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RADIOACTIVE CHARACTERISTICS OF SPENT FUELS AND REPROCESSING PRODUCTS IN THORIUM FUELED ALTERNATIVE CYCLES

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Radioactive characteristics of spent fuels and reprocessing products in thorium fueled alternative cycles*

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In order to provide one fundamental material for the evaluation of Th cycle, compositions of the spent fuels were calculated with the ORIGEN code on following fuel cycles : (1) PWR fueled with Th- enriched U, (2) PWR fueled with Th-denatured U, (3) CANDU fueled with Th-enriched U and (4) HTGR fueled with Th-enriched U. Using these data, product specifications on radioactivity for their reprocessing were calculated, based on a criterion that radioactivities due to foreign elements do not exceed those inherent in nuclear fuel elements,due to ^{232}U in bred U or ^{228}Th in recovered Th, respectively.

Conclusions are as the following :

(1) Because of very high contents of ^{232}U and ^{228}Th in the Th cycle fuels from water moderated reactors, especially from PWR, required decontamination factors for their reprocessing will be smaller by a factor of 10^3 to 10^4, compared with those from U-Pu fueled LWR cycle.

(2) These less stringent product specifications on the radioactivity of bred U and recovered Th will justify introduction of some low decontaminating process, with additional advantage of increased proliferation resistance.

(3) Decontamination factors required for HTGR fuel will be 10 to 30 times higher than for the other fuels, because of less ^{232}U and ^{228}Th generation, and higher burn-up in the fuel.

Keywords : Thorium cycle, Reprocessing, Uranium 232, Thorium 228, Product Specification, Spent Fuel, Radioactivities, PWR, HTGR, CANDU, Comparative Evaluation.

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* The present study was made in the author's stay at University of California, Berkeley, in 1977, by the guidance of Professor Thomas H. Pigford.

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代替トリウムサイクルにおける使用済燃料と再処理プロダクトの放射能特性*

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Thサイクル評価に資するため，下記燃料サイクルの使用済燃料組成を ORIGEN コードにより計算した。

(1) 高濃縮ウラン富化トリウム装荷PWR
(2) 変性ウラン・トリウム装荷 PWR
(3) 高濃縮ウラン富化トリウム装荷 CANDU
(4) 高濃縮ウラン富化トリウム装荷 HTGR

さらにこの結果を用い，外部不純物による放射能が，^{232} U，^{228} Th に起因する増殖ウラン，回収トリウムの内在放射能を上回らないとする仮定に基づき，上記Th 燃料再処理における不純物の放射能製品仕様を計算した。

主な結論は下記の通り。

(1) 水炉系とくにPWR からの燃料では^{232} U，^{228} Th 含有量が大きいため，これらの燃料を再処理する場合，所要の除染係数は，U－Pu サイクルに比べ 10^3～10^4程度小さくてよい。
(2) このU，Th の放射能製品仕様の緩和は，その再処理において低除染法の適用が許容され核拡散防止上も利点となる。
(3) 高温ガス炉の場合，^{232} Uの生成量が少く，燃焼度も高いため，他のTh 炉型に比べ，その所要除染係数は10～30倍高くなる。

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1. Introduction

Since the U.S. President's statements of 1977, increased emphasis of thorium utilization is suggested leading to renewed interests as an alternative fuel cycle with proliferation resistance. Although many studies[1] on the cycle have been reported and more presently[2], recent concern is overall performances of the cycle with the view of resource utilization. Considering that effective use of the resourse is expected by its recycling, evaluation of the back-end will be indispensable.

The principle of the Thorex flowsheet and the technological possibility have been proven in the pilot-scale at ORNL already in 1950s[3]. Many analyses and conceptual design studies[4] were made to apply the flowsheet to the reprocessing of thorium fuels from those reactors classifically significant. Pilot scale development of a more advanced process of "Acid-Thorex"[5] was alos carried out by General Atomic Co. along with thorium utilization program in HTGR[6]. Based on these back-ground, this technology is considered not to have large problems for recovering nuclear material from the spent thorium fuels[7]. Still, it should be noted that the present requirement for the technology will be different from those in the past, in proliferation resistance, minimum environmental release and minimum waste generation. Considering the renewed incentives for utilizing the resource, it will be important to review the technology through a detailed analysis on the fuel from those reactors which are already or almost commercialized. The purpose of the present study was to provide one material for its evaluation, i.e. characterizing radioactivities of the fuel and showing radioactivity product specifications required for the reprocessing.

Radioactivities of spent fuel were calculated with the ORIGEN code for the following fuel cycles in equilibrium ;

(1) PWR fueled with thorium and 93 % enriched uranium
(2) PWR fueled with thorium and denatured uranium
(3) CANDU fueled with thorium and 93 % enriched uranium
(4) HTGR fuelded with thorium and 93 % enriched uranium

They are selected from those fuel cycles which could be implemented in the reactors which are in the stage of commercial application. One significant features of the fuel is the intense radioactivity associated with ^{232}U or ^{228}Th, of which major generation pathway is

(n, 2n) reaction in ^{232}Th. Isotopic content of ^{232}U in bred U or of ^{228}Th in recovered Th will be different in the above fuel cycles, due to the difference in their neutron energy spectrum and in initial contents of Th and U in the charged fuels. This isotopic content will substantially influence the kind of technologies adaptable to the back-end of Th cycle.

With respects to radioactivity specifications for the reprocessing, which is related with recycling strategy of the fuel, the criterion will be different from U-Pu fueled LWR's because of the unique build-up and decay behavior of the activity. The criterion formerly accepted for U-Pu LWR cycle is to reduce the activity sufficiently for the direct refabrication and to prevent accumulation of hazardous actinides in the isotope enrichment process, although current proliferation issues may affect this criterion also.

In the case of thorium cycle fuel, however, it has been generally accepted that the most reasonable recycling of the fuel is to refabricate bred uranium quickly after reprocessing and, for recovered thorium, to store it for sufficient time to refabricate directly. Also, it was qualitatively recognized that less stringent product specifications would be possible in the reprocessing of Th cycle fuels. Nevertheless, the R & D carried out so far seems not to be well directed, not inconsistence with the above recognition, as seen in KILOROD project[8] aiming at semi-remote refabrication or in the Thorex flowsheet with three cycles[9] to obtain highly decontaminated products in the reprocessing of HTGR fuel. These inconsistent approaches done show that, only by the economical consideration, it is difficult to make any clear-cut of recycling strategy in this fuel cycle. Consideration of proliferation issues may lead this undefinite strategy positively toward the selection of low decontamination process.

Some conceptual design studies were made on low decontamination process for HTGR fuel by analyzing the required decontamination factors of fission products[10].

The present study was extended to the specification of U, Pu and Th as impurities in bred uranium and recovered thorium, so to show more detailed specifications on the product radioactivity. A comparison of the product specifications was made for the selected Th cycles to show the effect of reactor type on the recycling strategy. Some discussion was made on the proliferation resistance of the cycles, based on the results obtained.

2. Calculation of radioactivities of the discharged fuels

2.1 Reference fuel cycle

Fuel cycle performances of selected reactors are based on the Pigford and Yang's study[11]. Table 1 and Fig. 1 summerizes major reactor performances assued and material balance for nearly equilibrium cycle, which are used as inputs to the ORIGEN code in calculating compositions of the spent fuels. The model PWR is originally based on that described by Combustion Engineering[12]. A little higher burn-up assumed there, considers lower fuel density of ThO_2 than UO_2. For the calculation of the CANDU cycle, a higher value of 16 MWd/kg is used according to Till and Chang's study[13]. In some reactors, bred uranium can be recycled segregatedly from the make-up fuel for its more effective utilization. For simplifying the present discussion, however, it was assumed that both discharged fuels were mixed and reprocessed together.

