Recovery Yield of Nuclides Required for Satisfactory Actinide Recycling†

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アクチニドリサイクルでのアクチニド核種回収の目標像を描くために、必要とされるアクチニド核種の回収率についての粗い評価を行った。アクチニドリサイクル型の燃料サイクルからのアクチニド核種の流出の傾向に関して簡単なモデルを用いて検討した。日本の将来において生産されることになるアクチニド核種の総累積毒性について評価し、望まれるアクチニド閉じこめの度合いを調べた。燃料サイクルから発生する様々な廃棄物を経由してのアクチニド核種のロスの割合を推測し、MOX 高速炉サイクルの条件下で、それぞれのパスからのロスが、ロスの総計に対して与える影響を調べた。以上の結論として、MOX 高速炉サイクルにおいて、アクチニド閉じこめを高い度合いで実現するためには、中レベル廃棄物を経由してのアクチニドのロスを防止するための思い切った技術改良が必要であることを指摘した。

In order to establish a visual target of actinide recovery in the actinide recycling, a rough evaluation on the required recovery yield for actinide nuclides was performed. The tendency of actinide's flow-out from actinide recycling fuel cycle was studied with a simple mathematical model. Desired magnitude of confinement of actinides was studied with analysis on the cumulative radiotoxicity going to be produced in the Japanese future period. By estimating loss factors for actinides at various waste streams of the fuel cycle, their impacts on the magnitude of total loss was studied under realistic condition of MOX-FBR system. It was concluded that, for satisfying severe goal of confinement of actinides in the actinide recycling system using MOX-FBR, drastic improvements will be required for the loss at intermediate level waste streams of reprocessing and fabrication.

1. Introduction

Recently, variety of different actinide recycle concepts are proposed and studied. This is because, currently ongoing nuclear power generation system still does not have a perfect solution on the substantial issue of radioactive waste generation, especially on the handling of man-made long-lasting nuclides. Geological disposal technology has been developed for this issue. Although, at present, this is probably the nearest concrete way to provide a minimum necessary answer to the waste issue, it does not mean that the current nuclear system is already perfect. Doubtlessly, a system which makes as little waste as possible will be a better answer, hence, there must be "more" research activities pursuing "less" waste generating system.

Our actinide recycling concept in the future FBR fuel cycle is one approach to this ultimate challenge [1]. In comparison with the emerging concepts like actinide burning by transmutation reactors or accelerators, our proposal is rather simple. Because of necessity on obtaining domestic "renewable" energy source, which is important especially for Japan, we maintain the necessity on the development of FBR system. Our proposal is to strengthen FBR system to confine all TRUs (plutonium and minor actinides) in its cycle to achieve minimum production of them through their recycling in the system. It has

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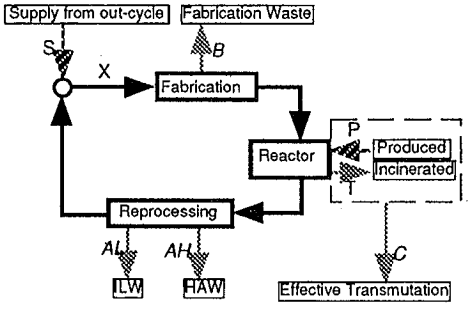
to be emphasized that this attitude does not necessarily pursue the reduction of exposure risk because the possibility of migration of actinides from deep geologic site is calculated to be very low [2]. Our motivation resides in a challenge against more substantial nature of nuclear system, which means the realization of "System producing as low man-made toxicity as possible".

In the Actinide Recycling FBR system, sufficient closure of the entire fuel cycle against flow-out of the actinide nuclides is strongly required. The magnitude of enclosure, namely magnitude of actinide recovery, has been a theme of discussion on transmutation technology. However, it has been likely to be limited to the recovery from high active liquid waste (HLW), which means loss of actinides through numbers of other possible paths has not been sufficiently discussed. In this article, in order to provide a basic guideline of actinide recovery in this type of fuel cycle, a very rough evaluation on the magnitude of recovery yield of actinides is conducted.

