CHAPTER III

MINOR ACTINIDES AND LONGLIVED FISSION PRODUCT CONDITIONING AND TRANSMUTATION

3.1. MINOR ACTINIDES. CONDITIONING AND FUEL FABRICATION

3.1.1. Homogeneous recycling processes

In order to assess the magnitude of the MA conditioning operations including : chemical purification, conversion to oxide, encapsulation and incorporation into fuel elements, it is necessary to quantify the total inventory of MA produced by reprocessing operations. By combining the data of table II and IV of Chapter I, the total production of MA resulting from the planned reprocessing facilities in OECD countries can be assessed.

Table III-1 gives an overviewof the data.

Table III-1.
Production capability of minor actinides in reprocessing plants as constructed or planned in OECD countries.

Nuclide	Specific yields g/THM	M.A. Annual production in kg/year		
		La Hague UP2 + UP3	Sellafield THORP	Tokai (Rokkasho)
Np 237	437	700	524	131(349)
Am (241 + 243)	380	608	456	114(304)
Cm (242-247)	1.5	2.4	1.8	0.45 (1.2)
Resid Pu(0.7%)	65	104	78	19.5(52)

3.1.1.1 Neptunium recycle

As already explained in Chapter II, the recovery of Np from reprocessing product-and waste-streams is feasible if certain improvements are introduced in the liquid extraction PUREX process. If this process scheme could be realized the Np stream will display more or less the same degree of contamination as the U-PU products. Under these circumstances the fuel fabrication plants could accept moderate quantities of Np in their PWR-MOX sections since Np is a pure alpha emitter.

The quantities of Np which could be mixed with Pu are controlled by reactor physics considerations and by the final Pu 238 concentration in the spent fuel element. For reasons of heat dissipation the Pu 238 concentration is limited to 5 %. An initial enrichment of 2.5 to 3.5 % is acceptable for FBR-MOX fuel [3.1].

The quantities which might be mixed with PWR-MOX fuel depend on the enrichment with U and Pu to counterbalance the initial antireactivity of Np. During the irradiation, Np transforms into Pu 238 with a higher fissile cross section. The initial Np concentration will not exceed 1 % in the PWR-MOX fuel assemblies (30 % of the total PWR-core). [3.2].

From the quantities of Np produced by the reprocessing plants and taking into account the acceptable concentrations in fuel assemblies, we can deduce the MOX production capacity required to treat the potentially upcoming Np flux. The total amount of Np to be produced in the operational and planned reprocessing facilities amounts to 1,700 kg/year. Taking a 2 % loading in PWR-MOX fuel and 2.5 % in FBR-MOX fuel a capacity of 85 THM respectively 68 THM will be involved with U-Pu-Np fuel fabrication activities.

Since the Np recovery is not yet operational in the reprocessing plants a gradual introduction of Np into the PWR-MOX fuel fabrication plants could be envisaged in three stages.
A pilot stage during which the technological problems are investigated (preferable Np fuel type, mixing of powders, homogeneity of Np distribution in the fuel . ..) followed by a semi industrial activity centered around one

reprocessing and one fuel fabrication plant. The third stage being the industrial one, requires a MOX fuel production capacity of 85 T/year. This capacity will be available at the turn of the century in Europe and Japan. Preferably the U-Pu-Np MOX fuel fabrication activity could be inserted into the existing industrial units or concentrated into one or two especially designed facilities.

However it has to be kept in mind that once Np 237 has been added to the fresh fuel, a second recycling of the MOX spent fuel containing 3 to 5 % Pu 238, into the conventionally built MOX fuel fabrication plants, becomes much more difficult because the higher heat and neutron emission which will accompany the increased Pu 238 content, will interfere with conventional glove box manipulations.

Repeated recycling of PWR-MOX fuel in a thermal reactor type is a practice which looks difficult to accomplish any way because the reduction in fissile isotope content (Pu 239+241) is significant [3.3]. Table II I-2 shows the data for PWR-MOX fuel during the first, second and third recycle without external Np addition.

Table III-2

Percentage Isotopic content of recycled spent PWR-MOX fuel

(data taken from [3.3])

Nuclide	Pu 238	Pu 239	Pu 240	Pu 241	pu 242
First recycle	2.5	49.7	27	16.2	7.1
Second recycle	3.1	44.6	38.7	17.2	9.5
Third recycle	3.7	42.1	29.4	17.4	11.1

Spent fuel from first recycle MOX fuel, can in principle be reprocessed in conventional reprocessing plants if homogeneous “dilution” of PWR-MOX fuel elements among conventional PWR-UO_2 fuel elements is accepted by the safety authorities.

Industrial amounts of spent PWR-MOX fuel elements can preferably be reprocessed in specially designed facilities which cope with the increased criticality risk and the reduced Pu volubility issue.

The buildup of higher isotopic mass concentrations of Pu in the recovered Pu streams from first and subsequent recycle operations will diminish its fissile worth in thermal reactors.

Economic reuse of "second recycle" Pu requires almost unavoidably a FBR scheme by which not only the uneven but also the even numbered Pu masses become fissionable. This problem will be further treated in more detail in section 3.2 on transmutation.

Alternatively to this advanced management scheme spent fuel from last recycle origin will have to be stored any way in a suitable repository which is the opposite to the primary goals of any P & T option.

3.1.1.2. Americium- Curium recycle

Due to the very nature of the radioactive alpha-beta-gamma emission of Am and Cm actinides, their processing in a conventional fuel fabrication plant is excluded.

In this section we will hypothyze that the Am-Cm fraction can be extracted from HLLW and purified up to a sufficiently high purity grade in order to use it as oxide in MOX fuel. Up to now this basic assumption has not yet been realized and may well take a number of years before a neat process will become available.

The very high similarity between Trivalent Actinides and Rare Earths will make the separation difficult and consequently expensive. According to present day technique, separations from HLLW lead, even on laboratory scale, to Actinide-Rare Earth (RE)- mixtures. In order to assess the problems involved, three levels of separation will be supposed: Actinides + 50 % RE fraction; Actinides + 10 % RE fraction; Chemically pure Actinide fraction (≃ 99 %). The corresponding compositions are given in table III-3.

Table III-3.
Composition of Am-Cm fractions contaminated with R.E.
(5 y after discharge)

	g/THM	Ci/THM	Watt/THM
Am + Cm + 50 % RE fraction	400 5119	2663 40370	91.3 97
Am + Cm + 10 % RE fraction	400 1023	2663 8074	91.3 19.4
Am + Cm 1%RE fraction	400 (loo)	2663 (800)	91.3

According to table III-1 the total potential of Am - Cm produced by reprocessing plants in OECD countries amounts to 1,490 kg /year which would be available for a comprehensive P & T option. The fuel fabrication plant which would receive a mixture of Am + Cm + RE 50 % ought te be designed as an annex of the reprocessing plant since the RE activity is about 15 times higher than that of the Am - Cm fraction. The use of such a hypothetical mixture as the basis for transmutation of Am - Cm would lead to very high neutron consumption in thermal reactors.

Even in the case of the most optimistic purification of Am - Cm/RE-l % the radioactivity due to strong gamma emitters eg Ce144 will require heavy shielding, remote handling and maintenance...

The chemical and metallurgical processing of such mixtures is difficult but not impossible. The-preparation of isotopic heat" sources based on Cm 244 is a good example of what is required to implement such an option, but upscaling to industrial quantities is problematic.

Systematic research has been done by the Trans Uranium Institute at Karlsruhe on the development of FBR fuel with MA incorporation and on the irradiation behaviour of different fuel types [3.4].

The minor actinides Am-Cm would make up 2 % of the FBR driver fuel if homogeneous recycling is considered. About 800 kg of Am-Cm mixture could

this way be mixed with the FBR core fuel of a 1 GWe FBR power plant. A fuel fabrication capacity of 75 THM/year would be required to transform all Am - Cm produced by OECD reprocessing plants (= 1500 kg per year) into a U-Pu- 0.02 Am - Cm fuel form. Such a plant located at a reprocessing site, could receive prepurified Am - Cm solution from other reprocessing plants and carry out a final purification step to homogenize the composition especially the RE concentration. Transfer of highly active liquors between different countries is yet a difficult endeavour for which very strict exchange rules are to be established in order to avoid discussions about waste- and fissile material property rights and ultimate responsibility. The preparation of MOX fuel by successive oxalate and/or ammonia precipitation, followed by drying, sintering and pressing is by itself standard technology operational in many fuel fabrication plants.

The aqueous part of the fuel preparation process must be housed in a separate building in which only non fissile M.A. are processed. Once the Am-Cm (RE) fraction has been transformed into a dry powder, transfer to the fuel manufacturing plant becomes acceptable. However the greatest uncertainty lies with the introduction of the remote handling- and maintenance technology on processes which up to now have been done manually. A second factor to be assessed thorougly is the heat output by MA on the process equipment and materials (gloves, plastic structures etc...).

Homogeneisation of master blends and other powder mixtures, takes place in relatively large powder mixers which have to be designed specially to cope with increased heat dissipation; the use of water as coolant being excluded for reasons of criticality risk.

Some proposals have been made to incorporate much higher MA concentrations i.e. 10 and 15 % in addition to the already accepted 2 % Am 241 level. [3.5]. These high MA levels would involve the introduction of 4500 kg and even up to 6730 kg MA in one FBR core. The heat emitted by such fuel mixtures, essentially due to Am 241, amounts to 0.5 MWth resp. 0.76 MWth. The question arises whether such large heat outputs can be tolerated in a fuel fabrication plant with industrial size and aim.

Increasing amounts of heavier actinides will also increase the neutron emission due to the α-n reaction in the fuel matrix and due to spontaneous fission of some actinides (Bk,Cf) associated and coprocessed with Am and Cm fractions.

A thorough analysis should be made, e.g. on the basis of a conceptual design exercice, of all the new aspects of fuel fabrication involving MA and especially Am 241 before any reliable evaluation can be made of the cost increase associated with this new option.

The radiological impact of MA recycling depends on the capital expenditure allocated to this operation but will be similar to the vitrification process of HLLW or to reprocessing activities in general.

3.1.2. Heterogeneous recycling processes

The specifications of the fuel and the fuel pins underlie the strong limitation of MA concentration in MOX fuel as well for LWR as FBR's. Heterogeneous recycling of MA involves the use of specially designed fuel pins and assemblies which contain a much higher MA concentration per unit length of fuel pin.

The first step in this direction is the '"development of MA-fuel mixtures with a high specific content of each of the nuclides concerned: Np, Am, Cm. The handling of Np does not raise particular problems and could be performed in classical glove-box-type facilities.

For Am and Cm, as already discussed, special handling techniques have to be developed to cope with the strong neutron; gamma-and heat emissions. Sol-gel techniques developed for $(U\text{-}Pu)O_2$ fuel [3.6] are very well suited for these purposes but will have to be further improved and upscaled to be useful at the 100 - 1000 kg/year fuel fabrication level. Remote handling techniques coupled to remote maintenance of the facility are to be implemented in pilot scale installations before any industrial activity is conceivable [3.7]. Vibrocompaction of fuel particles is an alternative way to fill fuel pins if the classical pellet pressing, sintering and grinding

sequence can not be used. The same technique is also applicable to dilu-
'ion '[ith] '[0]2' '[1]2[0]3' '[r0]2 'r 'e" [3.8] in order to improve heat dissipation.

At the laboratory level most of the fuel fabrication work involving MA has been carried out at the Trans Uranium Institute of Karlsruhe. [3.4] [3.9]. Work was concentrated on the preparation of small samples for irradiation in several prototype fast reactors (KNK II, PFR and PHENIX)
Pellets with 50 % NpO_2,Am_2O_3 were diluted with natural UO_2 in order to reduce the local heat-flux and prevent melting. Undiluted samples have to be irradiated in the blanket of a FBR.

Similar work at slightly higher activity level was reported byHEDL. [3.10]

Segmented pins were designed for incorporation of a number of actinide isotopes to be irradiated in PFR. Specifications for pellet specimens were developed on technical grounds which guarantee their compatibility with nuclear fuel oxides. The pellets were hot pressed and handled in very special remote handling facilities at ORNL equiped for work with high neutron emitting materials and isotopic heat sources. Dose rates around a $^{244}Cm_2O_3$ pellet amount to 6 Sv/h essentially due to neutrons from the (n,α) reaction. Work with 241 Am_2O_3 pellets revealed to be much easier since only ^{7}irradiation was to cope with.

A very important experiment for the demonstration of the heterogeneous recycling of MA has been undertaken by the joint TUI-CEA institutes under the acronym SUPERFACT [3.11].
The fuel was prepared by Sol-Gel method and the resulting microsphere were compressed into pellets. Two pins contained a mixture of 44.8 % Np and two were loaded with 21 % Np + 18 % Am.

The capsule is a standard PHENIX capsule, composed of 19 pins. The pins are 1793 mm long with ID of 5.65-6-58 mm, some pins intended to be experimental examples of heterogeneous recycling, have a fuel column of 400 mm long. In the same subassembly also homogeneous recycling of pins with 1.5 % Np and 1.8 % Am was demonstrated as reference. Irradiation took place

between Oct. 1986 and January 1988 and reached a burnup of 8.5 at% after 383 days of Eff. Full Power. Post irradiation examination took place in 1991 and did not reveal particular problems except for very high He pressures encountered in some Am pins. Experimental analysis of Pu 238, higher Actinides and Fission products is currently under investigation at CEA-FAR.

One of the major advantages of heterogeneous recycling of MA is the possibility to produce fuel pins along a specially designed fuel fabrication process in tailor made, relatively low throughput, fuel fabrication facilities. However the transmutation capacity of "lumped" fuel in separate fuel assemblies ought to be compared with the homogeneous recycling in FBR's particularly the overall burnup and recycle frequency.