2.2 Application of the ORIGEN code

Since ORIGEN nuclear data library does not include data for Th fueled PWR and CANDU, preparation of them are required, especially on nuclides with high contents to consider the different self-shielding effect on their resonance integrals, from U fueled PWR's. Nordeheim method[14] is applied to the calculation of major actinides, i.e. ^{232}Th, ^{233}Pa, ^{233}U, ^{235}U, ^{238}U and ^{239}Pu. For the other nuclides with lower compositions, data originally included for U fueled PWR were employed, because of possibly lower self-shielding effect on them.

With respects to flux parameters such as RES and FAST that must be also input in ORIGEN calculation, data by a burn up calculation[15] with multi-group code were applied for PWR, while a method developed by T. H. Pigford[16] were used for CANDU.

Table 2 shows an example of nuclear data used and a comparison of calculated compositions. Since their agreement among ORIGEN and the burn-up calculation remains within 20 %, it should be noted that the following discussions should be limited to the accuracy of the above order.

2.3 Radioactive characteristics of the spent fuels

Table 1 and table 3 summerize calculated results on compositions of the spent fuels discharged from the selected thorium cycles, after one year cooling.

Principal characteristics of their radioactivities are ;

1) more production, by a factor of ten, of ^{232}U and ^{228}Th in PWR than in HTGR, because of more fast neutron above the (n, 2n) threshhold, due to greater fuel lumping and closed packed lattice in PWR core,
2) more production of Pu in denatured U fueled PWR, because of higher ^{238}U content in the charged fuel, compared with the other thorium cycles topped with enriched U,
3) smaller Pu production and higher isotopic content of ^{238}Pu in the Th cycles fueled with enriched uranium, especially in CANDU, which might discourage economical incentives to recover plutonium, although the safeguard issues will not justify to throwaway it as high level wastes,
4) higher contents of fission products and actinides in HTGR, because of the higher burn-up assumed,

and

5) great ammounts of graphite waste from HTGR, which are qualitatively quite different from those water moderated reactors.

2.4 Radioactive characteristics of product uranium and thorium

Table 4 summerizes isotopic compositions of U and Th in the selected thorium cycles. Generally, predominant nuclides that affect the total radioactivity of the product are ^{232}U and ^{237}U in bred uranium, and ^{228}Th and ^{234}Th in recovered thorium, respectively. However, activities of ^{237}U and ^{234}Th become ignorable in the longer cooling time than a half year, because of their short half lives, which are 6.75 days for ^{237}U and 24.1 days for ^{234}Th. Therefore, the total radioactivities of U and of Th can be represented only by the isotopic content of ^{232}U and ^{228}Th, respectively.

Decay chain of ^{232}U and ^{228}Th are shown in Fig. 2[17]. ^{232}U makes α-decay to ^{228}Th which also decays emmitting α-particle. Since daughters from ^{228}Th are all short-lived, they reach secular equilibrium with the parent within a few days. Furthermore, most of these daughters,

especially ^{208}Tl and ^{212}Bi, release γ radiation with high energy, which presents one of the most significant effects on the selection of process technologies for Th cycle back-end.

Build-up rates of their gross activities are calculated in detail[18]. Considering penetrating power of the radiations, activity growths of ^{232}U and ^{228}Th were recalculated in the units of γ-MeV·Ci and γ-Ci, as a function of time after separation, t_F. Fig. 3 shows the radioactivity of unit kg U and Th with isotopic content of one ppm ^{232}U and ^{228}Th, respectively. In the time scale of day, the radioactivity growth of γ-MeV·Ci from unit Ci ^{232}U and ^{228}Th, $a_\ell(t_F)$, is given approximately as following[17] ;

$$a_\ell(t_F) = 0.00063\ B_{08,\ell}(t_F) + 1.36\ B_{224,\ell}(t_F)$$

where ℓ = index for nuclides, ℓ = 08 and 22 indicate ^{228}Th and ^{232}U, respectively.

$$B_{08,22}(t_F) = \left(\frac{\lambda_{08}}{\lambda_{08}-\lambda_{22}}\right)\left(e^{-\lambda_{22}\cdot t_F} - e^{-\lambda_{08}\cdot t_F}\right)$$

$$B_{224,22}(t_F) = \left(\frac{\lambda_{224}\cdot\lambda_{08}}{\lambda_{224}-\lambda_{08}}\right)\left[\frac{1-e^{-\lambda_{08}\cdot t_F}}{\lambda_{08}} - \frac{1-e^{-\lambda_{224}\cdot t_F}}{\lambda_{224}}\right]$$

$$B_{08,08}(t_F) = e^{-\lambda_{08}\cdot t_F}$$

$$B_{224,08}(t_F) = \left(\frac{\lambda_{224}}{\lambda_{224}-\lambda_{08}}\right)\left(e^{-\lambda_{08}\cdot t_F} - e^{-\lambda_{224}\cdot t_F}\right)$$

λ_{08}, λ_{22}, λ_{224} = decay constants of ^{228}Th, ^{232}U and ^{224}Ra, respectively. [day^{-1}]

From the previous discussion, total γ-MeV·Ci radioactivities of product uranium and thorium, $[A(t_F)]\ell_o$ (ℓ_o = U or Th), can be approximated as following ;

$$[A(t_F)]\ell_o = \alpha_\ell\ a_\ell(t_F)\ Y_\ell \qquad (2)$$

where α_ℓ = unit conversion factor of nuclide ℓ, [Ci-ℓ/kg-ℓ]

y_ℓ = isotopic content of key nuclide ℓ ; ^{232}U for U and ^{228}Th

for Th [wt. fraction]

3. Product specifications on the radioactivity in reprocessing Th cycle fuels

3.1 Definition of the criteria to give the product specification

Basic criterion used to define the allowable concentration of an impurity element is to compare the radioactivity or the hazard index with that inherent in the fuel element. An activity unit in γ-MeV·Ci was used for determining the specification of impurity Th, U and FPs. Concerning Pu, hazard index was employed for the comparison, considering required confinement for relevant facilities and equipments. These criteria are summerized as following.

(1) Criteria for Th, U and fission products

Total inherent activities in fuel element ℓ_o (ℓ_o = Th or U), $[A(t)]_{\ell_o}$, does not exceed sum of activities due to impurity element or isotope ℓ_F(ℓ_F = Th, U or FP i), $[A(t)]_{\ell_F}$, i.e.

$$[A(t)]_{\ell_o} \geq [A(t)]_{\ell_F} \tag{3}$$

with

$$[A(t)]_{\ell_o} = \alpha_{\ell} \cdot y_{\ell} (\Sigma_j B_{j,\ell}(t) \cdot \bar{E}_j) \quad (\ell_o = \text{U or Th})$$

$$[A(t)]_{\ell_F} = \alpha_{\ell} \cdot y_{\ell} \cdot x_{\ell_F} (\Sigma_j B_{j,\ell}(t) \cdot \bar{E}_j) \quad (\ell_F = \text{Th or U})$$

with respects to FPs,

$$[A(t)]_{FP-i} = Z_i \ (\Sigma_j B_{j,i}(t) \cdot \bar{E}_i)$$

where

$B_{j,\ell}(t)$, $B_{j,i}(t)$ = built-up or decayed activity of nuclide j at time t per unit initial activity of parent nuclide ℓ or i, [Ci-j/Ci-ℓ or -i]

$\bar{E}_j$ = average γ energy emmitted from nuclide j, [MeV]

y_{ℓ} = initial isotopic content of key nuclide ℓ in elemnt

for Th [wt. fraction]