2. Evaluation on relative loss from recycling system

2.1 Method of evaluation

To visualize the efficiency of the recovery of actinides from waste streams, a very simple model of closed recycling system is proposed. A recycling system under equilibrium is assumed as **Figure 1**. In this model, amount X (per unit energy production) of actinide is fed to fabrication making some loss to waste with fraction B, and loaded to the FBR core. Fraction factors like B are defined as relative amount of loss compared with unit throughput (mass flow) at local points of the cycle, and are called Local Loss Factors. In the reactor core, amount T is transmuted while P is produced, giving effective transmutation factor C. The spent fuel is processed at reprocessing facility making some loss to HLW and other intermediate level waste (ILW) with fraction AH and AL, respectively. By supplementing the decreased amount with a supply from out-cycle (S from LWRs), the flow is kept constant to X. In this model, FBR cycle maintains its inventory constant while S is continuously accepted from LWRs at every unity of power production. The total loss of actinides to the wastes is represented by WL which is the summation of loss at fabrication, ILW and HLW. Because loss to three different paths are functions of local through-put at every point, total loss is a function of X and C.



X :Feed to Fabrication(≒Core Load)
S :Supply from out-cycle
P :Produced amount at Reactor
T :Incinerated amount at Reactor
WL :Total loss to all wastes
B :Local Loss factor at Fabrication
C :Effective transmutation factor
AL :Local Loss factor to ILW at Reprocessing
AH :Local Loss factor to HLW at Reprocessing
WR :Total Loss Factor

$$S = X\{1-(1- B)(1- C)(1- AH)(1- AL)\}$$
$$C = (T-P)/\{X(1 - B)\}$$
$$WL = X\ B+X(1- B)(1- C)\{AH+(1- AH)AL\}$$
$$WR = WL/(S+P)$$

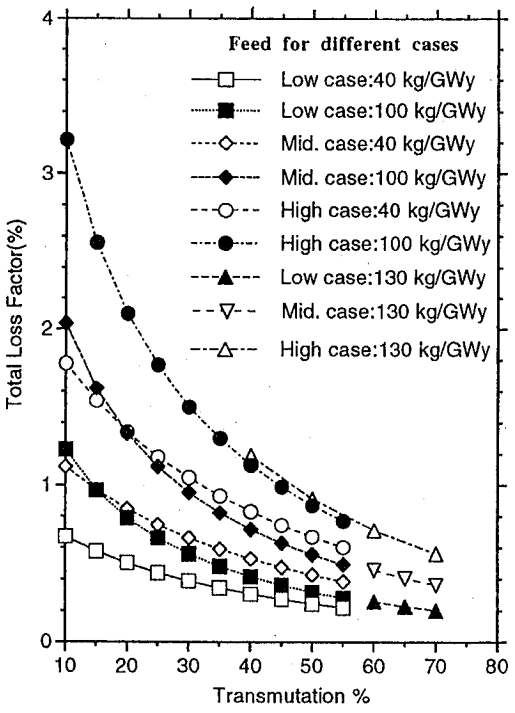
Fig. 1 Simple model of Recycling TRUs in the Cycle

In order to avoid the complexity in the evaluation of the resistance against the loss, "total loss factor" *WR* is defined as Figure 1. The denominator of "total loss factor" *WR* is S+P, and it gives the amount to be disposed if there is no recycling of actinides. In other words, under condition without actinide recycling, S as products of LWR having no acceptor, and P as a product of FBR operation have to be directly disposed as wastes. The loss to the wastes (WL) in the actinide recycling has to be far below the S+P. Therefore, *WR* gives relative magnitude of the loss in actinide recycling to the amount directly disposed by no actinide recycling case.

2.2 Results

According to the equations listed in Figure 1, the dependence of total loss factor on various combination of local loss factors, feed rate and transmutation rate was calculated as **Figure 2**. Apparently from Figure 2, high inventory system (much feed) with low transmutation rate makes rather large relative loss to the waste. In the case of high loss (loss at HLW = 0.2%, loss at ILW = 0.3% and loss at Fabrication = 0.3%), combination of low transmutation rate and high inventory produces more than 3 % of direct disposal, which is a far poor result to the primary purpose of actinide recycling.