Metallic type irradiation targets for MA irradiation in high flux reactors (HFR, BR2, OSIRIS, SILOE...) with flux levels of 2-5 10E14 n/s/cm^2 have been developed some decades ago in the frame work of the Pu 238 production. [3.12]. The technology to transform Np solution into Np-Al alloys is standard technical level for handling 100 to 500 g of Pu 238. In this case special attention must be given to the heat problem and to the integrity of irradiation capsules. If long term irradiations (one to several years) in a high neutron flux are required to reach a sufficiently high degree of target nuclide depletion the appropriate measures must be taken : to insure heat dissipation of the capsule or assembly, to cope with He buildup and to protect the reactor from contamination by double containment. However for industrial programmed on MA transmutation very large facilities would be required capable of irradiating 10 to 100 kg of each MA per year. Particularly for Np transmutation into Pu 238 this might be an interesting approach.

Irradiation subassemblies ought to be developed containing about 10 kg of MA each with an appropriate metallic diluant. These subassemblies could be irradiated during an extensive time period to deplete the target isotope with a factor 10 at least.

Recently the pyrometallurgical processing of metallic fuels for the Integral Fast Reactor (IFR) received considerable attention [3.13].

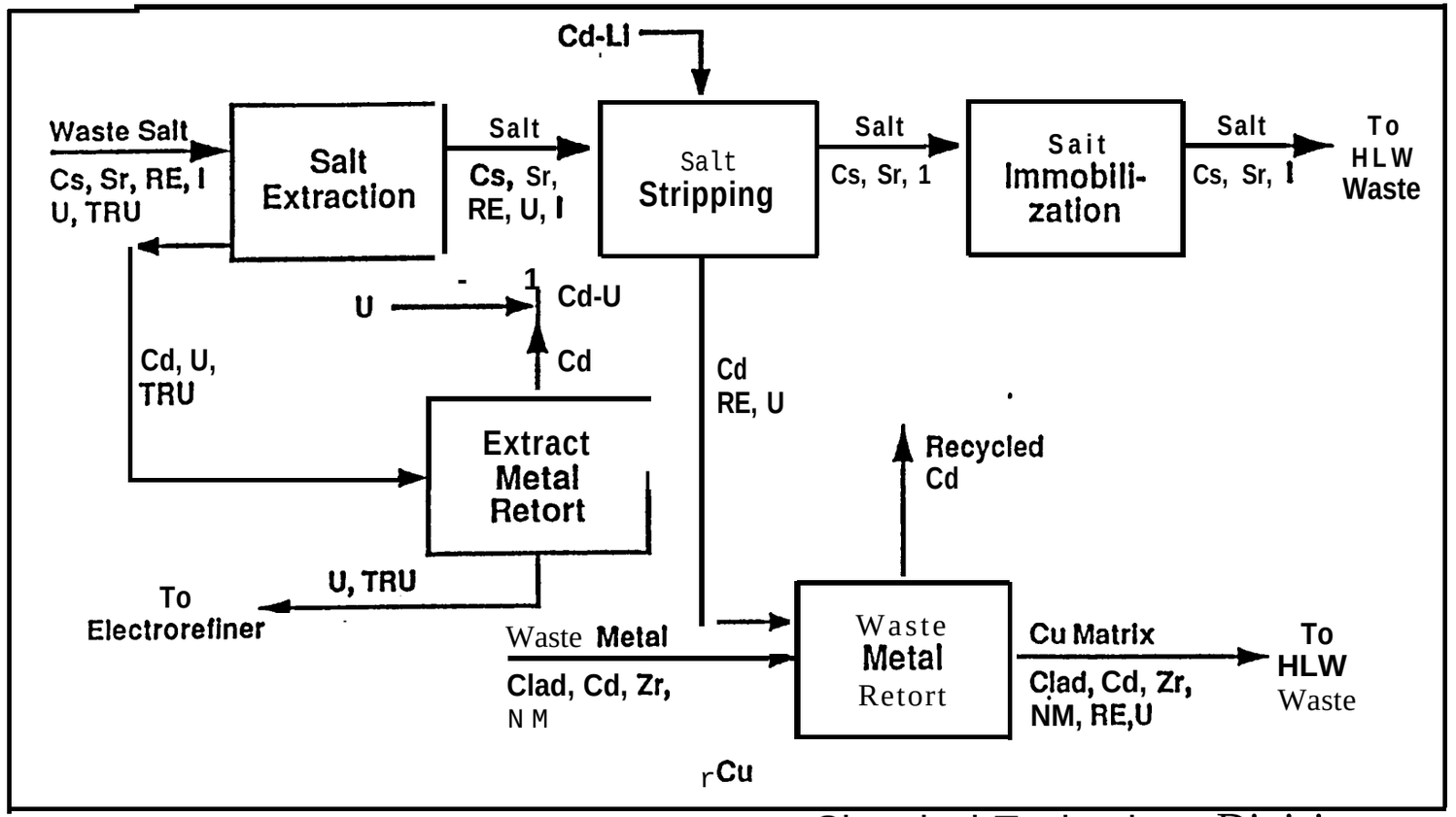
This process involves electrorefining of spent fuel with a Cadmium anode, solid and liquid cathodes, and a molten salt electrolyte (LiCl - KCl) at 500°C. The process employs a metallic fuel alloy of U-PU and Zr, clad with stainless steel. Due to the newness of this approach a some more detailed description follows:

The heart of the flowsheet is a pyre-electrochemical dissolution process of declad spent IFR fuel which is to be recycled on- site into fresh IFR fuel. The declad spent fuel is put into a basket and dissolved in a molten salt bath. After dissolution of the fuel $CdCl_2$ is added to the bath to oxidize alkali, alkaline earths and Rare earths elements which enter the salt layer. Pure U (free of Pu) is transported to the solid Cd cathode. A mixture of U-PU is electro transported to a second but liquid Cd cathode. During the electro refining process the MA follow the U-PU stream and need to be further separated for recycling.

A parallel and similar process has been developed for the recycling of Actinides from spent LWR fuel into a IFR for further incineration. [3.14]; the process is rather complicated and involves the following steps: reduction of UO_2 by electroreduction with Ca metal in a molten salt bath of $CaCl_2$- 20 % CaF_2; a molten alloy of Mg 42 % Cu is in contact with the molten salt: U precipitates in the metal CuMg bath and can be filtered off, Pu, Actinides and Rare Earths dissolve in the Cu-Mg alloy; TRU and Rare earths are extracted by molten $MgCl_2$ salt bath from the MgCu bath into a Zn-Mg bath; the TRU-Rare Earth fraction is separated from excess Zn-Mg alloy by heating in a retort; the separated TRU-Rare earth fraction is transferred to the electrorefiner for further purification and refabrication of IFR fuel elements. The conceptual flowsheet of the salt extraction is shown in fig 3.3. The fuel mixtures proposed for IFR are U10 % Zr and U19 % Pu 10 % Zr and are supposed to be fabricated by direct injection casting of the recycled semi purified TRU fraction.

A similar pyrometallurgical partitioning process but starting from aqueous HLW has been proposed by INOUE et al at CRIEPI [3.15]. In this process the HLW is first denigrated and calcined. The calcined oxides are chlorinated and the reductive extraction of TRU is carried out in a Li-$CdCl_2$ bath. The

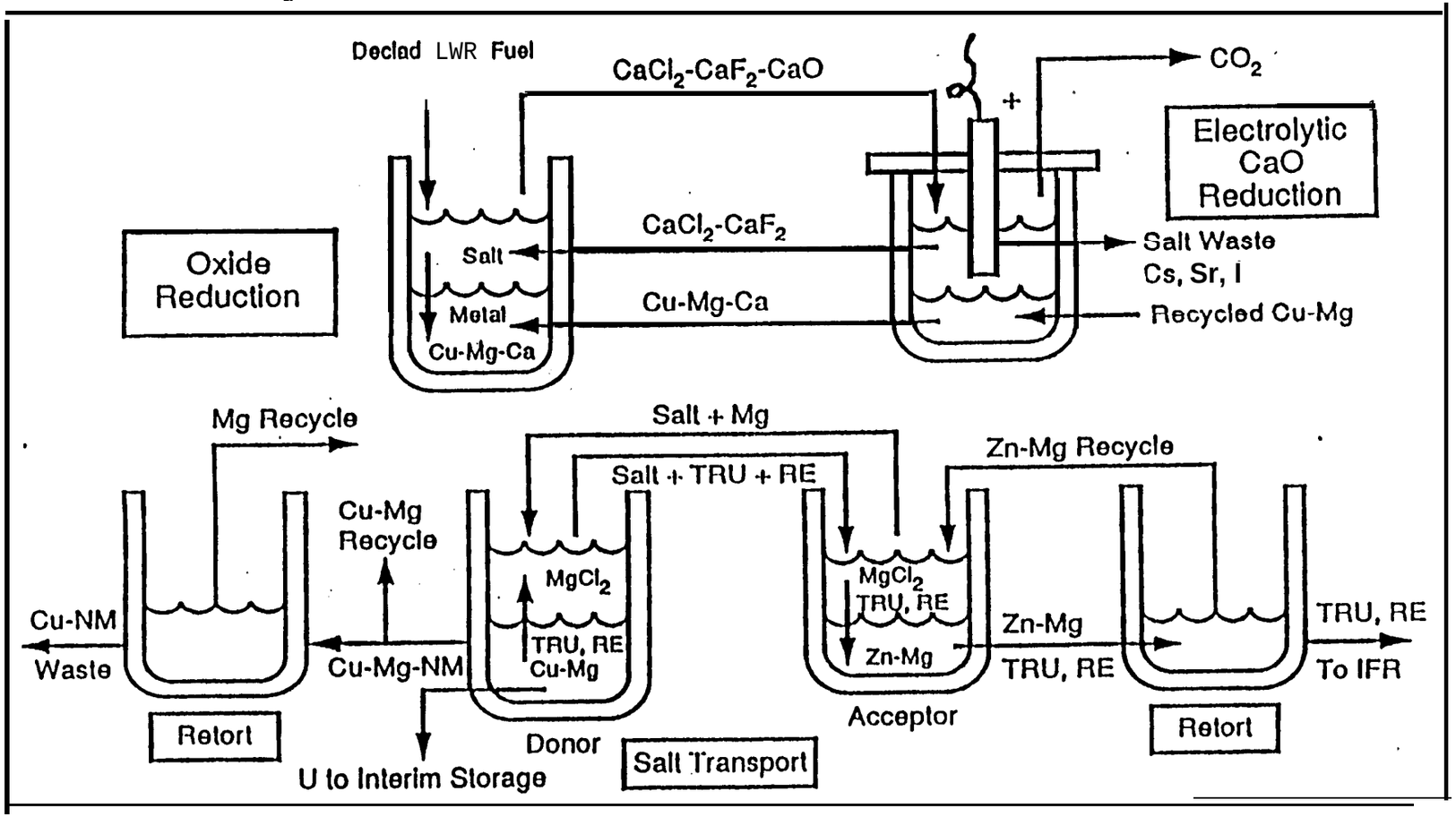
TRU Recovery and Waste Treatment



Chemical Technology Division

Fig. w.w.a

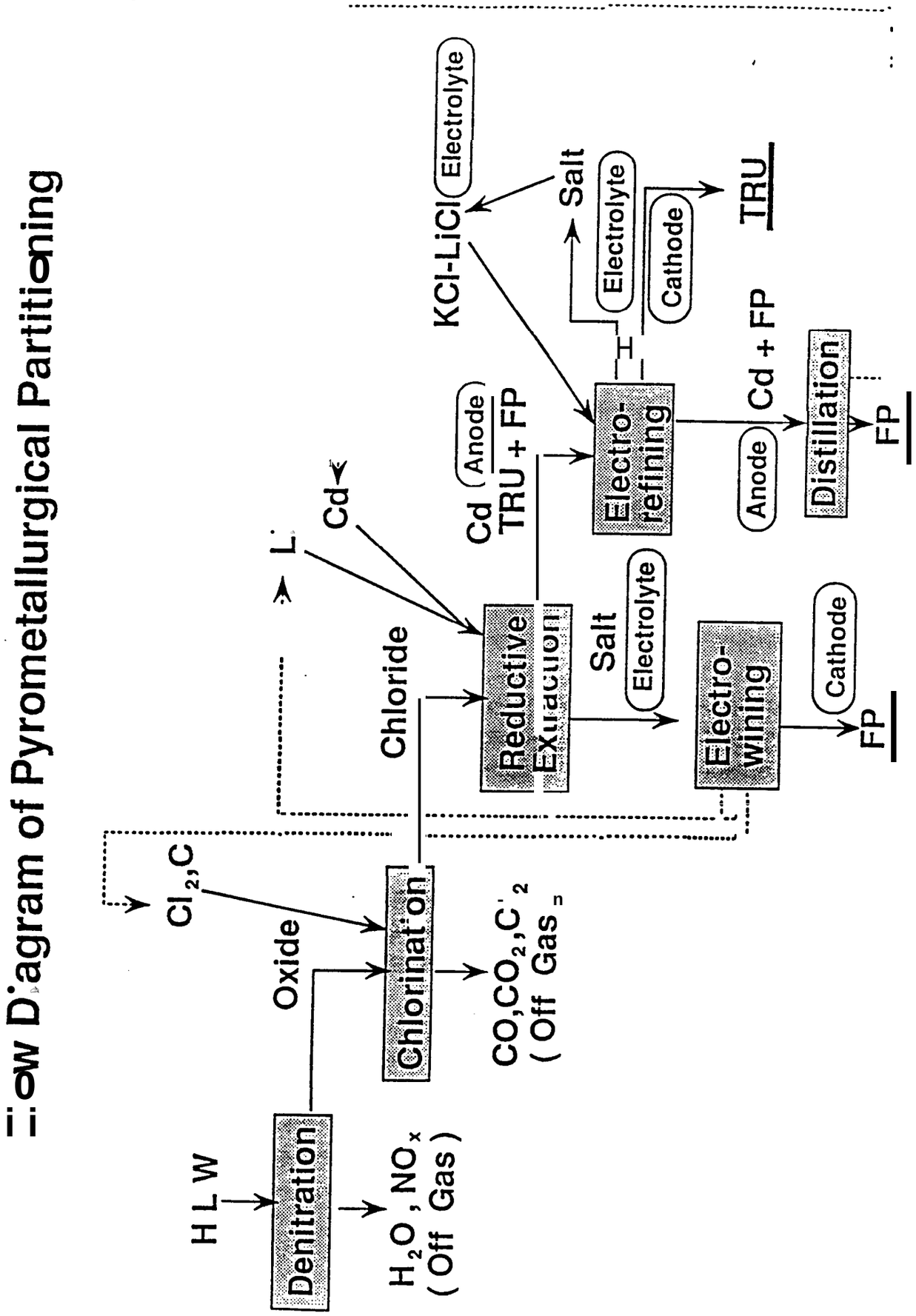
Salt Transport Conceptual Process



Argonne National Laboratory

Fig. 3.3.b

Fig. 3.4.