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with

$$[A(t)]_{\ell_o} = \alpha_\ell \cdot y_\ell \left(\sum_j B_{j,\ell}(t) \cdot \bar{E}_j\right) \quad (\ell_o = \text{U or Th})$$

$$[A(t)]_{\ell_F} = \alpha_\ell \cdot y_\ell \cdot x_{\ell_F} \left(\sum_j B_{j,\ell}(t) \cdot \bar{E}_j\right) \quad (\ell_F = \text{Th or U})$$

with respects to FPs,

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where

$B_{j,\ell}(t)$, $B_{j,i}(t)$ = built-up or decayed activity of nuclide j at time t per unit initial activity of parent nuclide ℓ or i, [Ci-j/Ci-ℓ or -i]

$\bar{E}_j$ = average γ energy emmitted from nuclide j, [MeV]

y_ℓ = initial isotopic content of key nuclide ℓ in elemnt

ℓ_F, [Wt. fraction]

x_{ℓ_F} = impurity composition of element ℓ_F, [Wt. fraction]

$Z_i(t)$ = activity composition of fission product i at time t per unit fuel element, [Ci-i/kg-fuel element]

(2) Criteria for Pu

Total hazard index inherent in fuel element ℓ_o $[H(t)]_{\ell_o}$, does not exceed that due to impurity Pu, $[H(t)]_{\ell_F}$;

$$[H(t)]_{\ell_o} \geq [H(t)]_{\ell_F} \quad (\ell_o = \text{U or Th}, \ \ell_F = \text{Pu}) \tag{4}$$

with

$$[H(t)]_{\ell_o} = \sum_j RHI_{j,\ell_o}(t)$$

$$[H(t)]_{\ell_F} = \chi_{\ell_F}\left(\sum_j RHI_{j,\ell_F}(t)\right)$$

$$RHI_j(t) = \frac{s_j \cdot y_j(t)}{(MPC)_{a,j}}$$

where

$y_j(t)$ = isotopic composition of nuclide j at time t, [wt. fraction]

s_j = specific activity of nuclide j, [Ci-j/kg-j]

$[MPC]_{a,j}$ = maximum permissible concentration of nuclide j in air, [Ci-j/cm^3]

3.2 Assumption to define product specification

(1) Cooling time

Cooling time t_c of one year is assumed a priori in the present study. An evaluation is made for the following criteria.

a. Decay of ^{131}I ; to reduce its activity to the level of ^{129}I considering the difference in their $(MPC)_a$

b. Decay of ^{233}Pa ; to reduce the potential ^{233}U loss less than 0.1 % of total potential fissile ^{233}U

Calculated results in Table 5 indicates that the decay of ^{131}I is the critical factor even in the thorium cycles studied. Considering current change in reprocessing incentives, different criteria could be

proposed, especially from the view of waste management. T. H. Pigford found that cooling time of 3.1 year minimizes ^{226}Ra activity, which could be one of the most significant radionuclide after 10^6 to 10^7 years[19].

(2) Storage time of recovered thorium

If recovered thorium is stored until the activity of ^{228}Th is no more greater than that of natural thorium by a factor ψ, the storage time T_s is given as following ;

$$T_s = \left(\frac{1}{\lambda_{08}}\right) \ell n \left[\left(\frac{y_{08}}{\psi}\right)\left(\frac{\lambda_{08}}{\lambda_{02}}\right)\right] \quad (5)$$

where

λ_{02} = decay constant of ^{232}Th

In this study, two cases are assumed (1) $\psi = 1$ as a lower limit and (2) $\psi = 5$ as an upper limit for direct refabrication[17]. Calculated T_s for the selected thorium cycles are included in Table 8.

(3) Refabrication time of bred uranium

Since the activity due to ^{232}U increases with time, the required specification on the radioactivity of bred U will be a function of time t_F before which the uranium is refabricated after the separation. In this study, two cases are assumed for t_F i.e. (1) one day and (2) ten days.

3.3 Calculated results

(1) Bred uranium

a. Decontamination of thorium

From the criteria defined by eq.3, the maximum concentration of thorium allowable in bred uranium x^*_o, which provides equivalent γ-MeV·Ci to that inherent in bred uranium after fabrication time t_F, is given as following :

$$x^*_o = \left(\frac{228}{232}\right)\left(\frac{\lambda_{22}}{\lambda_{08}}\right)\left(\frac{y_{22}}{y_{08}}\right) f(t_F) \quad (6)$$

with

$$f(t_F) = \left[\frac{\frac{1-e^{-\lambda_{08}t_F}}{\lambda_{08}} - \frac{1-e^{-\lambda_{244}t_F}}{\lambda_{244}}}{e^{-\lambda_{08}t_F} - e^{-\lambda_{244}t_F}} \right]$$

Equation (6) indicates that x^*_o is a function of the ratio of isotopic content and build-up rate, with respects to ^{228}Th and ^{232}U. Though ^{228}Th builds up its activity faster than ^{232}U, the effect of higher isotopic content of ^{232}U, compared with that of ^{228}Th, is more predominant to give very large x^*_0, i.e. low decontamination of Th from bred U, as shown in Fig. 4.

According to the generation mechanism of ^{228}Th, the ratio of the isotopic content y_{22}/y_{08} does not depend so much on the reactor type, except for the PWR fueled with denatured U, where ^{232}U is artificially diluted with ^{238}U. Therefore, required decontamination of Th from bred U become a little higher in this cycle, by a factor of ten, although it still belongs to low decontaminating value.

In conclusion, the criterion suggests that high separation of thorium from bred uranium is not necessary for all cycles selected. For increasing proliferation resistance, "Coprecessing" concepts could be applied to thorium cycle also. However, serious burdens associated with increased activities must be considered as suggested from x^*_o in the Fig. 4, as well as the risk of material modificability.

b. Decontamination of fission product

By the criteria of eqs. (2) and (3), the required decontamination factor $(DF)^*_i$ on FP-i is given as follows ;

$$(DF)^*_i = \left(\frac{1}{\alpha_{22}}\right)\left(\frac{Z_{i,o}\, E_i}{a_{22}(t_F)\, y_{22}}\right) \qquad (7)$$

where $Z_{i,o}$ is content in Ci of fission product i in spent fuels at t_F = o.

Table 6 compares calculated $(DF)^*_i$ on major fission products for the selected thorium cycles. Depending on assumed t_F, large reduction of decontamination factor are allowed, especially in the PWR fueled with Th-enriched U. In HTGR fuel, required DF become higher by a factor of thirty, because of lower isotopic content of ^{232}U in the bread uranium

and higher content of fission products in the spent fuel. Considering complex refabrication process for HTGR fuel, adaptation of high decontamination process, which will be effective only by quick refabrication after separation, might be justified.

c. Decontamination of plutonium

From eq. (4), maximum plutonium concentration x^*_3 is given as following;

$$x^*_3 = \left[\frac{\sum_j RHI_{j,U}(t_F)}{\sum_j RHI_{j,Pu}(t_F)} \right] \tag{8}$$

Calculated results in Table 7 indicate that required separation of plutonium also is not so high. Predominant radionuclides that affect on total RHI are ^{238}Pu, ^{241}Pu and ^{232}U. As in the case of FPs, HTGR fuel with less ^{232}U isotopic content requires higher decontamination of plutonium by a factor of ten than in the other cycles. With respects to PWR fueled with denatured uranium, in which isotopic content of ^{238}Pu is smaller, the effect is compensated by less ^{232}U generation, giving the almost equivalent value to the other Th cycles fueled with high enriched uranium.

