IMPACT OF FISSION PRODUCT PARTITIONING AND TRANSMUTATION OF Np-237, I-129 AND Tc-99 ON WASTE DISPOSAL STRATEGIES

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Introduction.

The separation of fission products as a means to simplify the HLW management dates back to the early sixties when it was assumed that by separating ^{137}Cs and ^{90}Sr from HLW the long-term problem was solved. Unfortunately, the presence of actinides in the HLW imposed a long-term solution for the storage and disposal of these wastes. Vitrification emerged as the ultimate solution for the conditioning of HLW and for indefinite storage in geological repositories.

In the early seventies however, the permanent presence of very toxic radionuclides for thousands of years in repositories was considered a risk the human race could not control and thus ought to be avoided. Alternative scenarios comprising separation of actinides from HLW and transmutation of these nuclides in LWR, FBR and HTGR received considerable attention, until it was recognized that geological disposal of radionuclides cannot be avoided and that transmutation scenarios end up with an actinide residue which has to be stored and disposed of.

Extensive information on all aspects of Partitioning and Transmutation (P.-T) was presented in the first [1] and second [2] conference held at Ispra in 1977 and 1980. A final assessment of the studies on transmutation by JRC was published in 1983 [3] and on separation on 1984 [4].

The overall conclusions were rather pessimistic with regard to feasibility, cost and risk reduction.

A US study carried out in the period 1976-80 [5] concluded on the theoretical feasibility of actinide partitioning, but drew the attention on the many non-resolved questions with regard to fuel reprocessing schemes, MOX fuel fabrication technology, type of transmutation reactor and auxiliary problems e.g. transportation of actinide wastes. The most important conclusion however was that in the most favorable option for P.-T. of actinides, the expected long-term benefit was only 3 man-Sievert/GWe, which is small compared to a threshold value of 46 man.Sv/GWe. This value is considered a lower limit level for justifying additional operations.

The benefits of ^{99}Tc and ^{129}I partitioning were estimated to be 300 man.Sv/GWe, which is 100 times more important than actinide partitioning and far above the 46 Man.Sv/GWe level. The incentive to investigate ^{99}Tc and ^{129}I partitioning-transmutation schemes is much more justified and its possibilities will be discussed in this paper. With respect to actinide partitioning and transmutation a selective approach might be more realistic. A limited effort on ^{237}Np separation and transmutation is worthwhile to be studied experimentally in order to ascertain whether a small change in the current reprocessing operations could alleviate the long-term hazard of this radionuclide.

In 1988, the Japanese government launched a very ambitious long-term programme, called OMEGA [6], incorporating not only radioactive product recovery (Cs, Sr, Actinides) but also noble metal (Ru, Rh, Pd) recovery with the major aim to reduce the long-term risk and at the same time to save precious resources [7]. This meeting will enable us to discuss the issues at stake and to lay down the grounds for international collaboration in the framework of the OECD.NEA.

Prospects and issues for a limited partitioning of ^{237}Np, ^{129}I, ^{135}Cs and ^{99}Tc.

Among the countries having taken the option of a nuclear fuel cycle with reprocessing, there is a general consensus on the following operational sequences.

LWR fuel discharged from the reactors at a burnup of 33,000 (up to 45,000) MWd/T is stored for at least one year in water ponds at the reactor site, transported to a central "Away from Reactor" pool, generally situated at the reprocessing plant sites (La Hague, Sellafield, Tokai). Three to five years after discharge from the reactor, the fuel is reprocessed leading to the recovery of valuable energy resources (U, Pu) and so-called waste products (HLW, MLW and LLW).

Apart from the principal actinides, U and Pu, very little attention was given to the fate of ^{237}Np which ends up partially in the HLLW and partially in the purified U or Pu fraction, depending on the specific separation flowsheet.

Among the HLW streams two separate fractions have to be considered :

- The insoluble residues from the dissolver-clarification step containing mainly noble metals (Ru, Rh, Pd) and Tc.
- The soluble HLLW which contains all the fission products and Actinides except U and Pu, which are removed for 98-99%.