On the contrary, in the region of high transmutation rate, dependence on the inventory and local loss factor is much smaller, but this high transmutation rate is only possible by hypothetical transmutation reactors. We tried to find the position of actinide recycling using MOX-FBR, by using an example of calculational results reported in a literature. According to the study done by M. Kiyota et al., 1600 MWth MOX-FBR produces 30 kg of minor actinides per cycle (15 months in 3 batches refueling) with no minor actinide loading [3]. In the case of heterogeneous loading of Am & Cm with homogeneous loading of Np in drivers, 205 kg charge at BOC (Beginning of Cycle) and 161 kg discharge at EOC (End of Cycle) are obtained per cycle. This case corresponds to ca. 21% transmutation with 85 kg feed/GWy [3], which suggests that quite low loss will be needed to attain smaller total loss factor than 1%. As can be seen in the following paragraphs, there are some cases that target level of recovery becomes less than 0.1%, suggesting that local loss factors at fabrication and reprocessing should be less than the lowest case of this



Local Loss Factor settings

Low Case	:HLW=0.1%,ILW=0.1%,Fab=0.1%
Mid Case	:HLW=0.1%,ILW=0.2%,Fab=0.2%
High Case	:HLW=0.2%,ILW=0.3%,Fab=0.3%

Fig. 2 Dependence of total loss factor on transmutation efficiency and feed rate

figure.

3. Target level of the cumulative toxicity leak-out

In addition to the main purpose of actinide recycling i. e. "reduction of the cumulative production of man-made actinides", we have another important objective that is "confinement of radiotoxicity into the cycle". This means that disposal of actinides has to be lower than a certain target level which is explicitly acceptable. A simple calculation study was done to clarify this target level.

3.1 Method of calculation

In order to provide an understandable measure of "satisfactory magnitude of actinide disposal", we evaluated cumulative production of actinides in the Japanese limited future. If the cumulative production of radiotoxicity of actinides during a certain period in Japan exceeds some commonly understandable or tolerable range, its disposal might be better to be postponed during this period, in order to see any better ultimate solution for its handling. The reason why we use cumulative mass over a certain period is because it provides a visual feeling on how much toxicity we are going to produce and dispose.

ORIGEN-II was used to obtain radiotoxicity production per unit power generation of three reactor types PWR, MOX loaded PWR (1/3-loading) and MOX-FBR. The nuclear electricity capacity in Japanese future was assumed to increase achieving ca. 150 GW at 2100. In this scenario, FBR starts to be employed from 2030 succeeding life-ended LWRs. As an index for radiotoxicity of nuclides, multiple of ALI (Annual Limit on Intake) was used. The assumed fuel conditions are summarized in **Table 1**. Period 2030 -2100 was selected as an understandable objective period for integral evaluation. To make the analysis simpler, it was assumed that cumulative toxicity throughout this period is produced at one time at 2100, hence decay of short-lived nuclides during this period is neglected.

Table 1 Calculation conditions for the cumulative production of toxicity

	LWR	Pu-LWR	FBR
Electric Power (GW)	1	1	1
Generation Efficiency(%)	34	34	40
Load Factor(%)	90.00	90.00	90.00
Burnup(MWd/t)	4.5	4.1	14.8
Annual fuel charge (tHM)	21.5	23.6	5.5
Specific Power (MW/tHM)	38.0	36.7	67.7
U-enrichment(%)	4.50		
Pu-enrichment(%)		2.97	18.75

3.2 Results

Cumulative generation of spent fuels from LWR, MOX-LWR and MOX-FBRs throughout this period is 83499 HMt, 9214 HMt and 34940 HMt, respectively. Produced cumulative radiotoxicity is shown in Figure 3. Data of Pu and U in **Figure 3** represent 0.5% of their total production, while other FPs and minor actinides are of 100%. Figure 3 is given as relative comparison to the toxicity of cumulative natural uranium and enriched uranium which are needed to realize this nuclear scenario. The cumulative amount of natural uranium and enriched uranium are 711065 t and 77630 t, respectively. The potential toxicity of these are the ones under sufficient radiochemical equilibrium, hence they include daughter's toxicity.

