CRITERIA FOR ACHIEVING ACTINIDE REDUCTION GOALS

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Abstract

In order to discuss various criteria for achieving actinide reduction goals, the goals for actinide reduction must be defined themselves. In this context the term actinides is interpreted to mean plutonium and the so called "minor actinides" neptunium, americium and curium, but also protactinium. Some possible goals and the reasons behind these will be presented.

On the basis of the suggested goals it is possible to analyze various types of devices for production of nuclear energy from uranium or thorium, such as thermal or fast reactors and accelerator driven systems, with their associated fuel cycles with regard to their ability to reach the actinide reduction goals.

The relation between necessary single cycle burn-up values, fuel cycle processing losses and losses to waste will be defined and discussed. Finally, an attempt is made to arrange the possible systems on order of performance with regard to their potential to reduce the actinide inventory and the actinide losses to wastes.

The actinide series of elements begins with actinium (element 89) and ends with lawrencium (element 103). Only the first four of these elements (actinium, thorium, protactinium and uranium) could be found on Earth in significant activities or concentrations before World War II. The development of the nuclear reactor and its widespread use as a large source of electricity have led to large scale synthesis of the other lower members of the actinide series, i.e., neptunium, plutonium, americium and curium. "Actinides" will in the following be used to refer only to the mentioned lower members of the actinide series. Some of these have found use, e.g., plutonium as a complementary nuclear fuel, as an energy source in space research, and as a constituent in nuclear weapons, and americium as radiation source in smoke detectors. However, at present the production is much larger than the consumption leading to increasing stockpiles, increasing amounts in stored spent nuclear fuel and as radioactive waste from reprocessing operations. Improved reprocessing technology might in the future reduce the amounts of actinides in the radioactive waste leading to an increased growth of stockpiles. In this situation means are sought to reduce production rates and to reduce stockpiles of these elements.

In general, there are two ways to reduce the amounts of actinide by: i) minimization of actinide generation in nuclear power reactors, and ii) destruction of actinides generated in nuclear power reactors. The last option is also frequently referred to as transmutation. The available means are:

i) changes in reactor operation modes,
ii) changes in fuel design,
iii) changes in fuel composition,
iv) changes in the nuclear fuel cycle,
v) introduction of improved (or novel) types of power reactors, and
vi) development and use of accelerator-driven subcritical power reactors.

At present, however, generation and utilization of plutonium is usually maximized for economic reasons. E.g., in a typical swedish BWR more than 30% of the energy comes from burning of the plutonium formed by neutron capture in ^{238}U.