(2) Recovered thorium

a. Decontamination of uranium

The maximum concentration of impurity U allowable in recovered Th x^*_2, is given as following ;

$$x^*_2 = \psi \left(\frac{\lambda_{02}}{g(t_s)} \right) \left(\frac{1}{y_{22}} \right) \tag{9}$$

with

$$g(t) = \left(\frac{\lambda_{08} \cdot \lambda_{22}}{\lambda_{08} - \lambda_{22}} \right) \left(e^{-\lambda_{22} t} - e^{-\lambda_{08} t} \right)$$

For direct refabrication case (ψ = 1 to 5), calculated results are given in Fig. 5 as a function of isotopic content of ^{232}U. It shows that a very fine separation of uranium is required to make a direct refabrication of Th, contrasting to the low decontamination allowed in the separation of thorium from uranium. The reason is the longer half

life of ^{232}U compared with ^{228}Th. With respects to HTGR and PWR fueled with denatured U, less stringent separation will be possible, because of their lower isotopic content of ^{232}U.

b. Decontamination of fission products

Required decontamination factor $(DF)^*_i$ of FP i is given by eq. (3) and (5).

$$(DF)^*_i = \frac{Z_{i,SF}(\Sigma_j B_{j,1}(t_s)\cdot\bar{E}_j)}{\alpha_{08}(\Sigma_j B_{j,08}(t_s)\cdot\bar{E}_j)\cdot y_{08}}$$

$$= (\frac{\psi}{1.4})(\frac{1}{\alpha_{08}})(\frac{\lambda_{02}}{\lambda_{08}})Z_{i,SF}\ (\Sigma_j B_{j,i}(t_s)\cdot\bar{E}_j) \qquad (10)$$

By the definition of ψ, which is related with the storage time t_s by eq. (5), $(DF)^*_i$ become independent of initial isotopic content of ^{228}Th. The effect of reactor type is shown in storage time t_s only. Table 8 summerizes the required decontamination factor on major fission products and storage time for corresponding ψ value. Although rather high DF is required for long lived nuclides such as ^{90}Sr, ^{137}Cs, ^{147}Pm and ^{154}Eu, the DF for ^{106}Ru which is one of the most troublesome fission product, is very small. With respects to ^{95}Zr, ^{95}Nb and ^{233}Pa also, required DF's can be negligible because of their short half lives.

c. Decontamination of plutonium

Eq. (8) is applied by replacing t_F with t_s for ℓ_o = Th.

Result in Table 7 indicate that high separation of plutonium also is required for thorium.

3.4 Discussion

Product specifications on radioactivity in two representative thorium cycles are summerized in Table 9.

Specifications of bred uranium are closely related with a refabrication time after separation, t_F. Although its rapid refabrication is preferred, longer t_F than ten days will be required if such practical considerations are made as hold-up of aged fuels or contamination of equipments by them. Therefore, low decontamination of Th, Pu and FPs will be acceptable, especially in PWR fueled with Th-enriched U

cycle. Compared with U-Pu fueled LWR's, possible reduction in decontamination factor will be more than 10^3 to 10^4.

However, the resultant activity of the product U become far higher than U-Pu fueled LWR's. This intense radioactivity will provide proliferation resistance with the fuel, comparable with such proposals as the spiked fuel concept[20] and the partial reprocessing[21]. The time-increasing character of the activity related with ^{232}U will be another advantage over the above proposals, with respects to available conversion time. If a simplified Thorex process with single cycle were introduced, the merit of process simplification could be compatible with the requirement for proliferation resistance, while this requirement will give additional burdens in U-Pu LWR cycle.

On the other hand, product specifications required for Th become a function of storage time t_s. The classical recycle policy consisting of "long-term-storage" and "direct refabrication" might be affected by the urgency for effective utilization of the resource. In order to clarify the effect of t_s or ψ on the required specification, calculations were made for Th-enriched U PWR cycle, by applying the same criterion as previsouly defined, to extended time scale of t_s. As shown in Fig. 6, the required decontamination increases with t_s until ^{228}Th activity decays to the level of ^{232}Th's. Necessarily, the highest decontamination is required in the case of direct refabrication which is possible for smaller ψ than about five. In the storage time shorter than ten years, required DF's for fission products do not exceed 10^3 because ^{228}Th does not decay substantially within the time, as shown in Fig. 3. Required DF's for those troublesome nuclides such as ^{103}Ru, ^{106}Ru and ^{95}Zr are kept within small values throughout the whole storage time, since their decay rates are larger than that of ^{228}Th. Although rather high separation is needed for long lived FPs like ^{90}Sr, ^{137}Cs and rare earths, it will be done without difficulties even in first cycle only.

If the Th is to be refabricated directly, a difficult problem is to remove the impurity Pu or U to a very high extent, because of the slow decay rates of ^{232}U and most Pu, and of ^{241}Am build-up. It might be better not to make any high decontamination of them at the time of reprocessing, but to make additional separation before the refabrication, since their separation can be done more easily at that time.

In conclusion, thorium cycle fuels have some advantageous features in proliferation resistance, because of possible adaptation of low

decontamination process justified in this cycle. Before the actual application, however, further analyses are needed such as flowsheet studies on the reprocessing process, evaluation of the shielding requirement and its effects on the fuel cycle cost. Also, it should be noted that, to attain final goal, it is essential to develop remote handling technologies and a refabrication process oriented to this direction.

4. Conclusion

In order to characterize the radioactivity of Th cycle fuels, compositions of spent fuel were calculated with the ORIGEN code on the following Th fueled reactors ; (1) PWR fueled with Th-enriched U, (2) PWR fueled with Th-denatured U, (3) CANDU fueled with Th-enriched U and (4) HTGR fueled with Th-enriched U. Using these data, product specifications on radioactivity in the reprocessing of these fuels were calculated by a criterion defined that radioactivities due to foreign element do not exceed those inherent in bred U, mainly due to ^{232}U or in recovered Th due to ^{228}Th.

Major conclusions are as the following:

(1) More ^{232}U and ^{228}Th are produced by a factor of ten in PWR's than in HTGR, because of harder spectrum in PWR core.

(2) This greater ^{232}U content in PWR justifies the introduction of low decontamination process to the reprocessing of bred U.

(3) Although the product specification of Th depends on its storage time, low decontamination process will be adaptable, even if sufficient storage time is assumed for direct refabrication.

(4) In the above case, however, additional separation of U and Pu from Th will be needed before their refabrication, since greater radioactivities or hazard due to long-lived ^{232}U and Pu isotopes are left after the long time storage.

(5) Effect of reactor type is rather small. The lowest decontamination is allowed for PWR fueled with Th-enriched U and the highest is for HTGR fueled with Th-enriched U, while the difference remains within a factor of ten to thirty.