Flow Diagram of Pyrometallurgical Partitioning
HLW
Denitration
H_2O, NO_x (Off Gas)
Oxide
Cl_2, C
Chlorination
CO, CO_2, C'_2 (Off Gas)
Chloride
Li
Cd
Reductive Extraction
Cd (Anode) TRU + FP
Salt
Electrolyte
Electro-winning
Cathode
FP
KCl-LiCl
Electrolyte
Salt
Electrolyte
Cathode
TRU
Electro-refining
Anode
Cd + FP
Distillation
FP

electrorefining step is identical to the IFR process; fig 3.4 shows the flow diagram of the process.

It is obvious that these processes are still in the exploratory stage and cannot yet be considered as proven, even at the conceptual level. The pyrochemical processes have the advantage not to involve any water mixture which is favorable for criticality safety but are very sensitive to oxygen and moisture ingress into the equipment. The combined pyrochemical IFR process is very complex as it includes at least 3 different molten-salt-molten alloy contractors which produce a waste stream made up of a very toxic Cd salt. The question arises what to do with the waste streams and how to reduce the environmental impact of fission products dissolved in a toxic salt mixture. Therefore it is premature to consider these processes as alternative fuel fabrication and TRU recycling methods.

3.2. MINOR ACTINIDES TRANSMUTATION

The generic term transmutation covers two distinct processes : the transformation of a nuclide into a heavier species by neutron capture and the "incineration" due to neutron induced fission.

The transformation of a long lived nuclide eg. Np 237, into a shorter lived decay product from neutron capture eg Pu 238 is a typical example of neutron induced shortening of the actinides 'lifespan. In thermal neutron systems transmutation by capture is by far the dominant process especially for Np 237 Am 241 and Am 243. Table 111.4 summarizes the thermal cross sections of the MA and some long lived fission products. [3.16] Incineration also occurs in thermal systems but generally in an indirect way by formation of a decay product with higher fissile cross section Pu 238, Pu 239 Am 242m, Cm 243 and Cm 245 are typical examples.

Introduction of nut"lides with predominantly neutron capture into a reactor core will increase the antireactivity and require a higher enrichment. During the life of the reactor core the reactivity may increase with the formation of fissile nuclides. The minor actinides play a role as burnable poisons at the Beginning of Cycle (BOC) and contribute to the reactivity in the course of the core life upto the end of Cycle (EOC) conditions.

TABLE 111.4. : THERMAL CROSS SECTIONS OF MA in barn

NUCLIDE	Np 237	Am 241	Am 242;	Am 243	Am 244 m g	Pu 238
σ°_{Y}	175.9	587 ± 12	(m) 2000 ± 600 (y) 2100 ± 200	75.1	(m) 1600 ± 300 (g) 2300 * 300	549
σ°_{f}	21.5 mb	3.2 b	(m) 6950 ± 280	74 ± 4 mb *		17
I_{Y}	640. ± 50	1425 ± 100		1820 ± 70	-	156
I_{f}	6.9 ± 1	14.4* 1	(m) 1800 ± 65	9 * 1	-	32

NUCLIDE	Cm 242	Cm 243	Cm 244	Cm 245	Cm 246	Cm 247	Cm 248
σ°_{Y}	16 ± 5	130 * 10	15.2 ± 1.2	369. ± 17	1.22 ± 0.16	57 * 10	2.63 ± 0.26
σ°_{f}	< 5	617 ± 20	1. *0.2	2145 ± 58	0.14 * 0.05	81.9 ± 4	0.37 * 0,05
I_{Y}	110. ± 20	215 * 20	650. ± 30	101. ± 8	121. * 7	530. •30	270. * 15
I_{f}	-	1570 * 100	12.5 ± 2.5	840 ± 40	10.2 * 0.4	'760 ± 50	15

THERMAL CROSS SECTIONS OF FISSION PRODUCTS (barns)

NUCLIDE	Se 79	Zr 93	Cs 135	I 129	Tc 99
U°_{y}		1.3 - 4	8.7 ± 0.5	188 ± 2 9 ± 1	22
σ°_{f}	-				
I_{Y}	.		62 ± 2	366 ± 4	300
*f	.				

Ref. NEUTRON CROSS SECTIONS, Vol. 1, part A & B: S.F. MUGHABGHAB; Acad. Press 1981.
* c. Wagemans et al., Nucl.Sc. & Eng. 101 (1989) 293 _ σ°_{f} = 74 ±4 mb

Direct incineration of MA can only be performed in a fast reactor spectrum. Table 111.5. gives the capture and fission cross section of the U-Pu-MA nuclides. [3.17]

At first glance it is clear that also in the case of a FBR the capture cross sections are important. For Np 237, Am 241, Am 243 and Cm 244 the capture. cross sections are prevailing. On the other side the fission cross sections are dominant for Pu 238, Am 242m, Cm 243 and Cm 245.

A conventional FBR will not only fission the MA but also shift their average mass to a higher number. However, the great nuclear advantage of the FBR is its insensitivity to thermal poisons (rare earths) and its potential to extract substantial fission energy from MA. Though absolute values of the fast neutron capture and fission cross sections are much lower than the thermal cross sections this effect is compensated by the fast flux which is 100 to 1000 times higher.

Two types of reactors are generally considered for actinide transmutation : LWR.PuO_2 and LMFBR's. These reactors are capable of using significant amounts of MA through a homogeneous recycling process.

At the end of the large R & D programme carried out by JRC in the 1975-1982 period, a comprehensive final report entitled "Assessment studies on Nuclear Transmutation of Byproduct Actinides" has been published in 1984 [3.18]. It constitutes a synthesis of the state of the art at that period and will be used as a starting point for this study.

TABLE 111.5. : CAPTURE AND FISSION CROSS SECTION FROM ORIGEN-2 CODE LIBRARY FOR FAST REACTOR FUEL CALCULATIONS (1990) AND DATA USED BY JRC (1980)

Nuclide	σ_c		σ_f	
	ORIGEN-2	JRC (1980)	ORIGEN-2	JRC (1980)
U 235	0.5303	0.642	1.865	2.070
U 236	0.5252	0.619	1.237	0.088
U 238	0.2794	0.327	0.052	0.039
Np 237	1.429	1.88	0.391	0.328
Pu 238	0.6932	0.505	1.188	1.160
Pu 239	0.4685	0.600	1.822	1.870
Pu 240	0.4847	0.595	0.424	0.350
Pu 241	0.4386	0.536	2.44	2.670
Pu 242	0.4046	0.413	0.307	0.264
Am 241	1.32	2.04	0.345	0.283
Am 242m	0.3646	0.510	3.896	3.18
Am 243	0.0502	1.39	0.271	0.295
Cm 242	0.3078	0.435	0.1985	0.140
Cm 243	0.2281	0.385	2.601	2.66
Cm 244	0.794	1.04	0.487	0.39
Cm 245	0.2981	0.371	2.594	2.84
Cm 246	0.2211	0.458	0.320	0.294

3.2.1. Homogeneous LWR recycling of MA in fission reactors

3.2.1.1. *Transmutation in LWR's*

A standard LWR (33 GWd/T), commissioned for Pu recycling can use up to 9 T of 5 % Pu enriched MOX fuel per year as only one third of a PWR core may be substituted by MOX fuel elements. In a maximum scenario a homogeneous Np enrichment of 2 % would be tolerated and 180 kg Np per year could be "consumed" by a 1000 MWe year PWR reactor. It will be gradually transformed into Pu 238 since the capture cross section of Np 237 is dominant (170 barns) compared to the very smal 1 (0.02 barn) fission cross section.

Neptunium behaves as a burnable poison and its presence must be compensated by a higher initial Pu enrichment. However due to the relatively low flux only a partial depletion can be achieved. For example an initial mass of Np 237 is according to Pilate [3.1] transmuted into Pu 238 and some Pu 239 ; but this transformation is limited to 54 % in a PWR-MOX core and 63 % in a PWR-UO_2 case after 4 years of irradiation and a burnup of 45 GWd/T. The residual 46 % of Np (83 kg) would still be present in PWR-MOX spent fuel and such an approach does not yield any significant radiological benefit, unless the recovered Pu is transferred to a FBR fuel cycle.

Recycling of Np in LWR-UO_2 is not a viable route since it would be difficult to co-process natural or slightly enriched U together with a very toxic alpha emitter such as Np 237 in the same plant which was conceived for U processing only. This incompatibility is yet limited to those process steps where open isotopic sources are handled. Once a fuel pin has been welded its radioactive content does not interfere with the other activities in the U fuel fabrication plant.

Similar computations have been made for Am 241 which can also be transmuted in a PWR-MOX reactor and in this case a transformation yield of 87,4 % is predicted. However other difficulties related to the highly active nature of this radionuclide interfere with such a venture : a.o. the separation of Am 241 from HLLW, the presence of non negligeable amounts of Rare Earths and the handling of highly active radionuclides in a conventional or even improved fuel fabrication plant.

It can be concluded that, even disregarding the inherent difficulties associated with the separation and conditioning processes, homogeneous recycling of Np and Am in PWR-MOX thermal reactors does not lead to a significant reduction of their concentration in spent MOX fuel. Moreover recycling of spent PWR-MOX fuel with residual Np + Am amounts, through a second reprocessing cycle produces a degraded Pu quality which can only efficiently be reused in a FBR option.

3.2.1.2. Homogeneous MA recycling in FBR.

The reference case considered in homogeneous recycling of MA in FBR's involves the following steps.

- Separation of MA (Np, Am, Cm) in LWR fuel reprocessing plant according to an improved method for quantitative Np recovery in the PUREX process and a new process for Am recovery from HLLW.

- Purification of MA (particularly removal of bulk Rare Earths) and transformation into MA-oxides with 1 to maximum 10 % impurities.

- Mixing of Np and Am-Cm with UO_2 PuO_2 powders in order to get a reference composition.
  UO_2 15-18% PuO_2 2 % Np 2 % AmCm
  The Am content listed is supplementary to the ingrowth of Am 241 from Pu 241.

- Fuel fabrication of FBR fuel.
  * if only Np is mixed with UO_2PuO_2 conventional fuel fabrication methods can be employed (sintering, pelletization)
  * if Am-Cm is also mixed, advanced fuel fabrication methods have to be used eg sol-gel processes and vibro-compaction-densification methods.

- Loading of MA containing core and axial blanket fuel assemblies into the reactor.

Table III.6

• **Mass of BPAs [g/THM] as a function of cycle number at BOC applying a homogeneous self -generated BPA recycle scheme in an LMFBR**

Isotope	Cycle number						
	1	2	3	6 "	10	15	20
Np-237	**3.96·10^{+2}**	**6.27·10^{+2}**	**7.61·10^{+2}**	*8.86.10'^{2}*	*9.46.10^{42}*	*9.50.10'^{2}*	*9.50.10"^{2}*
Am.241	5.63.10^{42}	8.89.10'^{2}	1.08.10'^{3}	**1.25·10^{+3}**	1.34.10'^{3}	1.35.10^{43}	1.35.10'^{3}
Am-242M	2.21.10"	5.67.10'^{1}	8.49.10'^{1}	1.17.10'^{2}	**1.35·10^{+2}**	1.37.10'^{2}	1.37.10'^{2}
Am-243	3.24.10'^{2}	5.42.10'^{2}	6.88.10^{42}	**8.54·10^{+2}**	9.74.10'2	**9.91·10^{+2}**	9.93.10'^{2}
Cm-242	**2.86·10^{+1}**	5.74.10"	7.44"10^{4}'	9.01.10"	9.76.10^{4}'	**9.81·10^{+1}**	**9.82·10^{+1}**
Cm-243	**1.66·10^{+0}**	**5.82·10^{+0}**	9.91.10'^{0}	**1.52·10^{+1}**	**1.87·10^{+1}**	**1.90·10^{+1}**	**1.90·10^{+1}**
Cm-244	**4.60·10^{+1}**	1.40.10'2	**2.44·10^{+2}**	4.22.10'^{2}	6.33.10'^{2}	6.80.10'^{2}	6.89.10'^{2}
Cm-245	3.58.10^{40}	**1.85·10^{+1}**	4.24"10'^{1}	9.69.10'^{1}	1.82.10^{42}	**2.04·10^{+2}**	2.09.10'^{2}
Cm-246	9.54" 10.2	9.66.10"^{1}	3.34.10 ^{10}	1,31.10^{4}'	4.59.10"	7.50.104 ^{1}	8.92.10"
Cm-247	215.10"3	**3.81·10^{-2}**	1.81.10^{1}	**1.02·10^{+0}**	5.68.10^{40}	**9.78·10^{+0}**	**1.23·10^{+1}**
Cm-248	6.18.10^{5}	**1.74·10^{-3}**	1.23e10.^{2}	1,16.1 (11	**1.31·10^{+0}**	**3.27·10^{+0}**	5.05.10'^{0}
Bk-249	8.09.10-7'	**4.57·** 10"^{5}	4$_{0}$17.10"4	'5.18e10"^{3}	7.57.10-^{2}	2.07.10"^{1}	3.32.10^{-1}
Total	**1.38·10^{+3}**	2.34e10'^{3}	2.98.10'^{3}	3.7510^{43}	4.38.10^{43}	**4.51·10^{+3}**	4.55.10^{43}

Irradiation during 3 cycles of 1 to 1.5 years to reach a burnup of 100.000 MWd T-l and a mean MA transmutation yield of at least 11 % per cycle.