The comparison of cumulative man-made toxicity (FP and actinides) with the cumula-

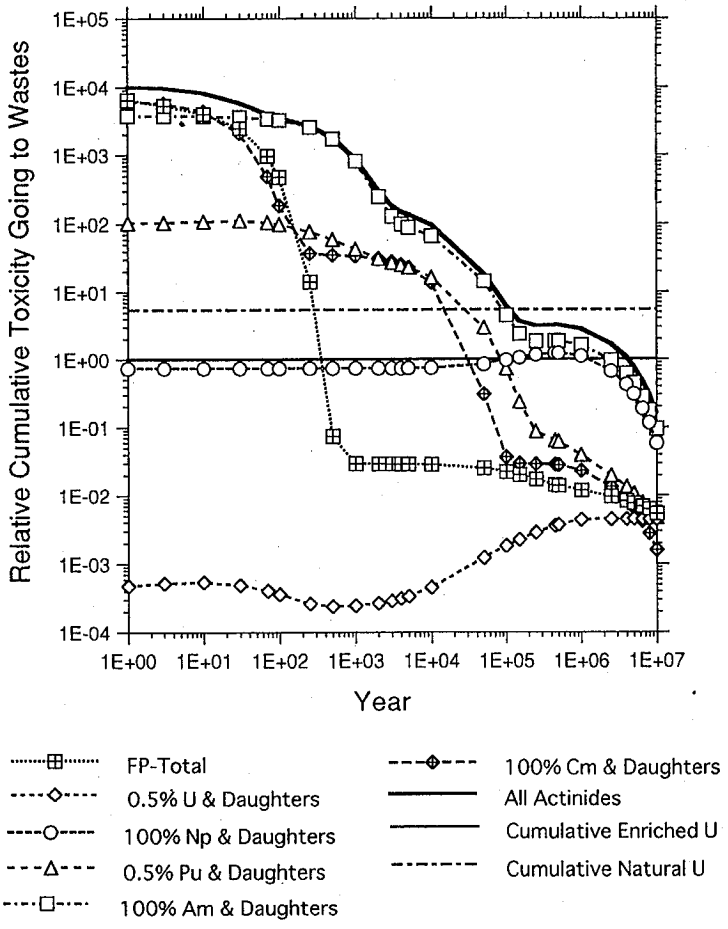


Fig. 3 Comulative toxicity going to wastes before year 2100

tive natural and enriched uranium provides how the produced toxicity is large or small compared with the raw materials that were used for getting energy gain. We believe that this comparison gives a qualitative reference level for the acceptance of the produced toxicity. Apparently, FP is quite high at the time of discharge, but it rapidly decreases to "less than uranium" before a thousand years pass. This means FPs have a feature somewhat tolerable if we can provide a certain reliable containment and isolation over ca. 1000 years. More over, even after 1000 years with consideration on long-lived FPs (^{99}Tc, ^{129}I, ^{135}Cs etc), it is far below the raw material level. This means that the disposal of FPs at present has less risk on possible future regret, therefore there will be no worry in speeding up the disposal of FPs.

On the other hand, the cumulative toxicity level of actinides, even with quite high recovery of Pu, maintains its level far higher than the uranium over a hundred thousand years. This means that these nuclides have a feature which requires more careful consideration on their handling compared with FPs. This is because we propose actinide recycling with FBR system. By confining actinides into the

cycle, we can delay the decision on their handling and can keep reducing its cumulative generation till the end of nuclear age comes [1].

If we postpone to make a decision on starting disposal of actinides to the environment, they must be well confined in the cycle. The tolerable level of the flow-out of actinides from confinement cycle has to be consistent with the above logic on FPs. Therefore, comparison with the level of uranium or FPs will be a qualitative target of the actinide confinement.

Figure 4 gives the recovery effect of actinides with different recovery yield on different nuclides. By enhancing recovery yield for Pu, Am, Cm and Np, namely lowering their loss factors to 0.01%, 0.05%, 0.1% and to 2% respectively, total toxicity of actinides becomes lower than the level of enriched uranium as is already achieved by FPs. This suggests that these levels of recovery are satisfactory for the confinement of the cycle. If the level of natural uranium is chosen as the reference, the desired recovery goal becomes much mitigated than this.