The MLW streams are generally very diluted and treated by chemical methods to reduce the environmental impact of liquid discharges. However, iodine, particularly ^{129}I, is separated from the other MLW streams at the very onset of the reprocessing operations. In the head-end step 99% of the iodine is transferred from dissolver off-gas (DOG) to scrubbing liquids or iodine absorbing filters. At present the iodine-containing liquid effluents are eventually discharged into the sea (UK, France) unless specific storage facilities are provided for iodine-loaded solid filtering materials (Zeolites).

The reference reprocessing flowsheet transforms all the HLLW

and the insoluble residues into borosilicate glass which is a waste form excluding further recovery processes. The canisters filled with glass are supposed to be stored till final disposal in a geologic repository. The eventual disposal in deep geological structures is supposed to confine all the radionuclides and protect the biosphere from radionuclide contamination for geological periods. However, the half life of four radionuclides combined to their radiological toxicity does not guarantee a perpetual confinement, whatever geological structure is considered. These are : ^{237}Np, ^{129}I, ^{135}Cs and ^{99}Tc.

Table I lists the most important characteristics.

Radiochemical characteristics of some long-lived radionuclides

	^{237}Np	^{129}I	^{135}Cs	^{99}Tc
Half-life	2.14 10^6 y	1.7 lo7y	3 10^6 y	2.12 10^5 y
Emission	α : 4.95 MeV	β : 0.189 MeV	β : 0.21 MeV	β : 1.7 MeV
Chemical form	Np IV-V and VI	I_2, I^-, I^{n+}	Cs^+	Tc(IV)-TcO_4^-(VII)
Ci/T HM (at 33 GWd/T)	0.31	0.0312	0.352	13.2
g/T	438-(820) ,	190	397	766
g element/T	438-(820)	230	(2470)-1330	766

Since ^{137}Cs is one of the major fission products, it is obvious that ^{135}Cs cannot be separated by any means from the FP before a cooling time of 500 years or more and therefore, it will not be discussed any further.

The partitioning of ^{237}Np from the other actinides is not very difficult but has to be conceived in such a way that the recovery yield of the major actinides (U, Pu) remains unaltered. It would be perferable to limit the amount of Np in the HLLW to as low-as-reasonably-achievable value. The recovery operation of Np (and other actinides) from HLLW is a very complex process [8] which needs important additional equipment.

From the long-term point of view, we have to take into account the ^{237}Np formation from ^{241}Am directly formed during reactor operation and the quantity building up as daughter product of ^{241}Pu [Fig. 1].
The total amount of $^{(241+243)}Am$ discharged after 5 years into HLLW amounts to 466 g/T. From this amount about 382 g ^{241}Am decays directly into ^{237}Np within a period of 10,000 years. This period coincides with the expected technical lifetime of the repository structures and confinement aids. However, beyond 10,000 years the newly formed ^{237}Np will constitute the long-term source for actinides in the geosphere. Its diffusion will largely depend on the long-term stability of the glass, but it is not reasonable to accept a glass stability beyong 10^6 years.
By complete transmutation of ^{237}Np, separated during reprocessing, the source term is only reduced from potentially 820 g/THM to 375 g/THM, or a reduction with a factor of 2.2. In order to further reduce the long-term ^{237}Np impact, it would be necessary to separate ^{241}Am from HLLW and to incinerate this radionuclide in a FBR, or to transmute ^{241}Am into higher Am-isotopes in a HFR, which leads to the formation of short-lived Np-isotopes as decay products. The drawbacks of this approach are : formation of secundary waste, safety problems with concentrated Am-targets in a reactor, etc. . . It is not obvious whether a further reduction of the Np source term through the separation of Am from HLLW will reduce the collective dose to the workers and the population at large.
In the reprocessing option Pu is for about 98-99% recycled as PuO_2 to be used as MOX fuel in LWRS and FBRs. If the recovered Pu from the PUREX reprocessing process is used immediately, the ingrowth of ^{241}Am is sufficiently low to neglect its contribution in a long-term prospect. Reirradiation of this MOX fuel results in a smaller Np formation (310 g/THM) instead of the reference value (480 g/THM). This is due to a lower contribution of the ^{236}U (n) $^{237}U \xrightarrow{\beta} {}^{237}Np$ reaction chain. But after several recycling the equilibrium value of 500-580 g/THM is attained. The formation depends

largely on the ^{240}Pu level, the residence time in the reactor, the neutron flux and spectrum.