- Cooling of spent fuel during 3 years.

- Reprocessing in specially designed facilities.

Purification of MA and transformation into oxides.

- Refabrication of second cycle UO_2 15-18 % PuO_2 2 % Np 2 % AmCm.

- Reloading in the reactor for 2^{d} recycle etc...

Early calculations by OLIVA [3.17] and SCHMIDT [3.18] of homogeneous self generated MA recycle in FBR's showed a gradual increase in MA concentration in the core fuel and a slightly decreasing Transmutation yield in the course of the repeated recycling (from 29.5 % at the first recycle to 28.7 % at the 20th cycle. These calculations were based on an irradiation time of 546 EFPD and 81.000 MWd/T. The reactor physics conclusion was a gradually increasing reactivity which reaches a steady level from the 10th recycle. It means that homogeneous self generated recycle of MA does not require additional enrichment with fissile material [3.17]. Table 111.6. taken from the JRC report [3.18] shows the concentration levels for all minor actinides.

The overall efficiency of transmutation η (capture and fission) was given by the relation.

$$\eta = \frac{(1-An)}{nW}$$

with An : Actinide concentration at the nth recycle
W : mass of MA produced per cycle.

By using the data of table 111.6. it results that the overall efficiency is 0.782 after 15 recycles and 0.835 after 20 recycles. Such a sequence would take 35 to 46 years to be accomplished. The global decontamination factor of 5 which results from this exercice shows the inherent difficulties of transmutation if the yield is limited by burnup or cladding performance.

Ultimately 20 % of the recycled MA remain in the spent fuel of the last recycle and has any way to be stored in a geologic repository to be kept from the biosphere for thousands of years.

Takano et al [3.19] investigated the overall feasibility of homogeneous recycling of MA discharged from LWR's into a generic FBR and its consequences on the reactor physics characteristics.

Recycling of MA generated by LWR's was investigated from reactor physics point of view under the hypothesis of a quantitative and homogeneous recycle of MA into FBR-MOX fuel.

The reactor computation based on the SRACLIB - JENDLE 3 cross section library was carried out with an adapted fissile Pu content of 11 % up to a burnup of 90.000 $MWdT^{-1}$. The most important conclusion from this study was that recycling of MA up to 8 times did not affect the fissile material content at BOC and that the burnupreactivity swing was only 14 %. The behaviour of MA as burnable poison suggested in earlier studies was, this way, confirmed especially for Np 237 and Am 241.

The coolant void reactivity which is very important for reactor safety showed to be a slightly positive coefficient (+2.7 %) In the case of MA recycling. This fact has to be taken into consideration when designing a FBR core with MA loading. So called flat cores (pan cake configurations) with a certain neutron leakage can cope with this problem. An advanced fast breeder concept with metallic fuel (MA-Pu-Zr) does not display this positive coolant void reactivity.

Recently quantitative calculations were carried out by Yamaoka et al. [3.20] on the transmutation characteristics of MA In a 1000 MWe FBR-MOX.

Table III.7.

Summary of TRU inventory and transmutation rate [3.20]

Core	Isotope	Amount of TRU loaded (kg/cycle)	TRU inventory		Amount of TRU discharged (kg/cycle)	Amount of TRU transmuted (kg/cycle)	TRU transmutation rate* (%)
			BOEC(kg)	EOEC(kg)			
Reference core	Np	0	6	11	5	-5	
(no TRU loaded)	Am	0	46	82	36	-36	
	Cm	0	4	8	6	-5	
	Total	0	66	101	46	46	
Homogeneous	Np	289	719	590	160	129	18.0
TRU-loaded core	Am	268	710	624	182	86	12.2
	Cm	31	142	173	63	-31	-22.1
	Total	589	1671	1387	404	184	11.7
Heterogeneous	Np	302	769	647	180	122	15.8
TRU-loaded core	Am	280	752	669	197	83	11.0
	Cm	33	140	168	61	-28	-20,4
	Total	614	1660	1485	438	176	10.6

*(Amount of TRU transmuted) / (TRU inventory at BOEC)

The isotopic composition in % of MA in LWR fuel was after 5 year cooling calculated with the ORIGEN 2 code and gave the following data : Np 49.1 %, Am 241 - 30 %, Am 243 15.5 % Cm244 5 %. The main data as reported by the authors are shown in table 111.7. and consist of three sets of data : a reference FBR MOX without MA loading, a homogeneous MA recycle and a heterogeneous actinide recycle.

The total MA load was set at 5 w/o amounting to 719 kg Np, 710 kg Am and 142 kg Cm in the core and axial blanket assemblies.

One of the most striking results with important consequences for further MA recycle scenario's is the positive breeding of Cm. When comparing the Cm (243 + 244 + 245) quantity at BOC an increase of 22% is observed at EOC.

The situation for Np 237 and Am (241 + 243) is much more attractive since 18 % respectively 12.2 % disappears by transmutation during a 456 days cycle of EFPD. If these data are transformed into calendar years with a 0.75 load factor, the yearly transmutation yield amounts to 10.8 % Np and 7.3 % Am.

A 1000 MWe $PWR-UO_2$ produces per year about 22 kg MA : 11.8 kg Np and 10.2 kg Am.

A homogeneously loaded FBR core with 2.5 % Np and 2.5 % Am would contain 719 kg Np and 710 kgAm. This corresponds to the total MA output of 60 GWe year $PWR-UO_2$ or roughly to the accumulated yearly capacity of a majour reprocessing plant (e.g. La Hague or Sellafield)

However the yearly transmutation yield which in this case means : incineration to fission products, corresponds to 77.6 kg Np and 51.8 kg Am. Compared to the MA output from the $PWR-UO_2$ reactor operation a yearly MA incineration in a 1000 MWe - FBR-MOX corresponds to the production of 6 units 1000 $MWe-PWR-UO_2$. This MA 'incineration' approach requires a very important FBR-MOX capacity to be installed throughout the world in order to transmute the output from the majour operating and projected reprocessing plants.

If we recall the data of Chapter I-Table I with the nuclear electric capacity, and Table III-1 with the MA production from reprocessing plants the following conclusions can be drawn :

A FBR-MOX reactorpark of 25 GWe is required to incinerate the total expected Np and Am output from the reprocessing plants constructed or planned in France, UK and Japan.

- The transmutation of Cm cannot be achieved in a FBR-MOX reactor. Other transmutation techniques have to be considered.

- The mean incineration period for MA varies from 9 years for Np 237 to 13.7 years for Am (241-243). This time interval is required to incinerate the initial loading of a FBR-MOX reactor loaded with 5 % (Np+Am).

- The financial implications of such an approach are very important and have to take into consideration : the supplementary electricity cost of a FBR-MOX compared to an equivalent LWR-UO_2 ractorpark, the additional reprocessing and fuel fabrication costs, the possible decrease in geologic disposal volume, the adequate use of Pu in a closed fuel cycle.

Any increase in transmutation yield will directly improve the economic appeal of such a scenario.

The incineration by a FBR-MOX reactor of MA is in the above sketched scenario limited to those countries having taken a positive option versus reprocessing as a whole and advanced reprocessing in particular. As has been explained in chapter I 50 % of the world nuclear energy production capacity have selected the non reprocessing option and due to delays in the reprocessing capacity construction compared to the nuclear electricity production every year increasing amounts of spent fuel are stored till a solution for their disposal is found.

3.2.2. Heterogeneous recycle transmutation

3.2.2.1. Transmutation in thermal reactors

In principle heterogeneous recycle could transmute roughly the same amount of MA as homogeneous recycle from pure reactor physics point of view. However the thermal characteristics of fuel assemblies Toaded with high concentrations of MA require the use of specially designed fuel pins and assemblies. Neutron shadowing is a very important factor in yield calculations as a function of time. In order to reduce additional MA formation from an U matrix a replacement by a nuclearly inert material e.g. ZrO_2 or MgO has been proposed. A systematic assessment study has been carried out by BUCCAFURNI and LANDEYRO [3.21] on generic cases involving the choice of the optimum matrix and the influence of neutron fluence on the final radiotoxicity of the actinide mixture with ORIGEN (2) as calculation code.

By substituting UO_2 with ZrO_2 an overall gain in transmutation efficiency with a factor 3 has been computed for an equivalent storage time and this conclusion is valid up to 5000 years.

At discharge after 1100 days of irradiation the radiological toxicity is nearly the same as at beginning of cycle. A significant radiotoxicity reduction factor of about 10 is obtained only after 11000 days under conventional nuclear power plant fluxes. After 22000 days (73 years) the overall toxicity is decreased by about a factor 400. Table 111.8. shows some typical results taken from this study.

The most important conclusion yet is the necessity to go to very high neutron fluences in order to beat the natural decay as a function of storage time. These very high neutron fluxes are not available in NPP's of the present generation.

High flux reactors with neutron fluxes of 2-7 10^{14} $n/cm^2.s$ are very interesting because the capture cross section is quite high and results either in a shorter lived actinide (e.g. Np 237 → Pu 238) or in a more fissionable activation product (e.g. Am 241 → Am 242).

Table 111.8.

Radiotoxicity ratio's of Actinides resulting from irradiation of MA in a ZrO_2 matrix at a concentration level 10 times the annual discharge of a LWR perTHM spent fuel (from 3.21).

Irradiation time EFPD	1100	3300	11000	22000
Total residual quantity (g MA)	"5.77 10^4	2.92 10^{4-}	3.06 10^4	898
Radiotoxicity index at fluxes n/cm²/s 2.8 10'* 2.5 10'⁴ 1 10'ˢ	1.337	0.767	0.089 9.4 10^{-5} 1.37 10^{-6}	0.0037 - -
Radiotoxicity ratio after 5000 years storage	0.00454	0.00301	0.00066	0.00022

3.2'.2.2. *Transmutation in fast reactors*

Heterogeneous recycle in FBR-MOX reactors has been investigated from the early beginning at JRC [3.18]. The main conclusions were that :

from reactor physics point of view very little influence was observed from the MA composition whether it resulted from the-first or the 20th recycle (see table 3.4)

at equilibrium composition the burnup swing reactivity loss between BOC and EOC is almost negligeable.

power peakings in target assemblies were predicted and computations were based on dilution of MA in MgO.
(0.36 gMA/cmmixedwith 1.7 g MgO).

two specific loadings (one central assembly and six surrounding assemblies) containing MA mixtures showed an inventory reduction of 37.4 % and 34.5 % per cycle which is substantially higher than in homogeneous recycle at the same neutron fluence.

A detailed analysis of heterogeneous recycling of MA mixed with UO_2 in a 1000 MWe FBR- MOX was carried out by YAMAOKA et al [3.20] and led to the following results :

- the transmutation yield is very much dependant on the location of the target assemblies in the core, dispersed location is preferable.

- the overall MA inventory is nearly the same as for homogeneous recycle i.e. a maximum of 5 % (see table 3.5)

- the transmutation yield i.e. kg of MA transmuted per cycle of 456 d is slightly lower than in homogeneous recycle.

- the swing of power peaking near the UO_2 - MA target elements becomes quite severe and critical for the thermal design criteria.

An experimental programme (SUPERFACT) was carried out in a joint venture between TUI and CEA-CADARACHE [3.11] and resulted in the fabrication and irradiation of Np/Am oxide fuel pins mixed with UO_2 in the PHENIX reactor. It could be concluded that the behaviour during irradiation was similar to that of $UPuO_2$ fuels. However up to now only a cumulative burnup of 8.3 % has been attained and this is relatively small in the context of MA transmutation.

If heterogeneous recycle would be adapted it seems unavoidable to substitute the UO_2 matrix by neutronic-neutral matrices eg. Al_2O_3, ZrO_2 or MgO.

The majour principal advantage of heterogeneous recycle lies with the fuel fabrication and reprocessing steps where tailor made facilites with relatively low throughput would be sufficient to accompany a reactor irradiation programme.

From reactor point of view there is no significant difference in transmutation yield compared to homogeneous recycle. Thermal problems may occur in Am loaded fuel assemblies. But the overall conclusions of § 3.2.1.2. (p. 99) remain valid and apply as wel 1 to the heterogeneous recycle route.

3.2.2.2. *Transmutation in IFR and ABR's.*

The Integral Fast Reactor (IFR) and the Minor Actinide Burner Reactor (M. ABR / P.ABR) are new answers to the transmutation issue by which the MA follow a complete separate route once they are discharged from the LWR - UO_2 reprocessing plants.

Since 1980 very important-progress has been made in the development of a fast reactor concept with very high mean neutron energy ($\geq$ 700 Kev). It was shown in earlier reports that the fission to capture ratio of MA increases with the mean neutron energy ; an increase with a factor of 3 is observed between 700 and 1070 keV. [3.22]. Under these circumstances the minor actinides are additional fissionable resources instead of waste materials as in the LWR fuel cycles.