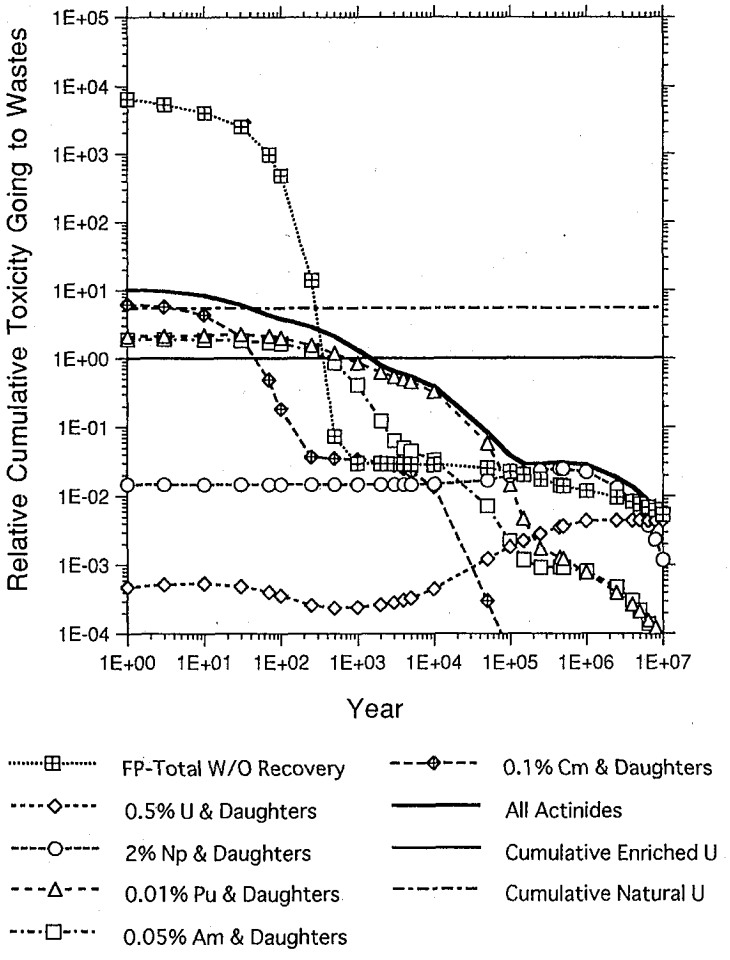


Fig. 4 Effect of actinide recovery on the total toxicity to wastes

However, in Japanese case, because all natural uranium is bought abroad, enriched uranium which has once been brought into Japan will be more visually understandable reference. It has to be emphasized that this reference level determined by cumulative enriched uranium is rather severe goal. However, it is still proper because finding an ultimate goal of actinide recycling will enable us to find a practical goal too. In addition, it should be noted that this reference never gives an acceptable level of safety, but only gives an understandable magnitude of potential toxicity production.

4. Analysis on the possible flow-out via various waste streams

4.1 Local loss factors of actinides

Paths of actinide's flow-out from the fuel cycle are listed in **Figure 5**. Local loss factor *B* in the above model covers three fabrication wastes and *AL* covers all intermediate level wastes of reprocessing. What is needed for analyzing necessary confinement level of actinide recycle is to evaluate most probable level of loss factors at every path of wastes. However, actually it is impossible to determine realistic loss factor of minor actinides because there is little data on the mechanism and magnitude of loss under plant conditions. Therefore, we tried to evaluate them very roughly using already reported data. For the data of minor actinides, the experienced loss in the current Pu utilization can be used as first-order approximation under condition that similar type of processing will be used for the minor actinide treatment.

