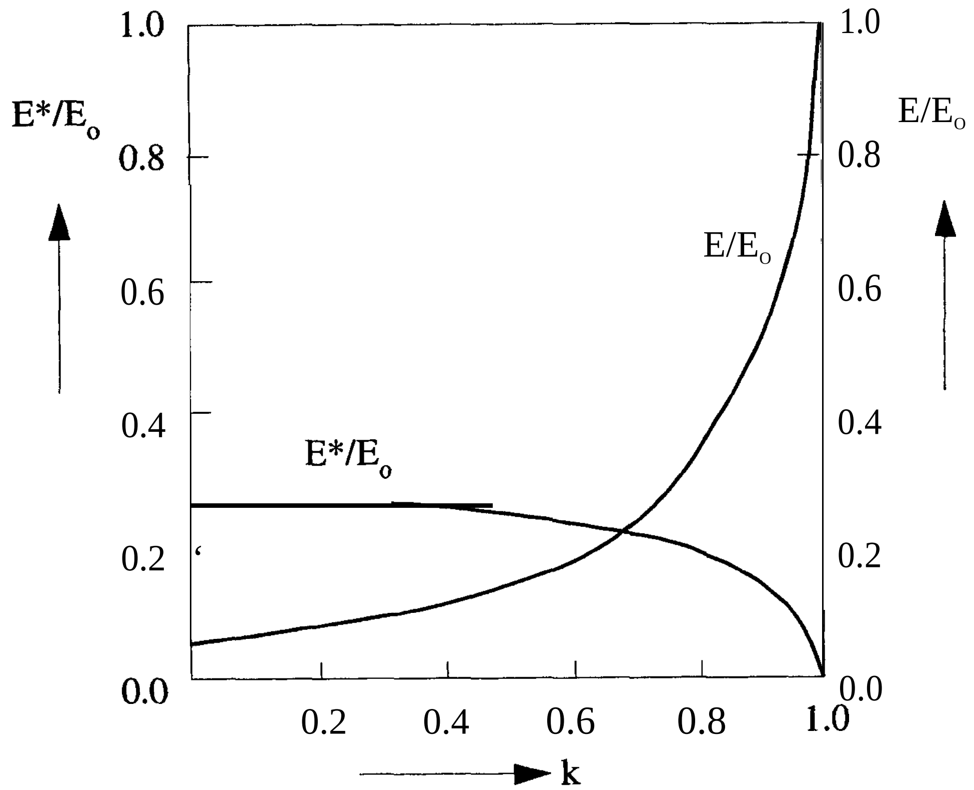


Fig. *7* ***Input energy E' and output energy E of the subcritical system, as functions of the multiplication factor k.***
E_0 *is* ***the energy output of one CANDU***

Figure 7 clearly shows that the real gain in efficiency is obtained with k very close to **unity.** Although the accelerator-driven system is promoted for its inherent safety, it should be realised that there will be a strong motivation to put up its reactivity as far as possible, rendering the system into a nearly critical system with safety problems of the same order as those of nuclear reactors. **In** connection with these safety aspects it should be noticed that - even if the system is designed such that it will remain subcritical under all possible circumstances - it cannot be considered as an inherently safe system. Reactivity-initiated reactor accidents form only a small **part** of all possible accidents. In most accidents decay-heat removal is of utmost importance and problems with this decay-heat removal are equally relevant for critical reactors and for (slightly) subcritical systems.

3. CONCLUSION

It is difficult to find a sound motivation for the transmutation of fission products with accelerator-driven systems. If there would be any hesitation in transmuting fission products in nuclear reactors, there would be an even stronger hesitation to use accelerator-driven systems, mainly because of their lower energy efficiency and their poor cost effectiveness. The use of accelerator-driven systems could become a 'meaningful' option only if nuclear energy would be banished completely. To make the option economically more attractive, the system will very much look like a (slightly subcritical) reactor, with the same safety-related problems.

We arrived at these conclusions by an analysis of scenarios for thermal, heavy-water moderated, systems. The conclusions are quite general, however, and apply equally well to other types of reactors and moderators.

REFERENCES

[1] K. Abrahams, Minimizing the Integrated Collective Radiation Dose and the Transmutation of Long-Lived Nuclear Waste.
Proc. Specialists' Meeting on Accelerator based Transmutation at PSI, Villigen, 24-26 March 1992. PSI-Proceedings 92-02, p. 619.

[2] C.D. Bowman et al., Nuclear Energy Generation and Waste Transmutation using an Accelerator-Driven Intense Thermal Neutron Source.
Nucl. Instr. and Meth. in Physics Research A320 (1992), p. 336.

[3] A.R. Dastur (AECL), Private Communication, May 1990.