Two concepts are under development : the Integral Fast Reactor (IFR) in the US and the minor Actinide Burner (ABR) with two options a metal cooled one (M.ABR) and a gascooled one (P. ABR) both in Japan.

The key features of the IFR concept include a pool type sodium cooled reactor with UPuZr alloy fuel, high internal-conversion ratio - core designs and very specifically a pyrometallurgical fuel reprocessing process integrated into the IFR plant site [3.23] [3.24].

The emphasis for the development of fast reactors has shifted in the US from the Pu breeding purpose to a Pu and actinide burning objective. Compared to the MOX fuelled fast reactor, which is burnup limited (200-250 GWd/THM), the IFR reactor fuel concept is fluence limited (< 3.10^{23} nvt). The metal fuel core is radially heterogeneous with subsequent layers of driver fuel, internal blankets and radial blankets which have to be re-processed simultaneously. The core residence time is about 3 years with annual partial refueling and associated pyro metallurgical reprocessing in view of removing fission products only. The reinsertion of pyrometalurgically reprocessed fuel is based on injection casting of metal fuel with Na bonding.

EBR-II Reactor and Fuel Cycle Facility

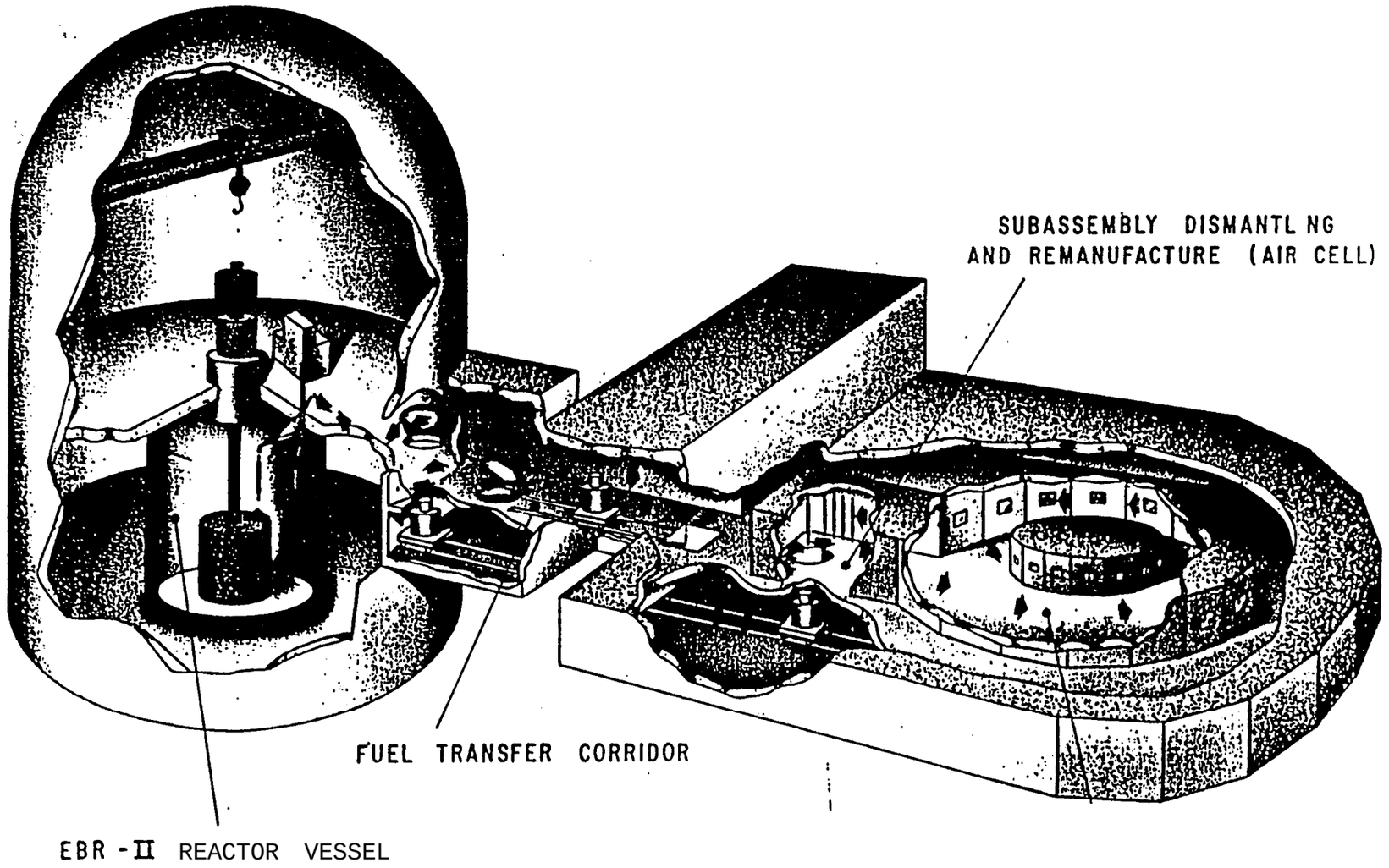


Fig. 3.5.

The IFR fuel cycle concept is represented schematically in fig 3.5. , it includes a Liquid Metal Fast Reactor with a breeding ratio of 1.17, a pyroprocessing unit based on direct electro refining of decanned spent fuel and a unit for remote fuel refabrication. The fuel behavior has been demonstrated in EBRII.

The IFR is conceived as a Pu burner with a significantly lower MA concentration in comparison with LWR's and is conceptually-capable of "burning" the Pu and Actinides discharged from LWR's. The fundamental drawback of the IFR fuel cycle is the coprocessing of MA and Rare Earths which are only partially separated from each other during the electrorefining process. This would in the end lead to a overenrichment of Rare Earths in the fuel mixture.

In order to transform the IFR concept into a Minor Actinide Burner Reactor concept any introduction of fertile material (U 238) would be avoided and the breeding ratio would be kept below one. Therefore the fuel should consist mainly of MA and Plutonium. The Minor Actinide Burner fast reactor concept based on MA-Pu fuel has been intensively studied in Japan [3.25] [3.26].

The first option is a sodium cooled MA metal fuel burner (abbreviated M-ABR) . In this reactor two types of fuel are used :

Np 22 % Pu 20 % Zr in the inner core
AmCm 35 % Pu 5 % Y in the outer core.

The reason for separate fuel elements is the limited solid volubility of Np and Am-Cm. Zr and Y are added to produce an alloy with a higher melting point. Pu is only added to the initial fuel inventory as a source of initially fissionable material which gradually depletes and is compensated by a reactivity gain of MA due to Transmutation of Np 237 into Pu 238-239 and Am 241 into Am 242 m.

The fuel irradiation parameters are similar to those of the IFR concept i.e. fluence limited reactor cycles (fluence 2.3 10^{23} nvt, flux 3.6 10^{15} n/cm².s, cycle length 730 EFPD) determine the cycle length.

Table 111.9.

Comparison of MA transmutation in various reactors [3.26]

		MA[1] Burner Reactors		PowerReactors		
		M-ABR	P-ABR	U-PWR	MOX-FBR	LMR
output	(MWt)	170	1200	3410	2600	2632
Cycle length[2]	(FPD)	730[2]	300[3]	850[4]	750[4]	900[4]
Core averaged Fast neutron flux	(X1O[15])	3.6	8.4	Q. 37	3.3	5.0
Mean neutron energy	(keV)	780	750	thermal	480	490
MA loaded	(kg)	666(*)	2065	180"	1450[2]	1200[5]
MA transmutation rate[6]	(%/cycle)	26.0	25.3	54.1	27.6	38.7
	(%/year)	10.7	25.3	19.1	11.0	12.9
MA burnup rate"	(%/cycle)	17.8	17.2	15.0	9.4	16.8
	(%/year)	7.3	17.2	5.3	3.8	5.6
MA burnup/reactor	(kg/year)	49	355	9.5	55	67
MA generated"	(kg/year)	—	---	26	35	30
Net MA burnup	(kg/year)	49	355	-16.5	20	37
MA burnup	(kg/1GWt·year)	287	296	-4.8	7.7	14

1) MA: mixture of minor actinides such as Np, Am, Cm (initial load 255 kg Pu, 199 kg Am.Cm 212 kg Np) (*)
2) Fuel irradiation time
3) Fluence limited
4) Burnup limited
5) Concentration of MA in fuel: 0.2% for U-PWR、5% for MOX-FBR and LMR
6) MA transmutation rate = $\frac{\text{MA(BOC)} - \text{MA(EOC)}}{\text{MA(BOC)}}$
7) MA burnup rate = $\frac{\text{MA fissioned}}{\text{MA(BOC)}}$
8) MA generated from fuel, ie U and Pu

In the present M-ABR designs a high neutron flux could not be attained due to the low thermal conductivity of the TRU alloy and its relatively low melting point. A modified version M3-ABR with smaller fuel pin diameters has been proposed but this thermal improvement has been obtained at the expense of the mean neutron energy which on his turn determines the direct fissioning rate of the MA. The He cooled particulate bed MA-Burner Reactor is a more recent version of ABR. The fuel is made of microsphere of MA-Nitrides coated with TiN which are kept-in concentric--fuel pins made of porous metal tubes through which the MA can permeate to cool the coated fuel particles.

The ^{14}C generation which will undoubtedly occur in this type of fuel may complicate the pyrochemical processing step and its occurrence as long lived activation product will negatively influence the overall radiological benefit resulting from the actinide transmutation. A comparative analysis was made of the Actinide transmutation and incineration potential of different types of nuclear reactors in comparison with ABR's. Table 111.9. show the data as reported by MUKAYAMA [3.26] and TANAKO [3.19].

A direct comparison cannot be made because the power output and cycle time differ from case to case. However the data expressed per GWth - 300 days are fully comparable and show that the ABR's have a far superior behavior in terms of transmutation and fissioning compared to PWR-UO_2 or MA-FBR's. The PWR-MOX produces more MA per cycle than can be transmuted. If in a futuristic plot the M.ABR concept would further be improved from fuel and reactorphysics point of view, these types of reactors would have an important MA incineration capacity :

MABR case 1000 MWth or 400 MWe at 80 % load factor 1000/170 x 454 kg Np-Am/170 MWth x 0.073 fissioning = 195 kg FP/year.

P.ABR case 1000 MWth or 400 MWe at 80 % load factor.
1000/1200 x 1363 kg/Np-Am 1200 MWth x 0.17 fissioning = 193 kg FP/year.

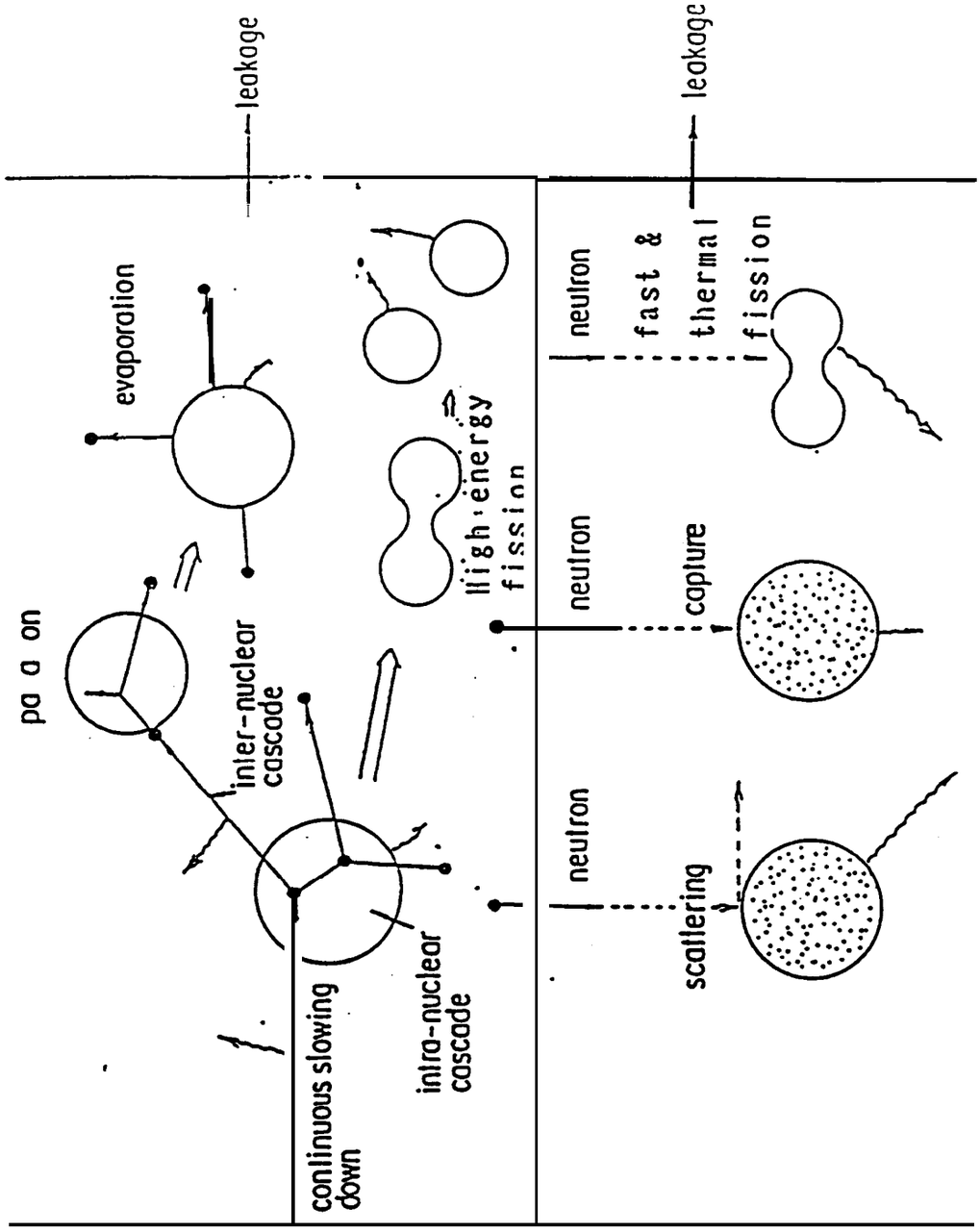
The electricity production of $2.88.10^9$ KWh. per reactor.year has to support financially the reactor operation, the fuel fabrication and the special reprocessing operations, further analyses should be carried out to verify whether this is sufficient to set up such MABR centers.