Recovery yield data for actinides by newly developed extraction process is reported in comprehensive manner as can be seen in **Table 2** [4-6]. There are four types extraction

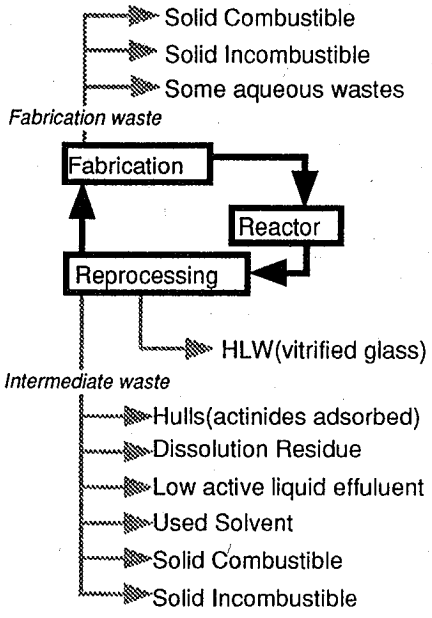


Fig. 5 Major possible paths of actinide loss to wastes

process whose recovery yields are actually observed by small scaled hot experiments. Except for the case in which valence control of Np was failed, the loss factors are satisfactorily low (<0.01%) [4]. This gives us a hope that actual local loss factor *AH* can be as low as 0.01% if its design is carefully performed. We can easily guess that paths which dominate Pu flow-out in the current reprocessing (Chop & Leach plus PUREX) will also dominate minor actinide's flow-out if same type of processing is used. For instance, if fuels containing minor actinides are dissolved in nitric acid and same kind of chemical adsorption occurs onto the surface of hulls, it can be estimated as large as or less than the case of Pu. This is simply because adsorption of tetravalent Pu cation to inorganic surface is expected to be much stronger than the case of tripositive TRU cations. In the past report about the Pu contamination

Table 2 Reported distribution of actinides to raffinate stream and solrent

Distribution to Raffinate Unit : %

	TRPO[1]	CMPO[1]	DIDPA[1]	DIAMIDE[3]	Representative[5]
U	0.019	0.001	0.005	-	0.001
Pu	0.200	0.025	0.667	-	0.025
Np	0.024	0.003	-	-	0.003
Am	0.005 2)	0.003 2)	0.025 2)	0.091	0.003
Cm	0.005 2)	0.003 2)	0.025 2)	0.125	0.003

Residue in Stripped Solvent Unit : %

U	0.100	0.100[4]	99.300	-	0.100
Pu	0.100	-	8.100	-	0.100
Np	0.500	0.100[4]	11.600	-	0.100
Am	0.000	0.400[4]	-	-	0.400
Cm	-	0.600[4]	-	-	0.600

1)Study by J.P.Glatz et al. (4)
2)Because DF observed is limitted by detection limit, observed DF for Nd is used as a very similarly behaving nuclide.
3)Study by C.Madic et al. (5)
4)Study by R.P.Bush et al. (6)
5)TRUEX(CMPO) is assumed.

on the rinsed hulls of LWR fuel after dissolution, it is reported that as much as 0.4% remains on the hulls while Cm/Pu ratio is about 1/2.7 of the initial spent fuel [7].

Another important path of actinide flow-out is the dissolution residue as was experienced in the case of conventional Pu recycling. Although undissolved fraction of Pu is thought to be less than 0.15% of the initial [9], there is no information on the possibility of similar phenomena on Am, Cm and Np. Remainder on the used solvent will probably be one of the critical path of flow-out. It is reported that the stripping of actinides from extractants is rather poor [4], however, there is no data about the efficiency of solvent scrubbing and its possible enhancement.

Probably, contaminated solid wastes from fuel fabrication facility will be the biggest path of actinide flow-out. It is natural to estimate that same order or more will be generated if same type of fabrication process and equipment is used for the fabrication of minor actinide fuels. However, actual flow-out data of MOX fabrication plant is very little. In the report about French MELOX plant [10], as high as one thousandth of the Pu through-put is anticipated. There will be much discussion on this, but the depression of the actinide distribution to the wastes at fabrication will certainly govern the total loss factor. Above rough estimation on the different waste streams are summarized in **Table 3.**