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Table 1 Summary of assumed characteristics on thorium fueled reactor and their fuel mass balance

Fuel cycle		PWR fueled with Th and enriched U	PWR fueled with Th and denatured U	CANDU fueled with Th and enriched U	HTGR fueled with Th and enriched U
Assumed reactor performances [a]					
Electric power,	MW(E)	1000	1000	1000	1160
Average sp. power,	MW/kg	30	30	23.2	65.1
Average burn-up,	MWd/kg	33	33	27	95
Thermal efficiency,	%	34.2	34.2	30.5	38.7
Load factor,	%	80	80	80	80
Annual charge in equilibrium [a]					
Thorium,	Mg/GW(E)	24.0	19.0	34.5	7.19
Uranium,	"	1.58	6.59	0.945	0.755
U-233,	"	0.43	0.40	0.47	0.18
U-235,	"	0.54	0.73	0.18	0.37
Total heavy metal,	"	25.6	25.6	35.5	7.95
Annual discharge in equilibrium [b]					
Thorium,	Mg/GW(E)	23.3	18.5	33.5	6.65
Protactium,	"	0.0018	0.0015	0.0036	5.5×10^{-4}
Uranium,	"	1.33	6.04	1.05	0.47
U-233,	"	0.49	0.43	0.62	0.19
U-235,	"	0.22	0.33	0.060	0.05
Neptunium	"	0.029	0.039	0.0075	0.020
Plutonium,	"	0.016	0.119	0.0036	0.015
Fissile Pu,	"	0.0048	0.090	9.8×10^{-4}	0.0033
Americium,	"	1.3×10^{-4}	2.5×10^{-3}	3.0×10^{-5}	4.1×10^{-4}
Curium,	"	1.9×10^{-5}	2.9×10^{-4}	3.7×10^{-6}	2.2×10^{-4}
Total heavy metals,	"	24.7	24.7	34.5	7.16
Fission products,	"	0.88	0.88	0.98	0.79
Hulls,	"	6.93 [c]	6.93 [c]	10.6 [c]	82.7 [d]

a) Assumed based on Pigford-Yang's report (ref.11) b) Calculated results with ORIGEN

c) Zircalloy d) Graphite and SiC

Table 2 Example of nuclear data used and comparison of calculated results with those by a burn-up code

Actinide	Nuclear data used for ORIGEN calculation[a]				Calculated Results on Actinide Composition[b]		
	Thermal Cross Section[d]		Resonance Integral		ORIGEN	CEPAK[c]	ORIGEN/CEPAK
	σ_c	σ_f	$(RI)_c$	$(RI)_f$			
^{232}Th	7.40	0.00	18.1	0.0	0.912	0.914	0.998
^{233}Pa	41.46	0.00	9.2	0.0	1.0×10^{-3}	1.25×10^{-3}	0.80
^{233}U	46.0	525.	100.	595.	0.018	0.0160	1.13
^{234}U	95.8	0.0	320.	0.	9.0×10^{-3}	9.23×10^{-3}	0.98
^{235}U	101.3	577	130.	260.	8.6×10^{-3}	7.55×10^{-3}	1.14
^{236}U	6.0	0.0	150.	0.	0.0137	0.0137	1.00
^{238}U	2.73	0.0	250.	0.	1.34×10^{-3}	1.37×10^{-3}	0.98
^{239}Pu	27.1	741.9	170.	250.	1.52×10^{-4}	—	—

a) In barn

b) Ton/ton-initially charged heavy metal

c) Calculated from Pigford-Yong's report (ref. 11)

d) Same as original ORIGEN data for U fueled PWR, except for ^{239}Pu

Table 3 Radioactive characteristics of discharged fuel from thorium cycle, after one year cooling.
(Unit : Ci/Mg-initial charged fuel)

Fuel cycle	PWR fueled with : Th and enriched U	PWR fueled with : Th and denatured U	CANDU fueled with: Th and enriched U	HTGR fueled with: Th and enriched U
Hulls [b]	1.4 (+4) [a]	1.2 (+4)	1.5 (+4)	5.2 (+2)
Actinides				
^{228}Th	2.5 (+3)	2.2 (+3)	5.8 (+2)	1.9 (+2)
^{232}Th	1.0 (-1)	7.9 (-2)	1.0 (-1)	9.2 (-2)
Total Th	3.1 (+3)	3.5 (+3)	1.5 (+4)	2.0 (+2)
^{233}Pa	1.8 (+3)	1.2 (+3)	1.7 (+3)	4.5 (+3)
Total Pa	1.9 (+3)	1.3 (+3)	1.8 (+3)	4.6 (+3)
^{232}U	2.9 (+3)	2.5 (+3)	8.1 (+2)	2.9 (+2)
^{233}U	1.8 (+2)	1.6 (+2)	1.7 (+2)	2.2 (+2)
^{234}U	5.6 (+1)	4.2 (+1)	3.1 (+1)	6.2 (+1)
^{237}U	9.1 (-1)	1.8 (+0)	1.4 (-2)	3.0 (-1)
Total U	3.1 (+3)	2.7 (+3)	1.1 (+3)	5.9 (+2)
^{239}Np	4.2 (-1)	6.6 (+0)	7.6 (-2)	8.7 (+0)
Total Np	1.3 (+0)	7.6 (+0)	2.3 (-1)	1.1 (+1)
^{238}Pu	7.2 (+3)	9.3 (+3)	1.1 (+3)	2.2 (+4)
^{241}Pu	3.9 (+3)	7.9 (+4)	6.0 (+2)	1.2 (+4)
Total Pu	1.2 (+4)	8.8 (+4)	1.7 (+3)	3.5 (+4)
Total Am	1.0 (+1)	2.1 (+2)	1.5 (+0)	5.5 (+1)
Total Actinides	3.8 (+4)	1.2 (+5)	1.5 (+4)	5.6 (+4)
Fission products				
^{3}H [c]	3.8 (+2)	4.4 (+2)	3.2 (+2)	1.1 (+3)
^{85}Kr	2.2 (+4)	1.9 (+4)	2.1 (+4)	5.9 (+4)
^{129}I	4.8 (-2)	4.5 (-2)	4.4 (-2)	1.3 (-1)
^{131}I	1.7 (-8)	1.8 (-8)	1.3 (-8)	3.6 (-8)
^{89}Sr	1.1 (+4)	9.0 (+3)	8.6 (+3)	2.3 (+4)
^{90}Sr, ^{90}Y	1.1 (+5)	9.7 (+4)	9.0 (+4)	2.9 (+5)
^{91}Y	1.9 (+4)	1.7 (+4)	1.5 (+4)	4.1 (+4)
^{95}Zr	3.1 (+4)	3.0 (+4)	2.4 (+4)	6.6 (+4)
^{95}Nb	6.5 (+4)	6.4 (+4)	5.1 (+4)	1.4 (+5)
^{103}Ru	9.2 (+2)	1.3 (+3)	6.3 (+2)	2.1 (+3)
^{106}Ru, ^{106}Rh	3.6 (+4)	1.1 (+5)	2.3 (+4)	9.9 (+4)
^{134}Cs	1.4 (+5)	1.4 (+5)	7.8 (+4)	5.8 (+5)
^{137}Cs	1.1 (+5)	1.0 (+5)	8.7 (+4)	3.0 (+5)
^{141}Ce	6.5 (+2)	6.3 (+2)	5.1 (+2)	1.5 (+2)
^{144}Ce	4.7 (+5)	4.6 (+5)	3.5 (+4)	1.1 (+6)
Total rare earths	1.0 (+6)	1.0 (+6)	7.9 (+5)	2.3 (+6)
Total β Ci	1.7 (+6)	1.8 (+6)	1.3 (+6)	4.3 (+6)
Total gross Ci	1.8 (+6)	1.9 (+6)	1.4 (+4)	4.6 (+6)

a) $1.4(+4) = 1.4 \times 10^4$ b) Not include activities due to broken fuel

c) Not consider generation due to impurities and coolant

Table 4 Isotopic content of bred U and recovered Th

Isotope	PWR		CANDU	HTGR
	Th-enriched U	Th-denatured U	Th-enriched U	Th-enriched U
^{232}U	0.27 wt.%	0.050 wt.%	0.12 wt.%	0.024 wt.%
^{233}U	37.2	7.15	59.4	40.0
^{234}U	17.2	2.85	16.9	16.8
^{235}U	16.6	5.50	5.69	10.9
^{236}U	26.1	7.61	12.5	25.5
^{237}U	2.1×10^{-9}	9.4×10^{-9}	5.8×10^{-10}	6.2×10^{-9}
^{238}U	2.6	76.8	5.16	6.59
^{228}Th	3.3×10^{-4}	3.7×10^{-4}	7.6×10^{-5}	2.8×10^{-5}
^{229}Th	9.3×10^{-5}	9.9×10^{-5}	6.7×10^{-5}	5.9×10^{-5}
^{230}Th	1.1×10^{-4}	1.1×10^{-4}	1.8×10^{-4}	8.5×10^{-3}
^{232}Th	99.99	99.99	99.99	99.99
^{234}Th	2.0×10^{-10}	4.8×10^{-10}	2.6×10^{-10}	1.1×10^{-7}

a) For nearly equilibrium cycle, 1 yr. cooling

Table 5 Comparison of cooling time criteria on thorium cycle fuel

Cooling time criteria	PWR		CANDU	HTGR
	Th-enriched U	Th-denatured U	Th-enriched U	Th-enriched U
Decay of ^{131}I a)	173 days	175 days	170 days	170 days
Recovery of ^{233}Pa b)	150	145	155	180

a) Relative ammount of ^{131}I in spent fuel to that of ^{129}I be less than the ratio of their $(MPC)_a$.