If the recovered Pu is not used immediately, ^{241}Pu will decay at a rate corresponding to its half-life of 14.4 years into ^{241}Am and the ^{241}Pu concentration will drop from 13.8% in fresh spent fuel to 8.3% after 10 years [9]. This 5.5% drop in (fissile) ^{241}Pu concentration has negative effects on the reactor economy, but has also a radiological impact on the fuel manufacturing process which becomes difficult without remote handling. Therefore, it would be advantageous to reprocess this "old" Pu before bringing the purified Pu into the MOX fuel manufacturing plant but this operation is very expensive (9 - 18 $/g Pu).

From waste disposal point of view this purification step would have the advantage to separate ^{241}Am in a pure isotopic form which is suitable for use in "incineration" or "transmutation" processes leading to a nuclide mixture which does not lead to ^{237}Np formation by decay.

As a conclusion we can say that whatever strategy is adopted towards ^{237}Np, a certain fraction will always be found in the final disposal route, either as residue from "on PurPose" transmutation or as decay product of ^{241}Am.

The separation of ^{129}I from Dissolver Off Gases is a standard procedure at the fuel reprocessing plants because of the potential local buildup of this radionuclide in the biosphere surrounding the plant and its danger for man.

The scrubber liquids contain all iodine compounds (^{127}I, ^{129}I and ^{131}I) and some nuclides entrained in the off gas stream as aerosols (Ru, Sb, ...). The saturated liquid can be used directly as the ^{129}I source for conditioning and storage or, as is the case in the UK and France, discharged into the sea under controlled conditions.

The techniques for trapping and conditioning iodine have been studied extensively in the past [11]. The most important alternative to caustic scrubbing is the direct sorption on silver impregnated sorbents. This option is very attractive from waste management point of view since the iodine is

very firmly held by a solid material which can be kept for as long as deemed desirable. In the long term the stored iodine loaded material will have to be disposed of in an underground repository for ultimate confinement. In both fully opposite management options : direct dilution and geologic confinement, the most appropriate one depends on the siting of the reprocessing plant and on the availability of a suitable repository.

The impact of direct discharge into the ocean has been assessed for reprocessing plants located in North Western Europe. The maximum individual thyroid dose due to molluscs and crustacea amounts to 0.17 mSv/y at a yearly discharge rate of 2 TBq. The regional collective dose remains very small in the first centuries but could reach 150 man.Sv after a long period of time. As long as the reprocessing capacity throughout the world remains at the present level, discharge of iodine-129 effluents could be an acceptable intermediate practice. However, in the long term the accumulation of ^{129}I in world's oceans will increase the doses to mankind. Without sedimentation the global collective dose commitment reaches a value of 2.5 10^4 man.Sv.

The alternative Iodine management option with storage of solid sorbents and eventual disposal has to take into account that Iodine is a very mobile element in the geosphere. In a clay repository the long-lived actinides (except ^{237}Np) can be confined for geological periods without noticeable dose to man [12] but recent calculations in the framework of the PACOMA project of the CEC have shown that iodine-129 cannot be neglected [13][Fig. 2].

In case iodine should be retained at the reprocessing plant and that the resulting iodine waste has to be disposed of in a geological repository (Boom clay in Belgium) the maximum calculated dose rate might rise to a value of 0.54 mSv/y. According to a normal scenario the ^{129}I activity would reach the neighboring aquifer after about 5,000 years and the maximum flux occurs around 5.4 10^4 years with a radionuclide flux resulting in an annual dose rate of 0.1 mSv/year, which is 10% of the natural background. The waste conditioning

form (cement) does not exert any influence on the release rate from a AgI type iodine waste source. As a conclusion we can state that geological disposal of ^{129}I in clay is not a suitable and final solution for this problem.