4.2 Recovery target of actinides in the MOX-FBR cycle

By using example results of case study on

Table 3 Roughly estimated Local Loss Factors to various waste streams

Unit : %

	Waste Streams	U	Pu	Am, Cm	Np
Reprocessing	PUREX Raffinate	0.040 1)	0.080 1)	100	100
	MA Recovery Raffinate	0.001	0.025	0.003	0.003
	HLW after MA Recovery	0.000 2)	0.000 2)	0.003 2)	0.003 2)
	Hulls	*	0.400 3)	0.148 3)	*
	Dissolver Residue	0.100 4)	0.150 4)	*	*
	Liquid ILW	* 5)	* 5)	**	**
	Used Solvent	0.000	0.000	0.400 6)	0.100 6)
	Solid Combustible	unknown	unknown	**	**
	Solid Incombustible	unknown	unknown	**	**
Fabrication	Solid Conbustible	0.100 7)	0.100 7)	0.100 8)	0.100 8)
	Solid Incombustible				

* no estimation
** depends on the process to be adapted

1)Design of UP-3 first cycle. Actually, much better performance was observed.(8)
2)Product of above two.
3)Average value of LWR fuel. Subject to differ in FBR cladding case.(7)
4)Design of FBR reprocessing test facility.(9)
5)No definite number, but generally very low.
6)Without solvent washing treatment. Should be much improved.(4)
7)Target of MELOX plant.(10)
8)Assumed to be same to Pu if conventional fabrication technique is employed.

actinide recycling in MOX-FBR, possible range of recovery (local loss factors) satisfying the above goal was analyzed. Applying refueling characteristics of Np and Am (**Table 4**) to the above simple mathematical model, dependence of total loss factor on local loss factors in the MOX system is calculated as **Figure 6** and **7**. Refueling conditions reported are listed in Table 4 [11]. Because transmutation factor at MOX core is restricted to ca. 30%, total loss strongly depends on the local loss factors.

Because the distribution of Am and Np to the high active raffinate is estimated very low, total loss of them is almost governed by loss at ILW and fabrication. For Np whose desired confinement goal is not severe (2%), recovery with local loss factor around 0.5% at ILW and fabrication will be sufficiently enough. On the contrary for Am, to achieve its quite high confinement goal (hopefully 0.05%), very high recovery at fabrication and ILW is needed. Both loss at fabrication and ILW have to be in the range less than 0.1% which is far beyond the experienced

Table 4 Characteristics of Minor Actinide Loaded MOX FBR[11]

Thermal Power	2520 MWth
Cycle Length	15 months(3 batches)
Fuel Burnup	90000 MWD/t
Minor Actinide loading	Homogeneous

Refueling Pattern	Np	Am
Core Inventory(kg BOC)	719	710
Core Inventory(kg EOC)	590	624
Charge(kg/cycle)	289	268
Discharge(kg/cycle)	160	182
Consumption(kg/cycle)	129	86
Transmutation Rate(%)	44.64	32.09