b) Undecayed fraction of ^{233}Pa be less than 0.1% to total potential ^{233}U, i.e. sum of ^{233}U and ^{233}Pa.

Table 6 Required decontamination factor of fission products for bred uranium from thorium cycles[a)]

Fabrication within	PWR		CANDU	HTGR
	Th-enriched U	Th-denatured U	Th-enriched U	Th-enriched U
1 day after separation				
Total FPs	4×10^4	2×10^5	6×10^4	2×10^6
^{95}Zr	1×10^3	5×10^3	2×10^3	5×10^4
^{95}Nb	5×10^3	2×10^4	8×10^3	1×10^5
^{103}Ru b)	4×10^1	3×10^2	6×10^1	1×10^2
^{106}Ru b)	1×10^3	2×10^4	2×10^3	1×10^5
^{134}Cs	2×10^4	1×10^5	3×10^4	1×10^6
Total rare earths	3×10^3	1×10^4	4×10^3	1×10^5
^{233}Pa	3×10^1	1×10^2	5×10^1	7×10^2
10 days after separation				
Total FPs	9×10^2	5×10^3	1×10^3	2×10^4
^{95}Zr	3×10^1	1×10^2	4×10^1	7×10^2
^{95}Nb	1×10^2	6×10^2	2×10^2	2×10^3
^{103}Ru b)	1×10^0	7×10^0	1×10^0	1×10^2
^{106}Ru b)	3×10^1	4×10^2	4×10^1	2×10^3
^{134}Cs	5×10^2	3×10^3	5×10^2	1×10^4
Total rare earths	6×10^1	3×10^2	9×10^1	1×10^3
^{233}Pa	5×10^{-1}	3×10^0	1×10^0	2×10^1

a) For compositions after one year cooling

b) Determined by γ-Ci from daughter

Table 7 Decontamination of plutonium from bred uranium and recovered Thorium

Pu concentration allowable in	PWR fueled Th-enriched U	PWR fueled Th-denatured U	CANDU fueled Th-enriched U	HTGR fueled Th-enriched U
Bred Uranium	400 ppm	300 ppm	100 ppm	40 ppm
Recovered Thorium				
$\Psi = 1$ [a)]	5 ppb	9 ppb	6 ppb	4 ppb
$\Psi = 5$	20	30	20	10

a) Ψ ; Activity ratio of ^{228}Th to ^{232}Th in recovered Th after long time storage

Table 8 Decontamination factor of long lived fission products in storaged thorium

Fuel Cycle		PWR fueled Th-enriched U	PWR fueled Th-denatured U	CANDU fueled Th-enriched U	HTGR fueled Th-enriched U
$\Psi = 1$ [a]					
Storage time		28 yrs.	28 yrs.	24 yrs.	21 yrs.
Decontamination Factor	^{90}Sr [b]	1×10^{6}	1×10^{6}	4×10^{5}	5×10^{6}
	^{106}Ru	1	1	1	2
	^{134}Cs	2×10^{2}	2×10^{2}	3×10^{2}	2×10^{4}
	^{137}Cs	1×10^{6}	2×10^{6}	1×10^{6}	6×10^{6}
	total R.E.	4×10^{6}	6×10^{6}	3×10^{4}	2×10^{5}
$\Psi = 5$					
Storage time		23 yrs.	24 yrs.	19 yrs.	16 yrs.
Decontamination Factor	^{90}Sr	7×10^{5}	8×10^{5}	2×10^{5}	3×10^{6}
	^{106}Ru	1	1	1	1
	^{134}Cs	5×10^{2}	5×10^{2}	6×10^{2}	3×10^{4}
	^{137}Cs	7×10^{5}	9×10^{5}	7×10^{5}	3×10^{6}
	Total R.E.	2×10^{4}	4×10^{4}	2×10^{4}	1×10^{5}

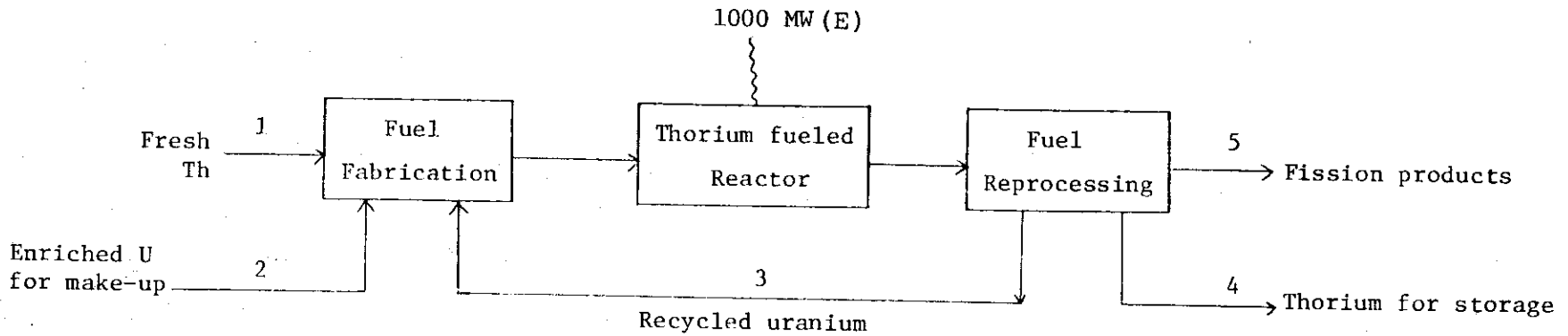
a) Ψ ; Activity ratio of ^{228}Th to ^{232}Th in storaged thorium

b) R.E.; Rare earth, c) Calculated by γ -activity due to the daughter

Table 9 Summary of required decontamination in reprocessing thorium cycle fuels

Fuel cycle		Low decontamination case (T_F=10 days, Ψ=5)[a]		High decontamination case (T_F=1 day, Ψ=1)[a]	
		PWR fueled with Th-95% ^{235}U	HTGR fueled with Th-93% ^{235}U	PWR fueled with Th-95% ^{235}U	HTGR fueled with Th-93% ^{235}U
Bread Uranium					
Radioactivity		530 γ-MeV·Ci/T	43 γ-MeV·Ci/T	11 γ-MeV·Ci/T	1 γ MeV·Ci/T
Total fissile		54 w/o	51 w/o	54 w/o	51 w/o
Decontamination	Th	1.3×10^4 ppm	1.4×10^4 ppm	1.0×10^4 ppm	1.1×10^4 ppm
	Pu	400 "	40 "	400 "	40 "
	Total FP	9×10^2 (DF)	2×10^4 (DF)	4×10^4 (DF)	2×10^6 (DF)
Recovered Thorium					
Storage time		23 yrs.	16 yrs.	28 yrs.	21 yrs.
Decontamination	U	6 ppm	90 ppm	1 ppm	20 ppm
	Pu	20 ppb	10 ppb	5 ppb	4 ppb
	Total FP	2×10^6 (DF)	1×10^7 (DF)	3×10^6 (DF)	2×10^4 (DF)

a) T_F; Time-after-separation before which the material is refabricated.
Ψ; Activity ratio of ^{228}Th in ^{232}Th in storaged fuel, defined as
$\Psi = [(\lambda_{08}N_{08}) / (\lambda_{02}N_{02})]$ in recovered Th.