ICRP recommends a dose limit of 1 mSv/year and for local contributions to the background 0.1 mSv/y. The potential contribution of ^{129}I in iodine waste equals or surpasses that criterion.

Transmutation of ^{129}I into a short-lived ^{130}I decaying into stable ^{130}Xe is an alternative strategy which ought to be assessed in order to investigate its feasibility and economics.

Technetium-99 is the third most important long-lived radionuclide which cannot be confined within a geologic repository because of its mobility as TcO_4^- coupled to its half-life of 2.1 10^5 years.

The reference option presently selected by the major reprocessors is the vitrification of the combined insoluble residues and HLLW. In this scenario all the insoluble residues which contain appreciable amounts of Pd, Rh and Ru are incorporated in a glassy matrix and practically unrecoverable in the future. Leaching of the glassy matrix will eventually transfer ^{99}Tc from the source to the environment. The problem is obviously much less preoccupying since the maximum dose rate due to ^{99}Tc is about 10^{-8} Sv/y and this level is only reached after 1.1 million years. However, when comparing with other nuclides leached from vitrified HLW, it is still the most important contribution to the radiologic burden of HLW after ^{129}, ^{237}Np and ^{135}Cs.

By simple chemical treatment (reduction or sulphide precipitation) the soluble “noble metals” including Tc can be separated from HLLW and stored together with the rest of the insoluble residues produced during the clarification. storage of this waste stream for several decades would eliminate the major radioactivity due to ^{106}Ru and could be used as a resource for noble metals [7]. At that stage the separation of Tc from Ru, Rh and Pd could be performed according

to existing precious metal refinery techniques.
A principle advantage of this scenario is the recovery for future generatiOnS of important mineral resources which would be available for industrial applications. The residual ^{99}Tc metal could be used as a target for transmutation in HFRs or FBRs. By neutron capture ^{99}Tc is transformed into ^{100}Tc (15.3 s) and decays to stable ^{100}Ru.

Separation and conditioning of Np, I and Tc for reactor irradiation.

1. Neptunium

Neptunium-237 constitutes 3% of the Pu mass which implies that its quantitative separation (12 to 13 kg/GWe year) requires an adaptation of the dissolution-extraction process during reprocessing in order to avoid a dispersion of Np throughout the different streams of the PUREX process.
After dissolution and valency adjustment of the feed solution Np is to the largest extent present as Np VI with some unextractable Np V. By adding an oxidant (e.g. $_{2}°_{5}$ in 3M HNO_3) at a specific level in the extraction column behind the extraction sections of U and Pu, a quantitative oxidation to Np VI takes place and its extraction in the subsequent stages of the extraction column becomes possible [14]. The organic phase contains U VI, Pu IV and Np VI as $TBP.NO_3$ complexes. In order to accommodate for quantitative Np recovery without loss of Pu, some slight modification has to be brought to the reference PUREX extraction cycle. But since the generalized use of U(IV) as reductant for Pu and the specific use of hydroxylamine nitrate as reductant for Np VI to Np V has been adopted, easy recovery within standard reprocessing equipment is possible [15].
The technology for transforming Np solution into Np.Al alloy is standard for several decades [16] in the framework of the ^{238}Pu production. However, in this case special attention must be given to the heat problem and to the integrity of

the irradiation capsule. The production of ^{238}Pu was optimized in cyclic irradiations to reduce the ^{236}Pu contribution and higher mass components ($^{239,240,\cdots}Pu$) and therefore the residence time of each capsule was limited to a few HFR cycles [17]. If longer irradiations, e.g. one to several years, in a high neutron flux are required, the capsule design and the proper choice of the encapsulation materials are of fundamental importance for the reactor safety.

Very large experience exists at the HFR sites which carried out research on material testing under extreme conditions during the past twenty years. The BR2 reactor at Mel, Belgium, with a mean flux of 2.10^{14} n/cm².s participated in the actinide irradiation programmed (Ra, Ac, Th) and the Transuranic elements development programme. The most important irradiations were done on the 100 g scale with $RaC0_3$.