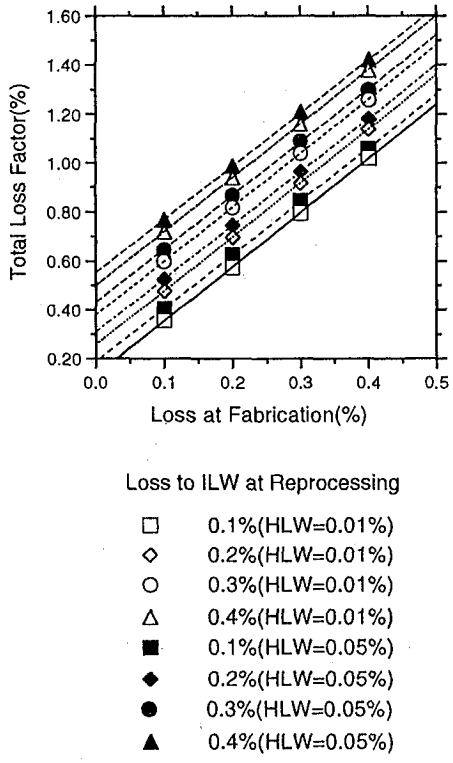


Fig. 6 Estimated total loss factor of Np in the MOX-FBR system

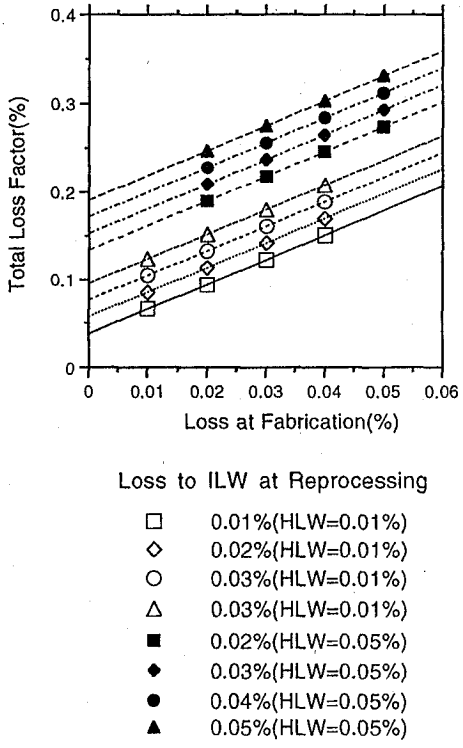


Fig. 7 Estimated total loss factor of Am in the MOX-FBR system

level in the current Pu recycling.

5. Discussion and conclusion

Through the above study on the recovery goal needed for satisfactory actinide recycling, a rough outline of the recovery goal covering entire cycle became clear. In order to satisfy the requests to keep the feature of wastes from actinide recycle to the range of used uranium, quite high recovery goal is needed for Pu and Am (total loss factor 0.01% and 0.05% respectively). Under a realistic condition of MOX-FBR system, in order to achieve this goal, quite high local recovery is also needed at fabrication and reprocessing. However, if currently experienced magnitude of loss at fabrication and ILW is applied to the actinide recycle, the goal can not be satisfied. This means there must be some drastic improvements in the waste generation mechanism both in the fabrication and reprocessing to realize satisfactory actinide recycling.

Through the above discussion, we can raise the following proposals to the technologies to be applied to actinide recycling. Firstly, both fabrication and reprocessing have to be so simple that they generates as little amount of wastes as possible. Secondly, standing on the fact that waste generation can not be zero, there must be a strong function to recover nuclides from once produced wastes. Figure

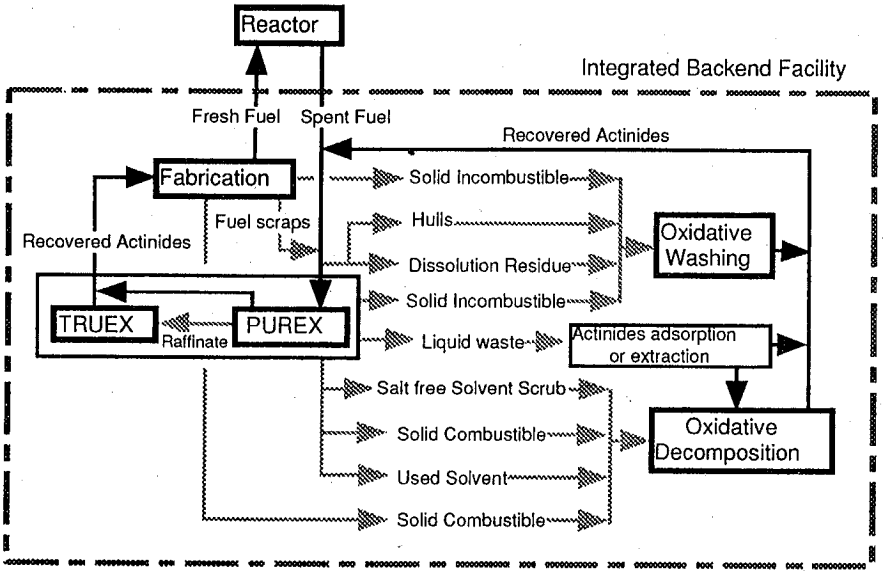


Fig. 8 Concept of actinide closing facility

8 provides a conceptual scheme of future integrated backend facility in which all waste streams are treated to recover actinides for feeding them back to the process streams. If decontamination factor of 20 to 30 at the integrated waste recovery process of **Figure** 8 is attained, the above-discussed ultimate goal can be achieved. Utilization of recently developed technologies like oxidative decomposition system using Ag (II) may hopefully meets the philosophy of this scheme [12].

6. References

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