In comparison with the FBR-MOX option, discussed above, this type of reactor would strongly reduce the required capacity to incinerate the MA. However a straightforward comparison is not possible because even in a FBR-MOX with a mean neutron energy of 480 KeV, transmutation into heavier nuclides is still very important whereas in ABR's at 700 KeV fissioning is the main process. The loading capacity of a 400 MWe M-ABR corresponds to 2670 kg Np - Am (Cm) and this is the output of 100 to 120 Units of 1000 MWe PWR-UO_2. The total electric capacity of the OECD countries produced by LWR-UO_2 is 134 GWe and the total reprocessing capacity (installed and planned) discharges a total of 3190 Kg Np-Am-Cm. One reactor of 500 MWe of the ABR type is sufficient to take up the future potential MA production per year from the combined reprocessing plants.

In order to estimate the required nuclear capacity of MA-burners we have to take into account the burnup rate per year. i.e. 7.3 % in a M-ABR and 17 % in a P. ABR. Per 1000 MWth this incineration yield is equivalent with 193-195 kg FP or the same amount of MA destroyed. Per unit of 1000 MWe LWR-UO_2 about 22 kg Np-Am (Cm) are formed and as a consequence the output of 8.7 units can be "incinerated" with one 1000 MWth (or 400 MWe) ABR. In order to destroy the entire Np-Am (Cm) production of the OECD countries about 16 GWth ABR capacity would be required. Expressed in reactor units of 400 MWe this capacity would be made up of 16 M-ABR units preferably situated near the three sites where conventional reprocessing plants have been erected or are planned (La Hague, Sellafield, Rokkasko - Tokai).
The bottleneck of this actinide incineration concept lies with the special reprocessing services and associated fuel fabrication plants which have to be built in order to keep the ABR's going.

The advanced Np - Pu fuel contains 22 % Pu and the Am (Cm) - Pu fuel contains 35 % Pu. The total fuel inventory which has to be discharged annually and transferred to the pyrochemical reprocessing units amounts to 2 THM per year per 400 MWe. When an equilibrium is reached between the MA-production from LWR - reprocessing plants and the M-ABR throughput a pyrochemical processing capacity of 80 THM/year should be made available. The other services eg fuel element storage (3 year) Pu storage and Vitrification of discarded FP are supposed to be available on the conventional

Fig. 3.6.



Spallation process in the target injected by high energy protons

reprocessing sites. Per site a capacity of 30 THM pyrochemical processing ought to be built in order to cope with the uncertainties of this new process.

Once the national LWR - UO_2 programme is halted, it takes an additional 14 years for MABR's and 7 years for P. ABR's to destroy the last amounts of MA loaded into the reactor.
Each unit discharge of 2 THM/year has to be recycled 7 times before the total M-ABR inventory is transmuted into fission products. If we assume 1 to 2 % losses per recycle the total "waste" inventory amounts to 7 resp 14 % of a unit charge (140 to 280 kg MA). During multiple recycle processes the nuclides which are not fissionable and not discarded with the F P stream will build up in the MA Pu charge. Therefore it is advisable to investigate pyrochemical techniques by which Rare Earths can effectively be separated from Am (Cm) streams.

Aqueous reprocessing processes which have acquired the largest industrial experience cannot be applied to this type of fuel. The large amounts of Cd necessary to carry out the pyrochemical electrorefining process will be contaminated with MA and cannot be vitrified according to the present borosilicate process. New ways should be investigated to solve this waste problem which is both radioactive and toxic.

3.2.3. Transmutation in accelerator driven systems

Incineration of Actinides by proton induced spallation and fission is one of the recently developed approaches which received considerable attention in Japan and the U.S.
Above the cut-off energy of 15 MeV the intra- and inter-nuclear cascade processes from proton bombardment of heavy nuclei produces a series of fragments which can be subdivided in three categories :

- light nucleons (n, d, 'T, He 3&4)
- fission products
- spallation products (fragmented nuclei).

Figure 3.6. shows schematically the nuclear processes involved.

The number of spallated nuclides per incident proton is relatively small and depends mainly on the proton energy. at 1.5 GeV only 5 nuclides are fragmented per impinging proton.

Among the light nuclei the neutrons play the most important role but the production of ^{3}T with a half life of 12.2 y deserves some attention in safety evaluations.

The number of neutrons produced during the spallation process is relatively high and depends on the--nature-of -the target---element -and on the proton energy. Table 111.10. summarizes the main data.

Table 111.10.

Spallated nuclides and neutron emission per impinging proton

Proton energy GeV	Directly spallated nuclides	Emitted neutrons per Proton			
		Pb	W	Np	Am
1	3	17	22	33	24
1.5	5	20	28	40	30
2	7	23	34	48	35
3	-	26	39	59	40

A beam of 1 A protons, with a unit charge of $1.6\ 10^{-19}$ C has a flux of $6.25\ 10^{18}$ p/s. The number of source neutrons emitted by such beam will range from 1.10^{20} to 3.710^{20} n/s depending on the target material and the proton energy.

The combination of high energy proton induced spallation and fission from secondary neutrons is therefore considered as an advantageous option for actinide incineration.

In view of assessing the potential of this option we have to consider the present state of the art in proton accelerators technology and the developments necessary to achieve an industrial level of operation.

The presently existing proton accelerators have energies between 500 and 800 MeVwith associated beam currents of 6.7 μA to 1 mA.

The neutron fluxes resulting from such machines range from 41013 to $6.25\ 10^{15}$ n/s.

An upscaling factor of 2 should be reached in proton energy (1.5 GeV) and a factor of 10 to 100 should be pursued on the beam current (10 to 100 mA).

Upscaling to higher energies (l-2 GeV) is easily feasible since very much technological development has been made in the design of high energy physics experiments and synchroton radiation machines. Most of the advanced technology used in high energy physics is directly available or can be adapted to the proton accelerator field. However a serious R&D effort will be required to develop a 1.5 GeV - 10 mA proton accelerator taking into account the special circumstances of the collocation of a pure physical machine with a highly radioactive target.

Vacuum sealing of the accelerator tubing is one of the most crucial problems to be solved either by designing a replaceable window between the accelerator and the target or by incorporating the highly radioactive target material into a single vacuum chamber.
The material test accelerator (MTA) at Livermore (U.S.) designed to produce highly energetic deuteron beams up to 500 MeV and 300 mA did not achieve its technical goals and realized only a 30 MeV - 50 MA beam.

In order to realize a 1.5 GeV - 100 MA proton accelerator coupled to a fuel irradiation and handling facility extensive research "and development will be needed. This option is anyhow a very long term objective with a lot of uncertainties and bleak economic outlooks.

Three different routes are presently under investigation.

I." Accelerator driven fast neutron incineration of metallic MA fuel (JAERI project).

2. Accelerator induced incineration of MOX type MA fuel (PHOENIX project).

3. Accelerator produced extremely high thermal neutron flux for transmutation of Actinides and fission products (LOS ALAMOS concept).

Each of these three options will be examined and compared with the direct reactor incineration of transmutation.

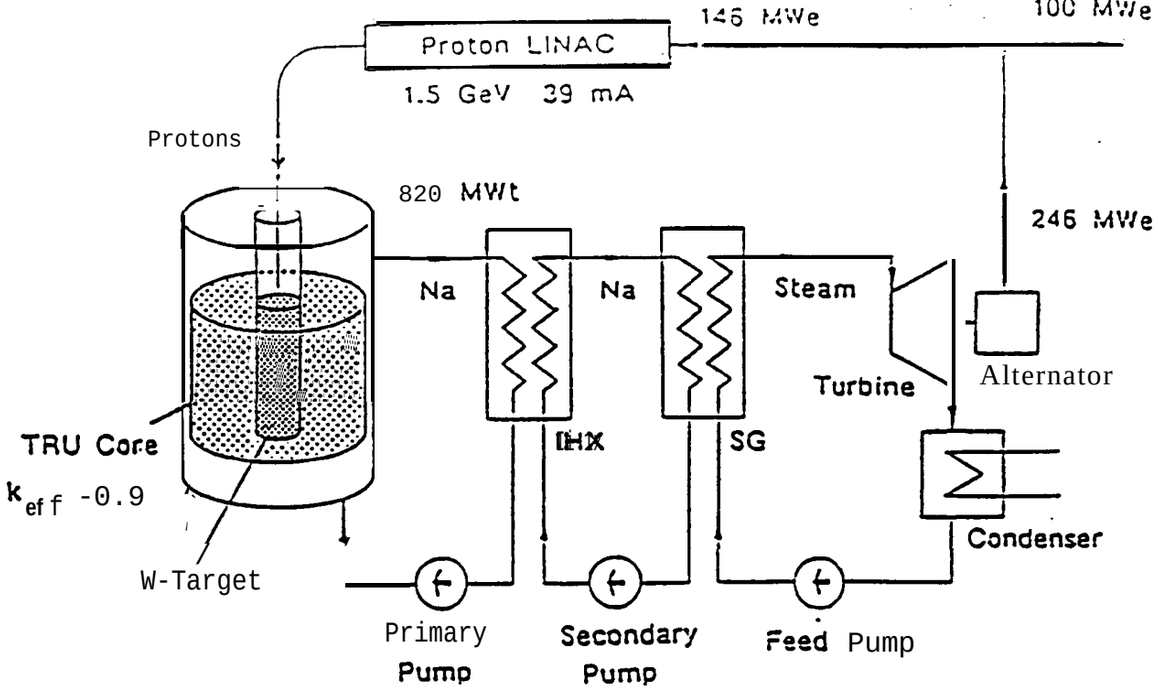


Fig.3.7 Conceptual flow diagram of actinide transmutation plant

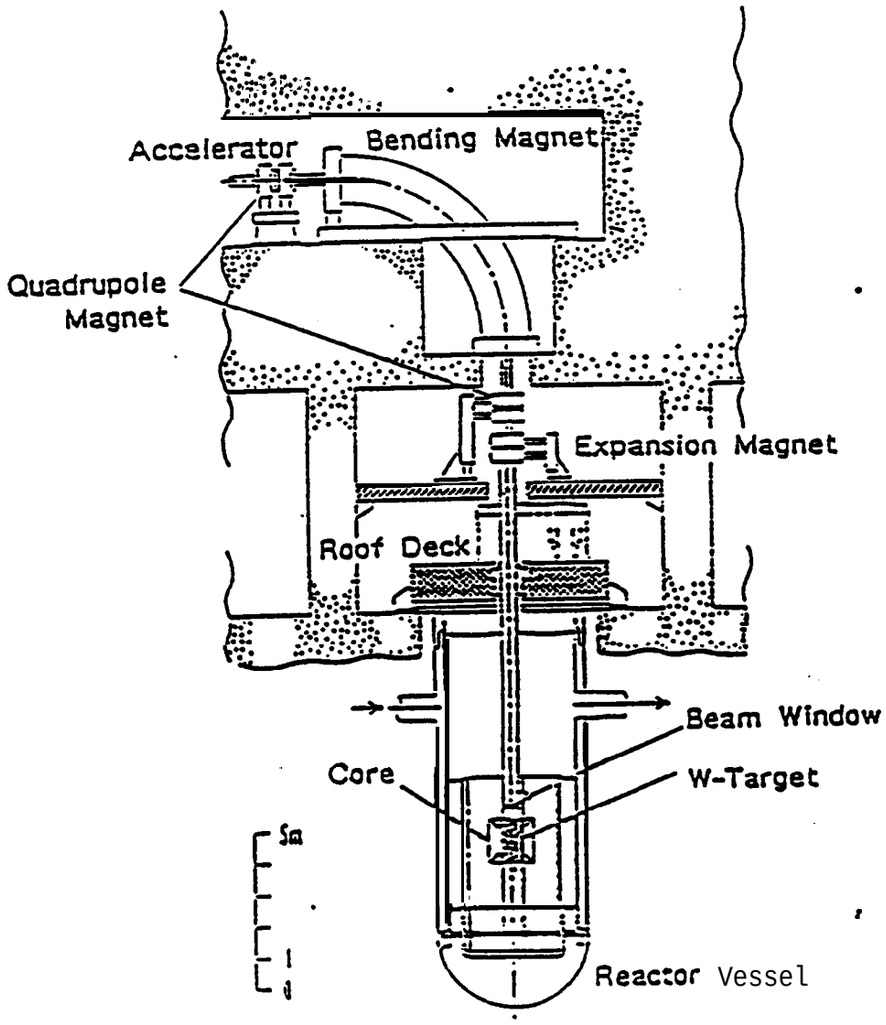


Fig.3.7 Reactor

3.2.3.1. *JAERI project*

In the frame work of the OMEGA project a comprehensive proton accelerator driven transmutation programme has been undertaken by the Japan Atomic Energy Research Institute at Tokai-Mura.

The conceptual flowsheet is shown in fig. 3.7. and refers in many instances to the fast reactor technology.