However, for industrial programmed e.g. transmutation of Np from fuel reprocessing into ^{238}Pu, large scale facilities capable of irradiating 10 to 100 kg per year ought to become available in the future. The present HFRs are not capable of handling such large quantities.

2. Iodine-129

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 1_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 ($\leq$ 2%).

Iodine can easily be transformed into iodate and precipitat-

ed 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 I 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. Technetium

^{99}Tc occurs for 50% in the insoluble residues, toaether 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 [7]. Some pyrochemical techniques based on a leaching with liquid magnesium and lead at temperatures of 950°C have been reported [18][19].

On the separation from HLLW no methods have been reported up to now. A group separation together with other noble metals might be envisaged.

In Japan work is in progress on the pyrometallurgical separation of Ru, Rh, Pd, Tc from insoluble residues [20].

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.

Irradiation of ^{237}Np, ^{129}I and 'gTc in HFR.

The transmutation of the targets discussed above cannot be achieved in a LWR flux, because otherwise extremely long irradiation periods are required. In a HFR with fluxes ranging from 1 to 5 10 14 n/cm²/sec the transmutation of 90% of the initial target does not seem impossible. Table II shows the cross section and the expected transmutation yields after the specified time periods.

Table II

Transmutation yields of ^{237}Np, ^{129}I and '^{9}Tc in HFRs.

	^{237}Np	^{129}I	^{99}Tc
σ_c (th)	170 b ± 5	19 b (to ^{130}I m)	22 b
σ_g	19 mb ± 3	9 b (to ^{130}I)	
RI	640 b	36 b	300 b
Irradiation time	Expected Transmut. Yields at 2.10^{14} n $cm^{-2}s^{-1}$		
200 days	17%	10%	18%
500 days	87%	23%	36%
1000 days	98%	41%	59%
2000 days	>99.9%	65%	83%
4000 days	>99.9%	88%	97%

From these data, which do not take into account the neutron shadowing, it is obvious that 90% ^{237}Np transmutation can be performed in a reasonable time. The transmutation of ^{129}I and ^{99}Tc is technically possible but requires extremely long irradiation times (about 10 calendar years) to reach the 90% target.

Taking into account the operational cost of HFRs and their limited loading capacity, it is very improbable that industrial scale transmutation of long-lived nuclides can be achieved in the present generation HFRs.

However, in view of validating some concepts on representative scale and in order to confirm theoretical transmutation yields, experimental irradiation for long periods of time in HFRs (type BR2) could bring important information on this subject.

The non-negligeable contribution of the epithermal flux in the resonance capture could be optimized to improve the yields.

Conclusions

1. From the point of view of waste management, three isotopes ^{237}Np, ^{129}I and ^{99}Tc deserve particular attention since they cannot be confined within any presently known geological barrier. The safety assessment studies showed that ^{129}I diffuses through 50 m of clay after 5000 years and creates a radiological burden which is superior to the other nuclides. ^{99}Tc and ^{237}Np emerge much later (million years) but cannot be confined in the repository.

2. Separation of ^{237}Np is already envisaged by the reprocessing plants and small modifications to the PUREX process permit to separate this nuclide quantitatively. However, the overall Np reduction factor cannot be higher than 3 to 4 because ^{237}Np is formed by alpha-decay of ^{241}Am which is immobilized in the glass. The irradiation technology to transmute ^{237}Np into ^{238}Pu is available.

3. Iodine is separated from the dissolver off-gases in the reference fuel cycle and could be transformed into a stable $Ba(IO_3)_2$ matrix which can serve as irradiation matrix to destroy ^{129}I.

4. Technetium is produced as insoluble residue and occurs in the waste streams together with Ru, Rh and Pd. It's recovery is still questionable and needs further developments.

5. Irradiation of small targets in HFRs can lead to a 90% transmutation of these long-lived isotopes but the irradiation periods (particularly for ^{129}I) are very long. The present HFR generation is not designed to handle large amounts of long-lived radionuclides and new more powerful units ought to be designed for that purpose.