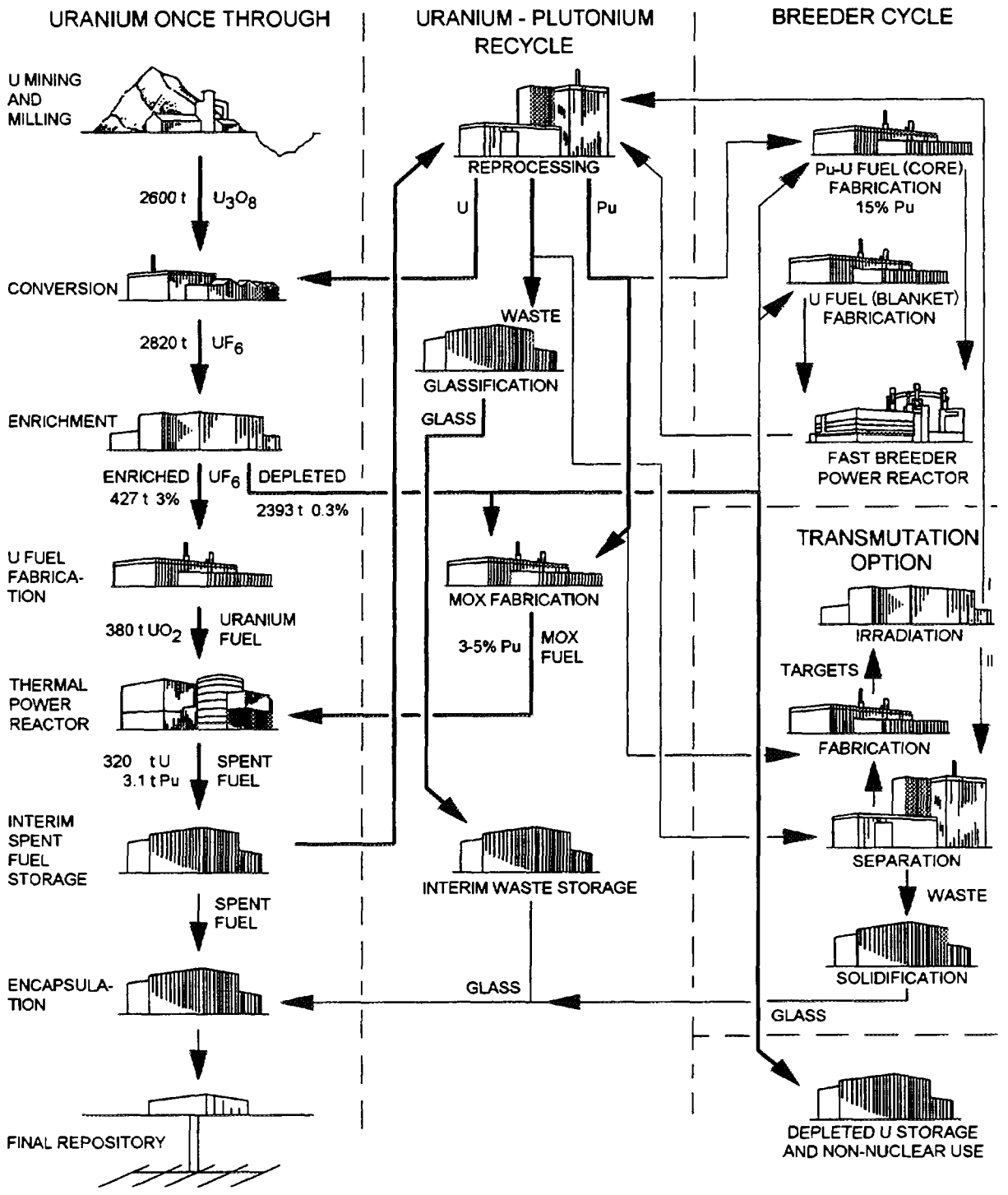

FIG. 1. The nuclear U/Pu fuel cycle with possible variations and extensions.

A trend towards thinner fuel pins in BWR and PWR reactors makes it possible to achieve a higher burn-up by increased conversion of ^{238}U to plutonium, partly compensating the consumption of ^{235}U, and burning of part of the plutonium produced. As plutonium is mainly formed (and burned) in the outer radial zone of a fuel pin, thinner pins yields a relatively higher percentage of plutonium in the spent fuel than thicker pins. However, the accompanying increase in the possible burn-up should in general more than compensate for the higher plutonium concentration in spent fuel. Thus one would expect that thinner fuel pins leads to less spent fuel, but with an increased plutonium content. The combined result could be a decrease in the total amount of plutonium in spent fuel from production of a given amount of electricity.

MOTIVES FOR A REDUCED ACTINIDE GENERATION

Why should actinide generation be reduced? Possible motives could be:

i) A reduced world inventory of plutonium. This could reduce the risk of proliferation. However, with this motive the incentive to reduce inventories of the other actinides should be rather small.

ii) A smaller potential hazard from long-lived nuclides in the final repositories for radioactive waste. In this case the are two different situations; final storage of unreprocessed spent fuel and storage of high level waste from reprocessing. With regard to storage of unreprocessed fuel there is only a very small reduction in actinide content possible. In the case of reprocessing waste, the actinide content depends mainly on the separation process used and its efficiency. However, in order to avoid a large inventory of actinides and corresponding cumulative losses to wastes it is also necessary to achieve a high degree of destruction of actinides during each recycle.

iii) Purely political. These motives could be e.g. to increase public acceptance of nuclear power by removing one of the arguments against its use. In this case we must ask if e.g. subcritical accelerator-driven reactors would be perceived as more acceptable than normal critical reactors, or if the Th/U-based fuel cycle is more acceptable by the general public than a U/Pu-based fuel cycle. An other argument sometimes presented is to reduce the possibility of a future large scale use of nuclear power by forcing a less efficient use of the available uranium resources. However, in this case it must be assured that spent fuel repositories will remain inaccessible for ever.