1. A proton LINAC of 1.5 GeV - 39 mA with an electric consumption of 146 MWe.

2. A tungsten target as the primary spallation source.

3. A core with metallic MA fuel elements surrounds the W target and contains a subcritical (k_{eff} = 0.9) lattice.

4. The Actinide inventory of the core is 3160 kg made up of fuel pins loaded with - Np 15 Pu 30 Zr
   - AmCm35 Pu 10 Y.

5. The designed neutron flux amounts to 4.10^{15} n/cm²/s with a mean neutron energy of 694 keV.

6. The burnup is about 8% or 250 kg Actinides per year of 290 EFPD.

The overall heat output is designed at 820 MWth or 246 MWe (at 30% efficiency). The accelerator consumes 146 MWe and the balance 100 MWe is delivered to the grid.
The Actinide fuel assemblies and the W target are upward cooled by liquid Na. Two core coolant loops coupled to intermediate heat exchangers transfer the extracted heat to a second double Na coolant loop which conveys the energy to the steam generator.

The cost of such a facility cannot yet be accurately established but it may be estimated that the overall investment cost will be twice that of a MAB reactor.

Many features are identical to the fast reactor technology : the fuel loading and unloading machine, the Ar gas blanket to protect the liquid Na from air ingress etc... One supplementary item needs further consideration : the accelerator must be separated from the reactor core by a beam window. This metallic membrane must be very radiation resistant (protons, neutrons) and display a very high thermal conductivity. Any failure of the beam window will shut down the accelerator. Advanced types of ferritic steels are being proposed capable of withstanding up to 130 dpa. but nonetheless it remains the weakest spot in the entire configuration.

The fuel burnup is limited to 8% which implies frequent reprocessing of "spent" metallic fuel pins similar to what has been explained in the section about the IFR concept. By improving the cladding material e.g. by using Oxide-dispersive-strengthened ferritic steels, higher burnups could be achieved and the reactivity due to Pu 238 compensated by a reduced proton beam current. This flexibility is an asset of this concept. The burnup rate being nearly the same (8% versus 7.3% for ABR) the reprocessing costs will be nearly the same. The only way to reduce these costs is by increasing the burnup to e.g. 10 - 12 atom %.

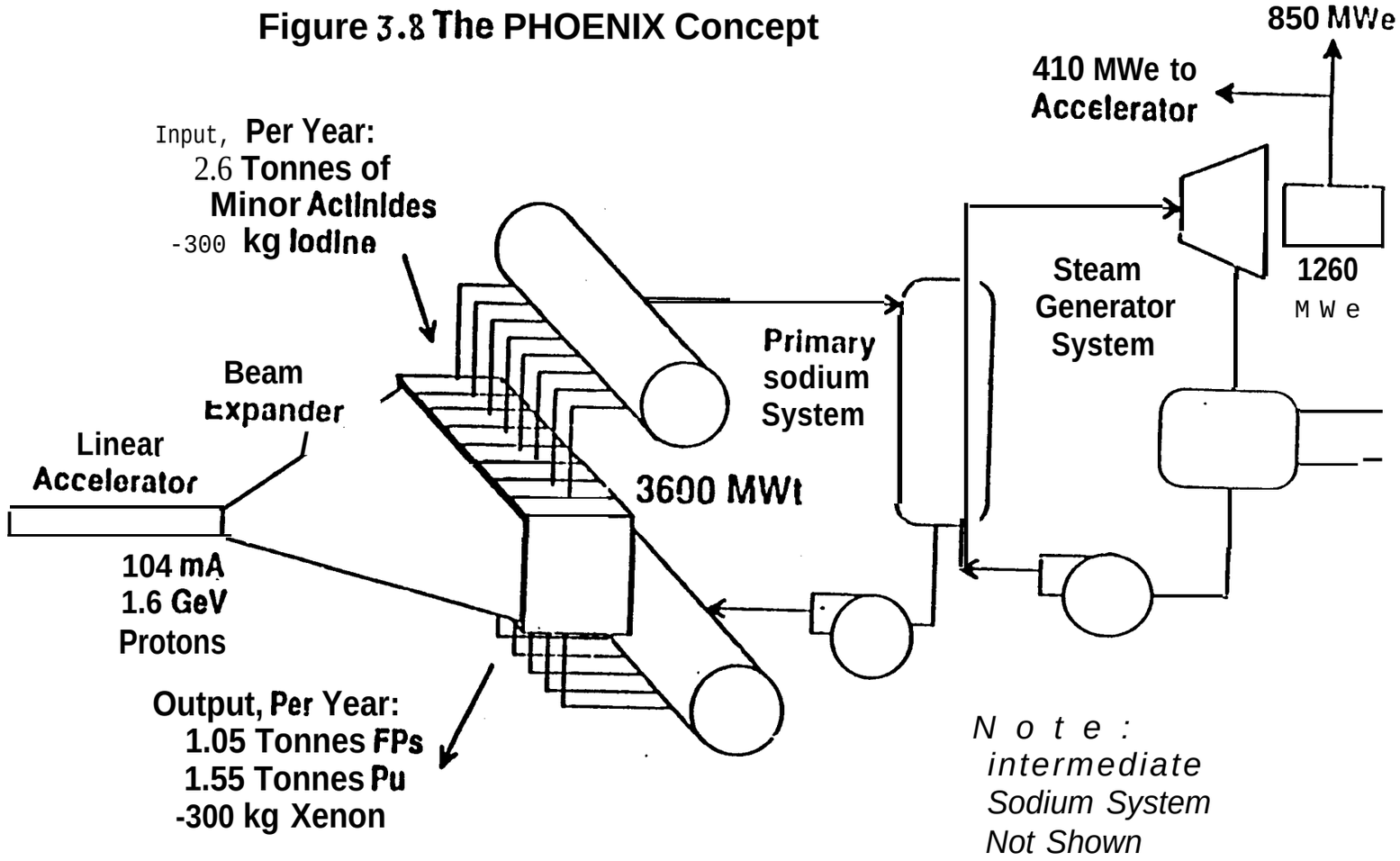
At first glance the incineration of MA with accelerator driven subcritical fast reactor cores will be at least 50% more expensive than the equivalent M.ABR concept. The R&D period to reach the goals of the Engineering Test Accelerator is very long and runs far into the first decade of the next century.

3.2.3.2. *The PHOENIX concept*

In the framework of a comprehensive waste management approach to nuclear electricity production, a project called "Clean Use of Reactor Energy" (CURE) emphasized the need for advanced reprocessing coupled to an accelerator driven transmutation strategy [3. 31].

The reason for this new look on fission energy in the US emerged from the now apparent challenges involved in characterizing and licensing a repository combined with specific regulations of the last several years.

Figure 3.8 The PHOENIX Concept



Real incentives for Actinide burning are more apparent than 10 years ago and will acrue during the next two decades. Current analysis for repositories show moreover that Tc 99, I 129 and Cs 135 are among the most significant nuclides affecting the long term safety.

The potential advantage of an accelerator driven transmutation system, called the PHOENIX concept is based on the coupling of a 1.6 GeV - 104 mA proton accelerator to a subcritical MA oxide-core in which the actinides and long lived fission products would be transmuted into short lived Actinides, fission products and stable isotopes [3.32].

The conceptual flowsheet of the PHOENIX concept is shown in fig. 3.8.
A 3600 MWth accelerator-MOX core, would transmute the MA by fast fission and neutron capture. the project is more ambitious than the JAERI project and is fundamentally different from it by the use of MA in a MOX fuel form and by the associated aqueous reprocessing schemes which are operable on this kind of fuel.

The subcritical core is composed of modules which are identical to a FFTF module with Na cooling. A modular buildup with 450 MWth units, assures a smoother operation of the whole system but is only adapted to a centralized fuel handling facility.

The system is made up of the following units :

1. a 1.6 GeV - 104 mA proton accelerator which requires a 400 MWe grid source to start operation;

2. a subcritical modular MA-MOX lattice composed of fuel pins filled with NpO_2, Am and Cm_2O_3. The lattice serves as a direct target for the 1.6 GeV impinging protons;

3. the neutron flux is expected to range between 4.10^{15} - 10^{16} n/cm²/s and has a somewhat lower mean neutron energy than the MA-metal core. The neutron multiplication factor is very high (450 n/p);

4. the modular target lattices are installed inside a single vacuum chamber avoiding the use of a beam window subject to radiation damage;

5. a secondary Na loop extracts the heat and transfers it to a steam generator of 3600 MWth producing 1260 MWe at 35% efficiency. About 850 MWe can be delivered to the grid;

6. the fission products (Tc 99, I 129 etc...) can be incorporated in special pins to counter balance the reactivity increase of the sub-critical MA.MOX lattice due to the formation of Pu 238, 239 and Am 242.

According to preliminary data the Pu 238 content at the end of a 2 year cycle would amount to 85% of the total Pu content in the transmuted target. Also some Pu 236 is likely to be formed by (n, 2n) reactions of Np 237. Nothing is up to now published on the fate Cm (242-243-244) which might also increase in concentration instead of undergoing fission.
The major merit of this concept is the combination of Actinide and fission product transmutation in a single facility.
The impact of such a concept on the fuel cycle operations is very important if applied to the US nuclear power production :

1. This strategy implies that all spent fuel in the US be reprocessed, an option which is presently not considered.

2. The implementation of advanced aqueous reprocessing (improved PUREX and upscaled TRUEX processes) on industrial quantities.

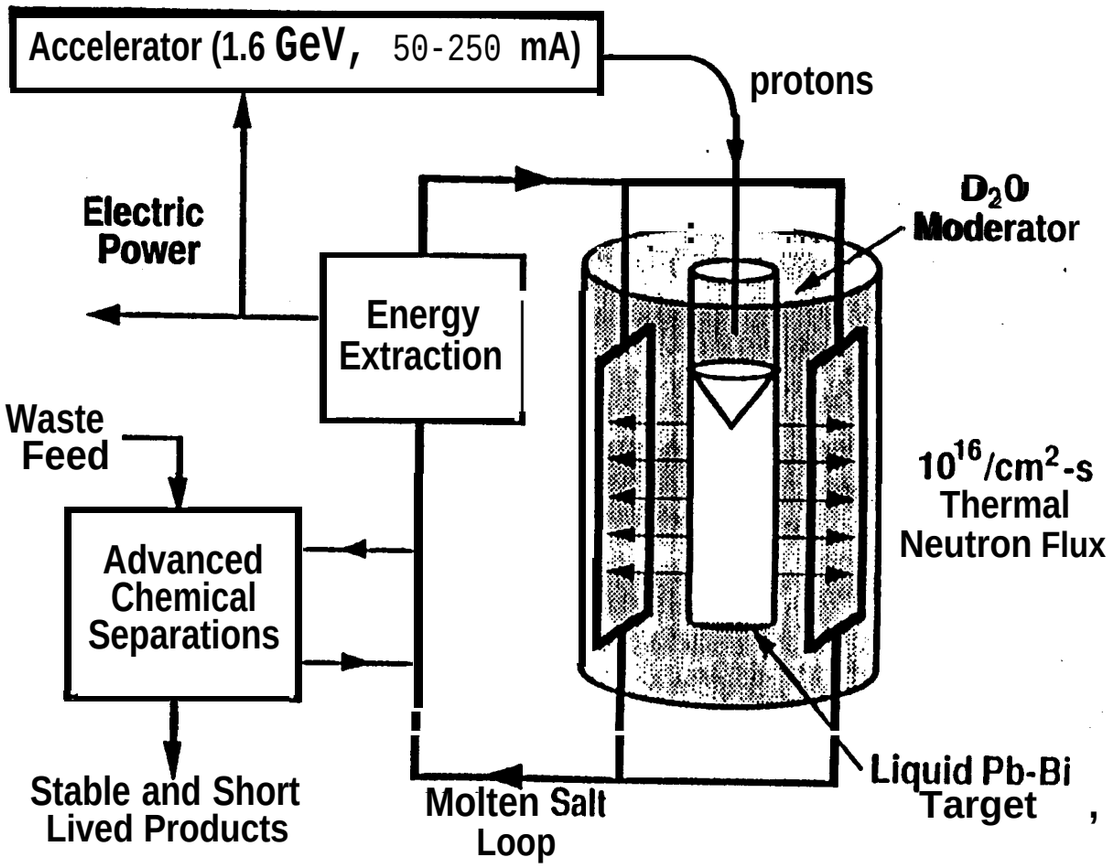
3. Fabrication of Np and Am (Cm) oxide fuel pins in specially designed facilities with remote handling and maintenance.

4. A specially equipped recycle facility of irradiated targets in order to refabricate new irradiation targets, to vitrify fission products and to condition Pu 238 for long term storage (≃ 800 years).