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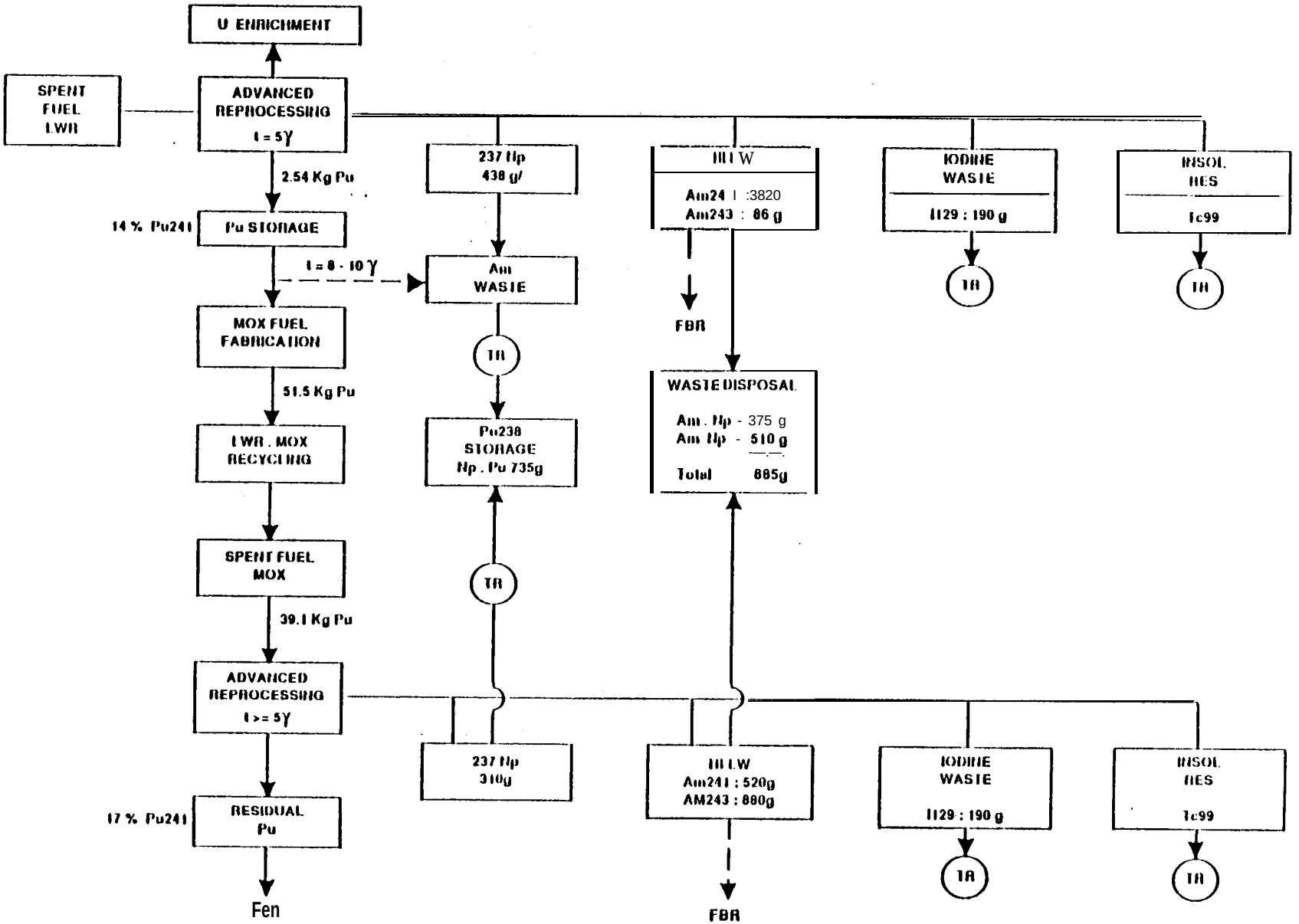