Stream No.	PWR		CANDU	HTGR
	Th-enriched U	Th-denatured U	Th-enriched U	Th-enriched U
1	24.2 Mg/y	19.2 Mg/y	34.8 Mg/y	7.26 Mg/y
2	0.28	0.68	0.13	0.29
3	1.32	5.98	0.89	0.47
4	23.1	18.3	33.5	6.65
5	0.9	0.9	1.0	0.79

Fig. 1 Reference fuel cycles and their mass flow per GW(E)·year

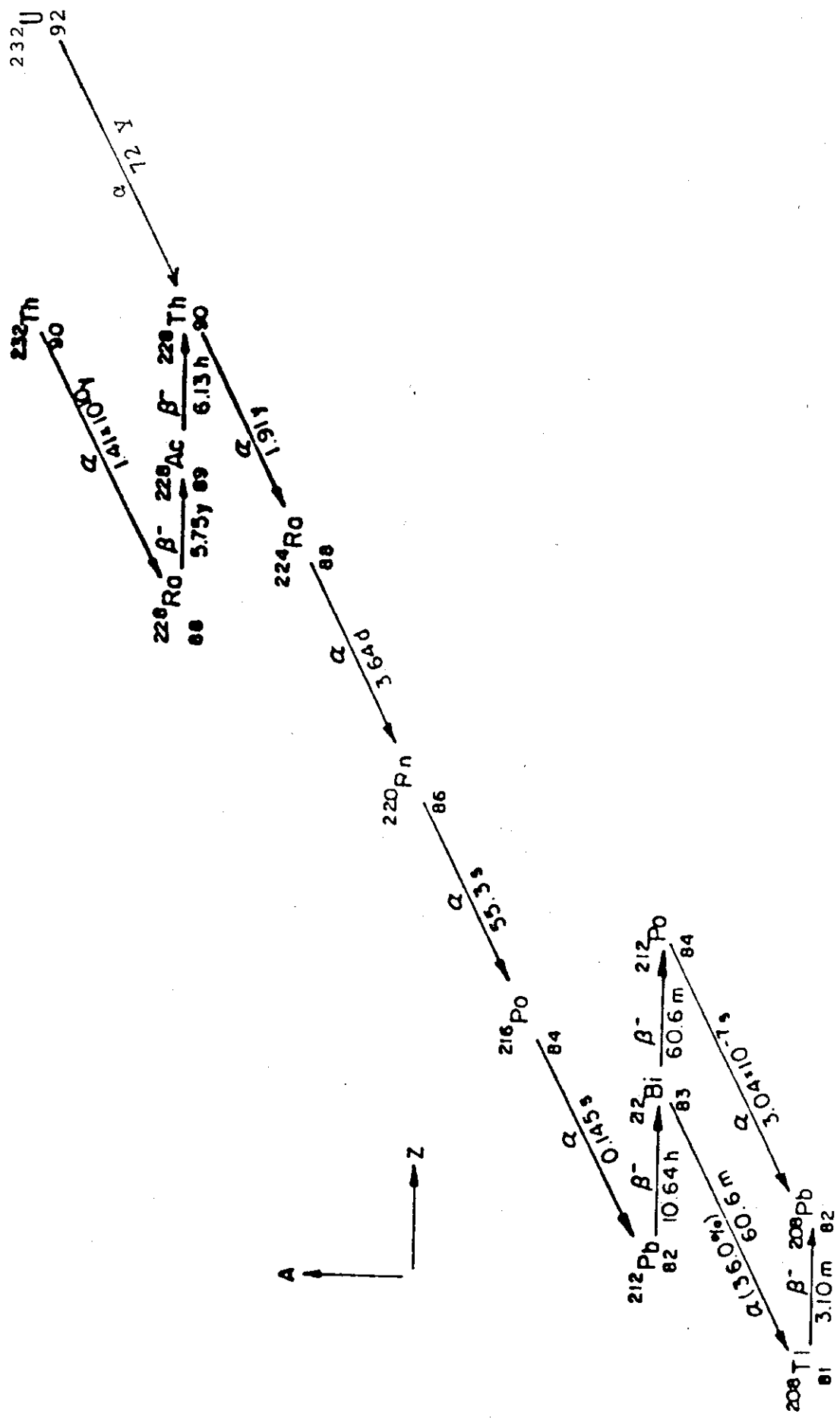


Fig. 2 Decay chain of ^{232}U and ^{228}Th (From Ref. 17)