THE EFFECT OF MINOR CHANGES IN FUEL DESIGN

What can be done with present light water reactors running on the U/Pu cycle? In case of BWRs some difference in actinide generation can be obtained by running the reactors at higher or lower void. It is also possible to vary the fuel pellet diameter in LWRs in order to affect the actinide concentration in spent fuel. The flux depression in the ^{238}U resonance capture region causes most conversion to occur near the pellet outer radius. Autoradiographs and other examination methods confirm that most of the plutonium is found in a thin layer near the pellet surface. Hence, a smaller pellet diameter yields a higher average concentration of plutonium than a larger diameter. This can be expressed as follows;

$$m_{Pu}/m_{fuel} = 4\, k_{Pu}\, a\, (d - a)\, d^{-2} + k_{Pu\text{-}avg}$$

where d is the pellet diameter, a is the zone thickness where most of the Pu is present, k_{Pu} is the excess concentration of Pu in the Pu-rich zone and $k_{Pu\text{-}avg}$ is the average Pu concentration in the pellet. The constants k_{Pu} and $k_{Pu\text{-}avg}$ depend of course on the reactor, operating conditions, burn-up, etc. If $a = 0.5$ mm and $k_{Pu} \gg k_{Pu\text{-}avg}$ are assumed and all other factors remain constant, a change from 10 mm pellets to 8 mm pellets would increase the concentration of Pu in the spent fuel by a factor ~ 1.23. However, a larger diameter most likely results in a fuel with a lower burn-up. This would result in more spent fuel for a given energy production, albeit with a smaller concentration of Pu, which might remove the gain. Careful calculations are needed in order to find an eventual optimum.

TABLE I. Comparison of calculated plutonium and minor actinide amounts ($g/GW_{th}d$) in spent fuel from reactors operating at 1000 MW_e and a load factor of 0.8 (based on data from [1], [2] and [3]).

	BWR	PWR	LMFBR	THTR
Protactinium	2.6×10^{-6}	1.0×10^{-5}	–	**0.21**
Neptunium	11	14	7.7	**24**
Plutonium	288	334	**3030**	18.6
Americium	3.8	5.1	**28**	0.5
Curium	0.79	**1.3**	0.64	0.24

The LMFBR (U/Pu cycle) and THTR (Th/U cycle) are assumed to operate in breeding mode.

THE EFFECTS OF REACTOR TYPE AND FUEL

What about differences between reactor types and fuels? Table I gives calculated amounts of actinides in spent fuel from four reactor types normalized to 1000 MWe and a load factor of 0.8. As can be seen, the largest concentrations of protactinium and neptunium are expected for a THTR operated in breeding mode on the Th/U cycle, the largest plutonium concentration for a LMFBR operated in breeding mode on the U/Pu cycle, and the largest curium concentration for a PWR operated on enriched uranium fuel.

FUEL CYCLES

Figure 1 illustrates the current and future options for the U/Pu fuel handling. The left part shows the activities involved with the once-through fuel option, the center part shows the activities and options involved in U/Pu recycle, the right upper part indicates the additional activities for a breeder cycle, and the right lower part illustrates the additional operations needed in case a transmutation option for actinides would be established. The Figure also indicates where reprocessing and separation processes have to be used. In this context it is important to understand the possibilities and limitations of these processes. The following facts are important:

i) The fuel/target material after irradiation is the input to the separation process.

ii) The process separates chemical elements regardless of their isotopic composition (isotope separation is possible, but costly, and must be added after the chemical separation).

iii) The primary output of the process is either new fuel/target material in the proper chemical/physical form or raw material to a separate fuel/target fabrication process.

iv) The secondary output from the process is waste for direct disposal or input to a separate waste treatment process.

TABLE II. Some typical losses (as fractions of input and % of input) in separation and fabrication.

	Reprocessing of spent fuel					
	standard PUREX		with modified PUREX process		TPu recovery process added	
Pa	1	100.%	1	100.%	1	100.%
U	0.005	0.5%	0.005	0.5%	0.001	0.1%
Np	1	100.%	0.05	5.%	0.01	1.%
Pu	0.003	0.3%	0.003	0.3%	0.001	0.1%
Am	1	100.%	1	100.%	0.01	1.%
Cm	1	100.%	1	100.%	0.01	1.%
	MOX and target fabrication					
	All actinides		0.0001	0.01%		

TOTAL LOSSES TO WASTE

Reprocessing/separation processes always have some loss of actinides to waste streams. Table II summarizes some typical data for existing reprocessing operations and for future extended actinide recovery operations. The Table also gives some data on typical losses in MOX fabrication and in target fabrication for actinide burning. All values, except for standard PUREX and MOX fabrication, are estimates.