The potential treatment capacity of this conceptual PHOENIX facility is tremendous (2.6 THM/y Np, Am, Cm) and corresponds to the total output of 100 GWe LWR units. The transformation of Np into Pu 238 call for further

Fig. 3.9

- Accelerator driven
- Central, intense neutron source
- Dilute, low material inventories
- Continuous material feed
- Advanced separations chemistry
- Energy production



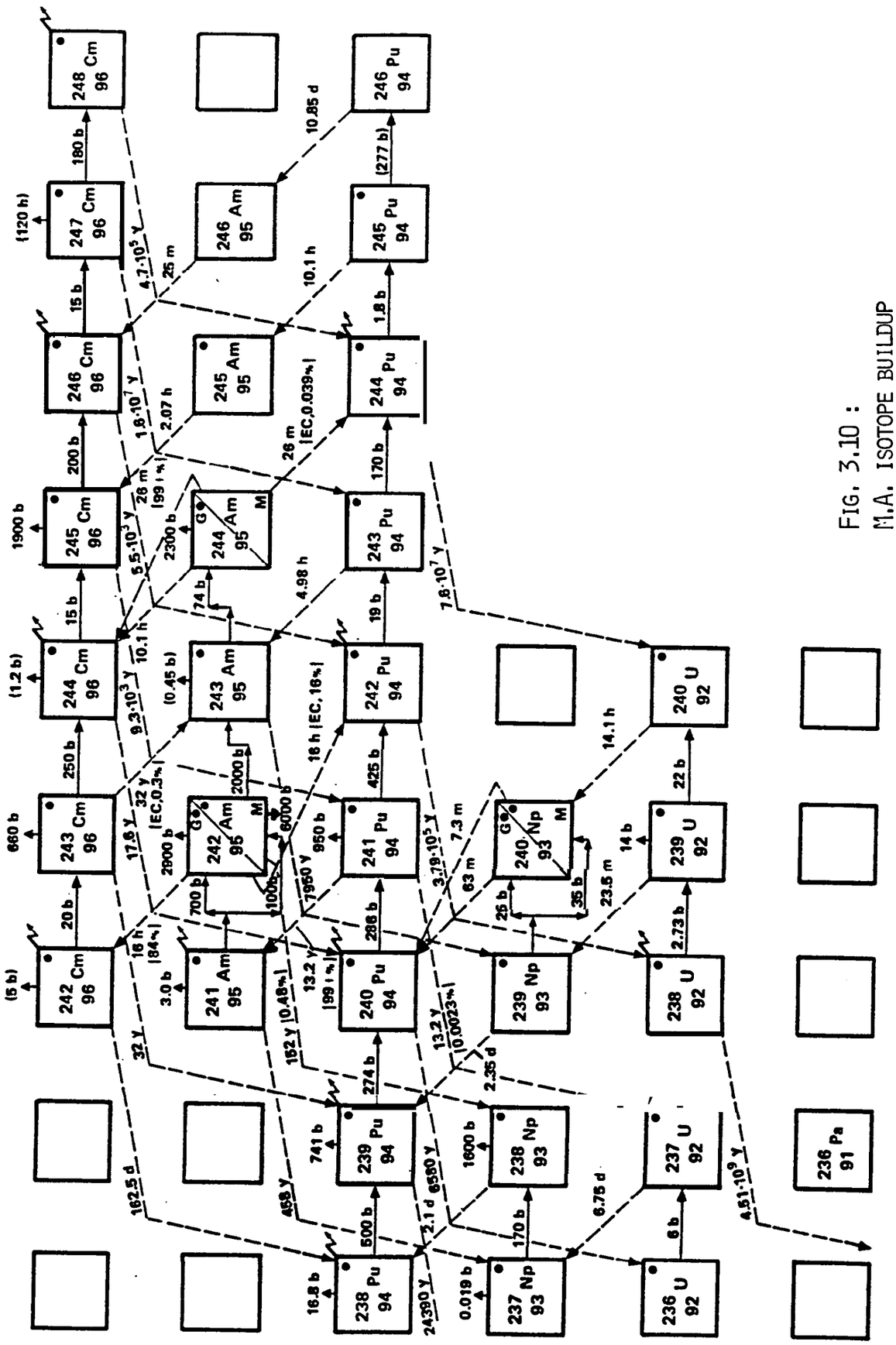


FIG. 3.10 :
M.A. ISOTOPE BUILDUP

study on the overall radiological impact and longterm benefit of this transmutation scenario. It undoubtedly relieves the long term radiological burden of Actinides since it reduces the life time to a mere 800 years instead of the standard *5.000* to *10.000* years adopted for geological disposal.
Further assessment on the economic feasibility of this option should be made in order to compare this concept with reactor incineration.

3.2.3.3. The LOS ALAMOS high thermal neutron incinerator

Recently a new colossal project has been introduced by LANL [3.33; 3.41; 3.42.; 3.43] it consists of the use of a 1.6 GeV - 250 mA proton accelerator coupled to a Pb-Bi spallation source and surrounded by a heavy water moderator.

Fig. *3.9.* shows the conceptual flowsheet.

In contrast with the two above discussed projects and proposals, this concept relies only on the very high thermal neutron flux induced by the protons in the Pb-Bi tank. A proton flux of 1.5 10^{18} can generate a thermal neutron flux of = 10^{16} if the spallation target is surrounded by a very effective neutron moderator-reflector. In this concept heavy water is proposed as moderator.

Another feature of the project is its low actinide inventory. In a very intense thermal neutron flux nuclides with low cross section or being present at very low transient concentration levels can efficiently be transmuted and/or fissioned.
For example in the case of the MA an intricate series of neutron captures and fissions occur in conjunction with the natural decay. Fig. 3.10. shows the data for the most important MA nucl ides [3.18].
The capture cross sections are arranged horizontally, the fission cross sections vertically and the dashed lines indicate the appropriate half life.

Particularly important are Np 238 with a fission cross section of 1600 b and a half life of 2.1 days, and Am 242 with a fission cross section of 2900 b and a half life of 16 h. These nuclides will be efficiently fissioned in such an intense neutron flux.

The n-2n and n-3n reactions which occur in fast neutron fluxes are not supposed to occur in this type of "activation" reactor.
Since the whole reactor vessel is free of fissile or fertile material only the radionuclides introduced into the systems for transmutation make up the inventory.

In order to provide sufficient residence time inside the reactor vessel a circulation loop is foreseen to keep the nuclear materials till a certain degree of depletion has been achieved.
In the present configuration 75 - 100 kg MA and 300 - 350 kg FP could be transmuted per year. From pure radiochemical point of view this is a remarkable proposal.

In comparison with the two other accelerator driven systems the concept is made up of the following main units.

1. A 1.6 GeV - 250 MA continuous-wave-proton accelerator which requires a 1000 MWe off site power source to start the reactor.

2. A Pb-Bi tank filled with molten lead-bismuth mixture which serves as spallation source with a neutron multiplication yield of 55 n/p.

3. The spallated neutrons are moderated in a heavy water tank of about 10 m³ which is cooled by a molten salt loop in which the actinides are dissolved to undergo fission. The neutron flux is about 10^{16} n/cm²/s and the transmutation yield 75 - 100 kg/year.

4. An aqueous flow of fission products can also be funnelled through the heavy water tank in one or more separate loops (Tc 99, I 129,...).

5. In the present proposal no fission power recovery has been considered. If a substantial amount (≃ 100 kg) of fertile or fissile nuclides could be accommodated in the "reactor vessel" a substantial amount of energy could recovered but it would very much affect the engineering of the proposed transmuter-incinerator.

It appears at first glance that the project is by nature very expensive since a 1000 MWe off site power source must be available to start up and operate the system. In case later on a subcritical reactor vessel would be added to the present proposal a certain fraction of the energy could be recovered but it would not equilibrate the present energy demand unless larger fissile material inventories are provided.

The intense neutron flux inside a heavy water moderator (1000 times higher than in CANDU reactors) will enhance the tritium production and the radiolysis.

In this present state of the proposal it is not obvious how a molten Pb-Bi target is kept separate from the accelerator vacuum chamber.

If a beam window has to be installed, the same type of problems as discussed in the JAERI proposal will have to be resolved but in a higher degree since the proton flux is 10 times higher and the resulting lifetime lower.

The molten salt chemistry of actinides is in principle very well known but with the multitude of radionuclides involved, it is a very complex issue.

The proposal seems to be more adequate for fission product transmutations than for actinides.

Independent of personnel and investment costs, this proposal would be outrageously expensive with the presently proposed Actinide throughput

$$\frac{1000\ \text{MWe} \times 8750}{100\ \text{kg} \times 0.075} = 6.5\ 10^{4}\ \text{MWh/kg}$$

or about 4-10 M$/kg fissioned actinide depending on the KWh cost of a dedicated plant.

The time span to realize an optimized concept with partial energy recovery is very long since RD&D work is to be done on the upscaling by a factor of 250 of the beam current. The time span between the present principle flowsheet and the industrial demonstration may easily last 10 to 20 years or more. The prospects and problems associated with Accelerator driven systems for transmutation purposes have been thoroughly assessed by Energy Research Advisory board on the basis of a proposal from Los Alamos National Laboratory for the construction of a Tritium production facility.

3.3. LONGLIVED FISSION PRODUCTS CONDITIONING AND TRANSMUTATION

3.3.1. Iodine 129 [3.34]

The transformation of iodine trapped in scrubbing liquids or on silver zeolites into pure target material ready for irradiation in a HFR is much more difficult than could be expected at first glance. The elements accompanying the iodine must be removed in order--to reduce and concentrate the iodine fraction and to reduce the neutron loss due to activation of chemical impurities.

iodine in scrubbing liquids can be removed from the scrubber solution as molecular I_2 by oxidative sparging;

iodine present in the AgI filter materials is very difficult to extract.

But since the bulk of the iodine is caught in the caustic scrubbers, it is not necessary to recover this residual amount (< 2 %).

Iodine can easily be transformed into iodate and precipitated with Ba or Pb salts. Because of its chemical stability and insolubility, preference should be given to Ba $(IO_3)_2$ as target material which can afterwards also be used as final storage matrix. $Ba(IO_3)_2$ is furthermore thermally stable till 700°C.

The low cross-section of the neutron capture and the low transformation yield does not impose an additional dilution of the product in the capsule.

The elemental dilution of Iodine in $Ba(IO_3)_2$ amounts to a factor of 4. which means that for 1 GWe year ^{129}I output a total amount of 25 kg $Ba(IO_3)_2$ has to be irradiated.

During the burnup in the reactor, stable ^{130}Xe is formed, which has to be vented in order to reduce the internal pressure in the capsule. A reliable venting system capable of withstanding very long residence times in the reactor has to be mounted on the capsule.

Very much attention should be paid to the temperature control of the capsule to avoid temperature excursion above 700°C which would destabilize the chemical compound and liberate highly corrosive iodine vapours.

3.3.2. Technetium 99

^{99}Tc occurs for 50 % in the insoluble residues, together with Pu, noble metals and short-lived fission products (^{103}Ru, ^{106}Ru). Taking into account the high radioactivity of these fission products, it is very difficult to carry out chemical separations before 25 years cooling [3.35]; Some pyrochemical techniques based on a leaching with liquid magnesium and lead at temperatures of 950°C have been reported [3.36] [3.37].
On the separation from HLLW work has been reported in Japan [2.23]. A group separation together with other noble metals might be envisaged as intermediate step before further elemental separation.

In Japan work is in progress on the pyrometallurgical separation of Ru, Rh, Pd, Tc from insoluble residues [3.38].
The transformation of any chemical Tc form into the very insoluble TcO_2 compound or into metallic Tc is the most obvious way to prepare irradiation targets for long duration irradiation in safe conditions.

The quantity of ^{99}Tc produced by the LWR fuel irradiation amounts to 21 kg/GWe year, such relatively large amounts require the development of new large high flux irradiation reactors capable of handling such large targets. Since ^{99}Tc decays into stable ^{99}Ru, no special requirements are imposed on the capsule design.

Spallation neutron sources [3.39] have been proposed to transmute isotopes with very low thermal neutron cross sections eg ^{99}Te and ^{129}I.

A 1.6 GeV, 250 m A proton accelerator produces a beam which generates a neutron source of 10^{20} n/s when impinging on a Pb-Bi target. After some thermalisation a local neutron flux of 10^{16} n/cm²/s can be achieved resulting in a very rapid transmutation (100 times faster than in a HFR.) of the isotopic target (Tc) dissolved in Pb-Bi alloy or transported through the moderator. Two dimensional neutron transport calculations have shown that with a 250 mA proton beam a transmutation rate of 500 kg/year could be obtained.

However the technological effort required to reach such high beam intensities is enormous and it will take several decades to accomplish this ambitious goal.

3.3.3 Cesium 135

Cesium ,135 is present in the HLLW as a result of the fission process which produces 324 g/THM or 8.75 kg/GWe.year.
This isotope has a half life of 2.10^6 years and is always accompanied by the overwhelming quantity of Cs 137 (1069 g or 28.8 kg/GWe year) emitting a γ ray of 0.6 MeV.
Isotopic separation between both isotopes is nearly impossible or extremely expensive. Both radionuclides together make up a Cs source of roughly 37.5 kg per GWe-year.
The separation from HLLW can be performed with inorganic ion exchangers but in order to prepare an irradiation target, separation with liquid extraction techniques is more versatile. A liquid extraction process [3.40] using Cobalt - dicarbollide dissolved in a polar solvent, has been developed at the Khlopin -Radium Institute of Leningrad. The process has been upscaled to pilot scale level and produces relatively pure Cs 137 + 135.

Due to the high activity of Cs 137 (95.000 Ci/THM) and the half life of 30 years, delayed reprocessing or aging of the HLLW has no influence on the technology to be employed i.e. heavily shielded hot cells, remote manipulation and maintenance and very active as well as hot targets.

Cesium can be transformed into CsCl or Cs_2O/CsOH or CSI. The very low cross section of Cs 135 ie 8.7b is much too small to be useful in thermal reactors unless fluxes of 10^{16} n/cm²/s are available.

Other transmutation devices should be assessed in order to determine what can be done in such a difficult case.
Transmutation by photonuclear reaction is one of the possibilities since linear electron accelerators at GeV energy levels are readily available in the high energy physics facilities.

The inverse compton method [3.41] produces a tunable quasi mono energetic gamma photon. The impinging γ ray interacts with the Cs atoms and results into a neutron emission from the Cs nucleus which decays into stable Ba./No data are available for Cs135 and this subject should be studied in order to assess the potential of this method.

eg. $^{137}Cs\ (\gamma\ n)\ ^{136}Cs \rightarrow\ ^{136}Ba$ (stable)

However the main problem to be solved lies in general with the current stability, the intensity of the beam (a few mA presently) and with the interaction probability between a high energetic photon and the Cs nucleus. A combination of these drawbacks will substantially decrease the transmutation rate and increase the specific energy consumption.

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