Fig. 1 : **Partial Partitioning Transmutation** Scheme

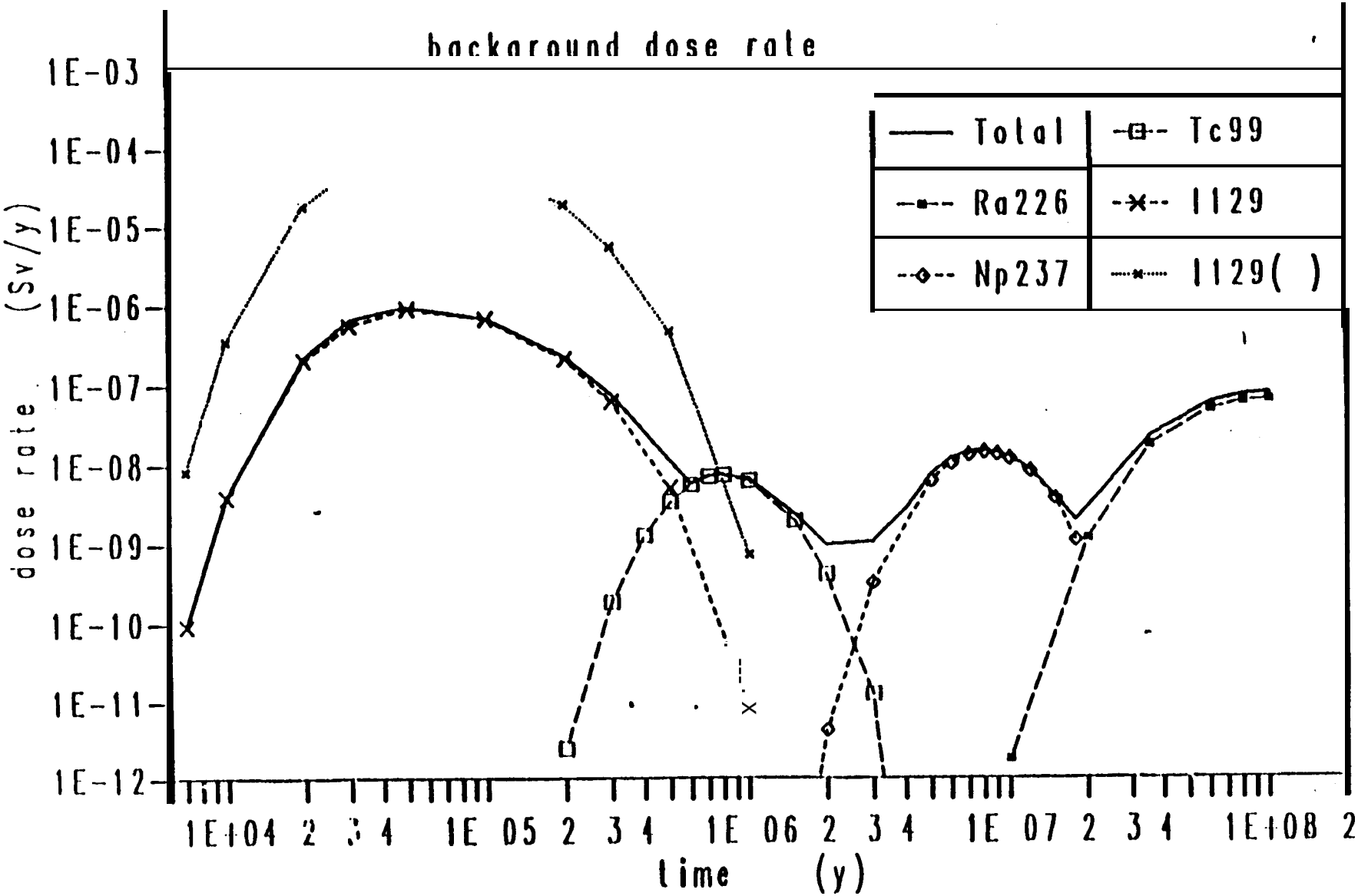

Fig. 2 : PACOMA STUDY
Calculated Total Individual Dose Rate for the Water Well Pathway of I-129, Np-237, Tc-99 and Ra-226
...x... Total Iodine Waste Inventory
---x--- Residual (1%) Iodine in MLW