Recycling of actinides can in principle be divided into two cases; co-processing and separate processing. In co-processing the irradiated recycled material is mixed with spent un-recycled fuel and is treated in a common reprocessing plant. By following a unit mass of any actinide through the reprocessing plant, fabrication of targets from recovered material, new irradiation, and back to the reprocessing plant, cycle after cycle, until all material is transmuted after an infinite number of recycles it is possible to compute how much of the original material is lost as waste. This type of recycling is shown schematically in the upper part of Figure 2 for the first cycle. The total fraction of any element, originally present in the spent fuel, which is lost to waste, w, is given by:

$$w = a[\sum_{n=0}^{\infty}(1-a)^n(1-b)^n\{\prod_{m=0}^{n}(1-c_m)\}] + b[\sum_{n=0}^{\infty}(1-a)^{n+1}(1-b)^n\{\prod_{m=0}^{n}(1-c_m)\}]$$

where a, b, and c_m are the fraction of element lost to wastes in reprocessing, in target fabrication, and the fraction burned/transmuted in recycle pass m (c_0 = 0), respectively. As can be seen, it is important in this case to keep the losses to waste in reprocessing very low and the fraction transmuted per recycle high. Otherwise a considerable fraction of the initial amount of any recycled element will ultimately become waste.

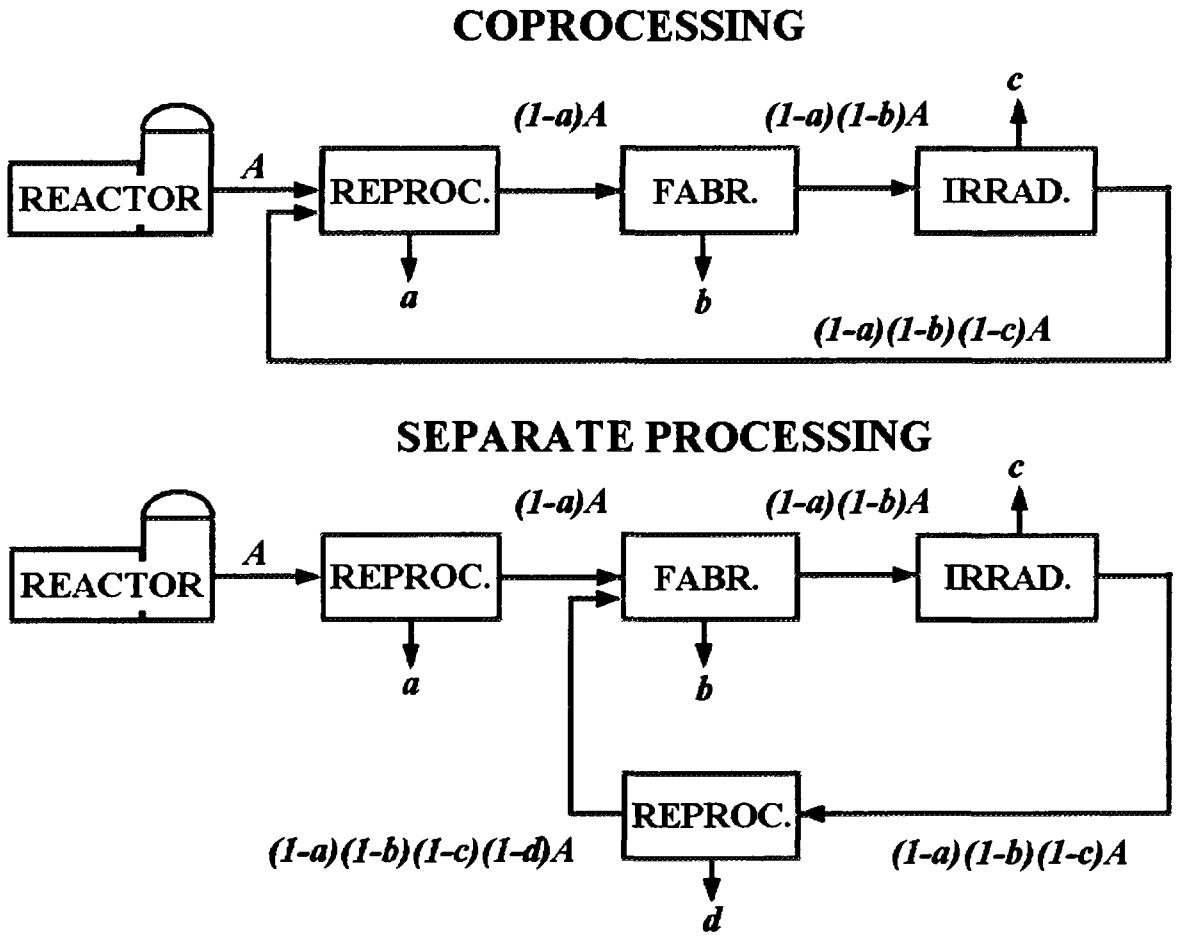

FIG. 2. Recycling options and fractions lost to waste in various operations.

When a separate plant is used to process/separate irradiated recycles material, as indicated in the lower part of Figure 2, the total fraction lost to waste after an infinite number of recycles is given by:

$$w = a + (1-a)[b\sum_{n=0}^{\infty}(1-b)^n(1-d)^n\{\Pi_{m=0}^{n}(1-c_m)\} + d\sum_{n=1}^{\infty}(1-b)^n(1-d)^{n-1}\{\Pi_{m=0}^{n}(1-c_m)\}]$$

where d is the fraction lost in the separation process. In this case the losses in normal reprocessing only occur once as an important term in the equation. Thus, slightly larger losses can be tolerated than in the case of co-processing. However, also in this case it is necessary to achieve low losses in the separate processing and manufacturing processes as well as a high burn-up in each recycle.

For a simple demonstration we can assume that the same fraction is transmuted in each irradiation, i.e. c_m = constant = c. After simplification with the assumption that a, b, c, and d are all $\ll 1$ we obtain the approximate equations:

$$w \approx (a + b)/(a + b + c) \qquad \text{for co-processing}$$

and

$$w \approx a + (b + d)/(b + c + d) \qquad \text{for separate processing}$$

For a meaningful destruction we must in both cases require that the denominator is dominated by c.

If we consider co-processing using a modified PUREX process with an add-on TPu separation process as an example, Table II gives the following data for neptunium: a = 0.01 and b = 0.0001. For a total loss to wastes of not more than 5% of all Np we must transmute at least 19.2% of Np in each irradiation.

Consider instead the case of separate processing after a modified PUREX with the same data as above. Furthermore, assume that the separation process only has a 0.5% loss of Np to wastes. In