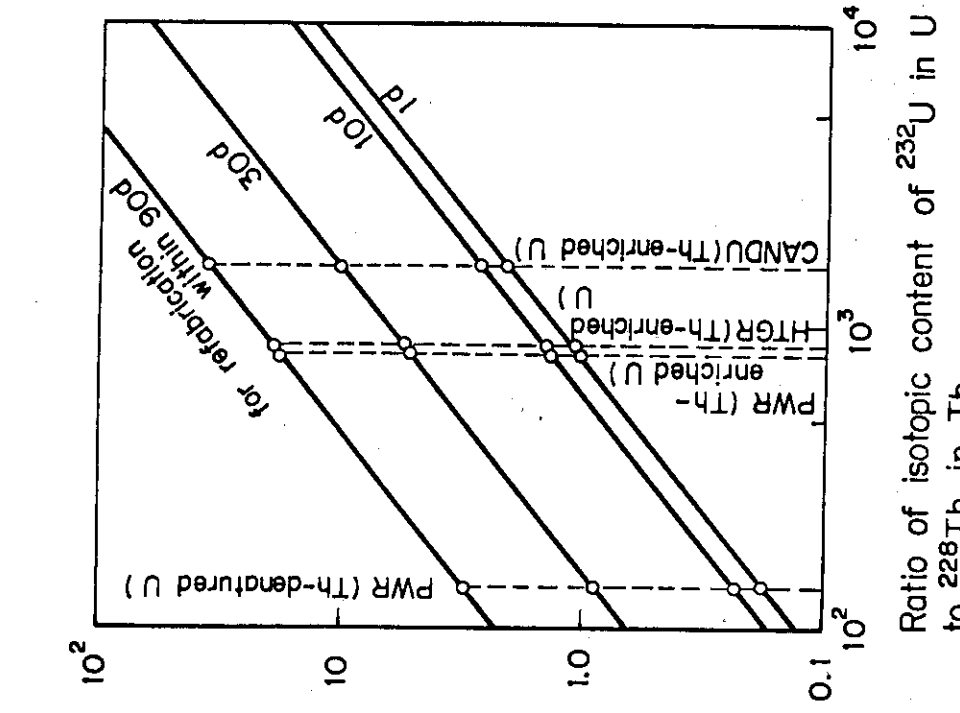


Fig. 4 REQUIRED DECONTAMINATION OF THORIUM FROM BRED URANIUM

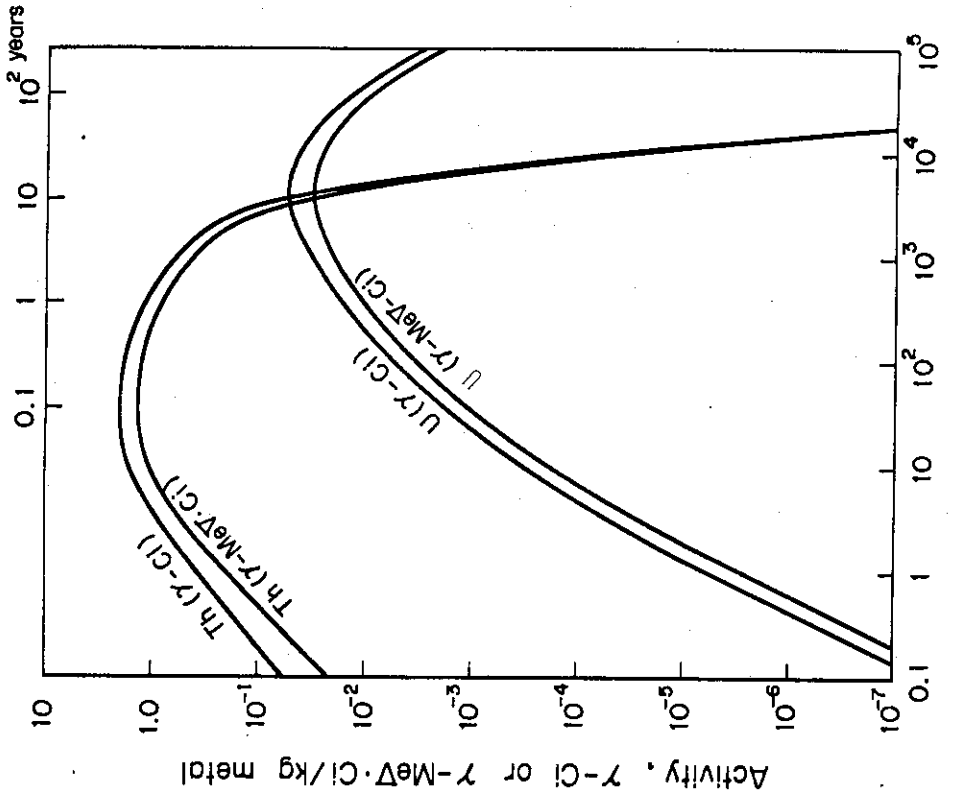


Fig. 3 RADIOACTIVITY BUILD-UP AND DECAY OF UNIT KG URANIUM AND THORIUM WITH ISOTOPIC CONTENT OF ONE PPM ^{232}U AND ^{228}Th, RESPECTIVELY

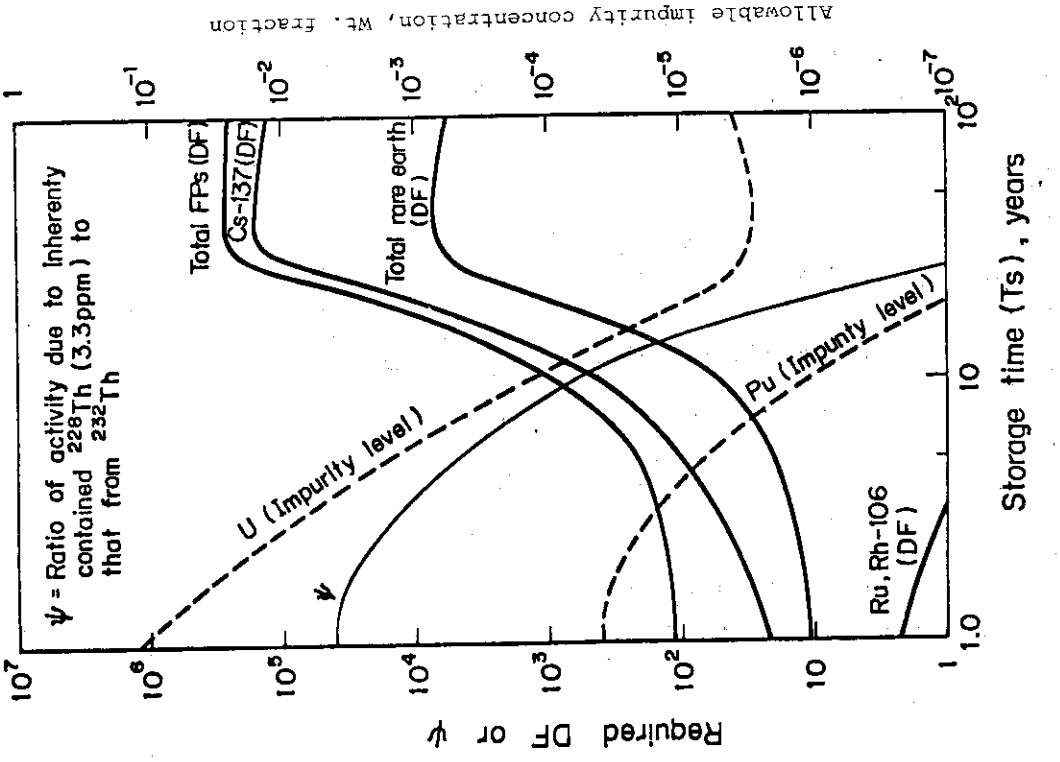


Fig. 6 CHANGE OF REQUIRED DECONTAMINATION IN REPROCESSING LONG-TERM STORAGED THORIUM

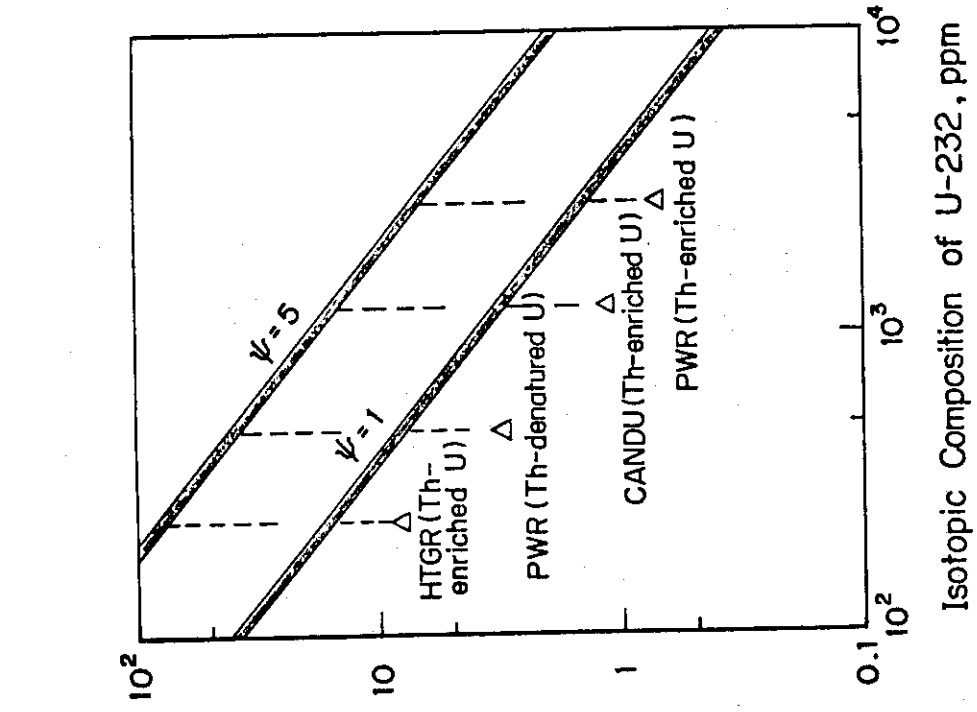


Fig. 5 REQUIRED DECONTAMINATION OF URANIUM FROM RECOVERED THORIUM