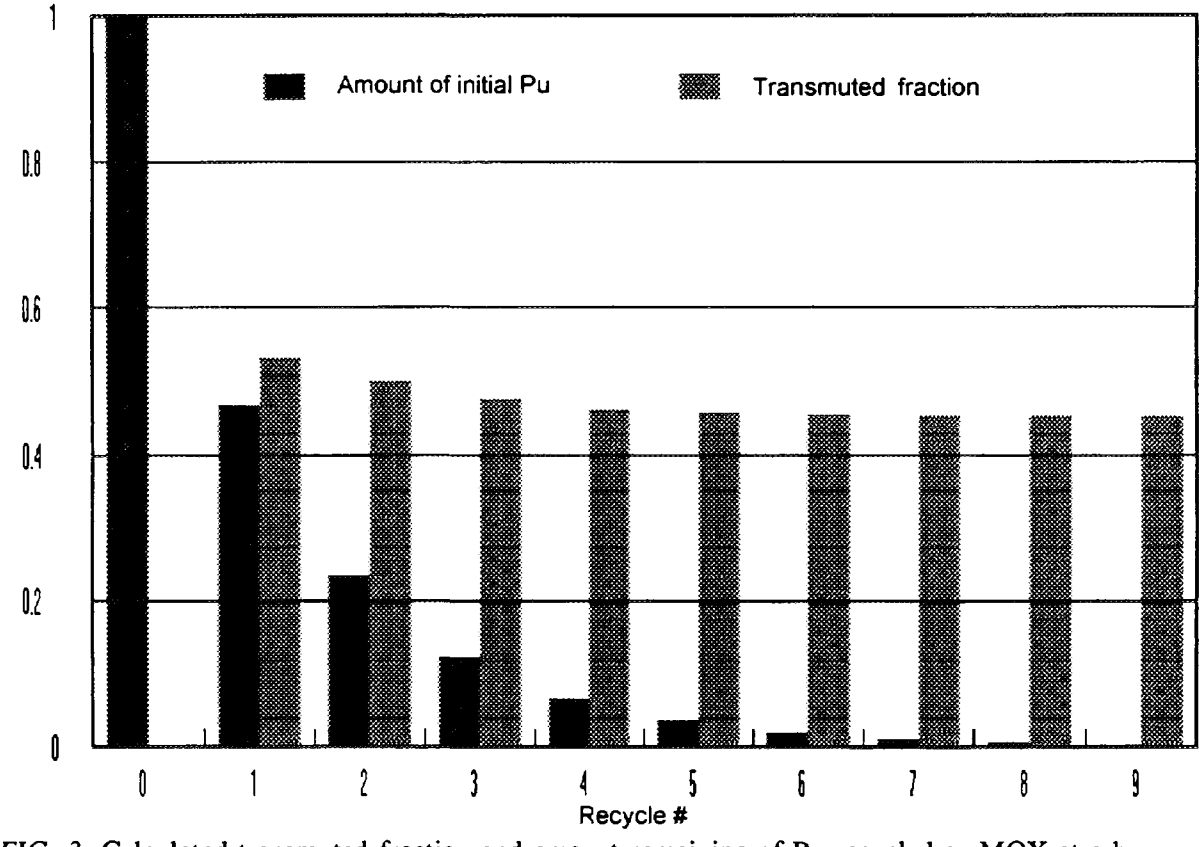

FIG. 3. Calculated transmuted fraction and amount remaining of Pu recycled as MOX at a burn-up of 33 MWd/kg.

this case we would require a transmutation of at least 12.2% to keep a loss of Np to all wastes of 5% or less.

Are such high transmutation fractions achievable? A simple calculation of the transmutation of plutonium when recycled to a light water reactor as MOX with 5% Pu and a burn-up of 33 MWd/kg gave the data shown in Figure 3. As can be seen, the transmuted fraction decreases somewhat with the number of recycles due to a changing isotopic composition of the recycled Pu. However, the single cycle transmuted fraction seems to be large enough to make a nuclear incineration feasible with rather small Pu losses to wastes.

THE TOTAL INVENTORY

If we denote the turn-around time of one recycle for the co-processing alternative by t_c, and the amount of an actinide fed to the process during t_c by A_c we can calculate the total inventory, M_c, of that actinide being circulated at equilibrium from the following equation.

$$M_c = A_c \left(1 + \sum_{n=1}^{\infty} (1-a)^n (1-b)^n \prod_{m=0}^{n-1} (1-c_m)\right)$$

If we now make the simplification that c_m = constant = c and use the data from the previous co-processing example (a = 0.01, b = 0.0001, and c = 0.192) the total circulating Np inventory would be about 5 times the amount of Np entering the recycling process during t_c.

CONCLUSIONS

The following general conclusions might be drawn regarding the available options to reduce actinide generation and actinide inventory:

1 - Countries who intend to store unprocessed spent fuel in a final repository.
- Only marginal interest in actinide reduction methods which involve reprocessing.
- Maybe BWRs are marginally better than PWRs with regard to actinide generation.

2 - Countries who accept reprocessing of spent fuel but desire to keep to current reactor types.
- Only recycling/burning as MOX fuel or special target pins is of interest.

3 - Countries who accept reprocessing of spent fuel and "new" reactor types.
- The Th/U fuel cycle has some benefits, but probably requires "new" reactor types and certainly requires new reprocessing facilities.
- FBR technology is also of interest as such reactors can be used in burner mode.

4 - Countries who have political problems with poor acceptance of current light water reactors.
- The idea of accelerator-driven systems might be more acceptable.

5 - Reduction of stock-piles of weapons-grade material.
- High enriched uranium fuel could be used, which would lead to minimal production of transuranics. However, this may pose another safeguard problem.
- ^{239}Pu could be used as MOX fuel in available power reactors or in special Pu burners, e.g. ATR or FBR.
- Accelerator-driven systems may also have some benefits as Pu burners.

6 - Most important
- To develop reprocessing and separation technology towards smaller losses to wastes.

- To be wary of new processes not proven on an industrial scale (remember the Mid-West Fuel Recovery Plant).
- To develop target materials for high burn-up and suitable for the separation process.

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