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DECAY AND STORAGE OF IRRADIATED FUEL

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DECAY AND STORAGE OF IRRADIATED FUEL

ABSTRACT

Reasons for storage to allow decay of irradiated fuel elements and possible methods of storage are given. The effects on storage and inventory costs of fuel element composition, plant size, element geometry, reactor type, methods of irradiation and recycle, and type of metallurgical handling are discussed. Estimates and comparisons are included for the decay for several typical fuels. The special problems associated with thorium fuel elements are considered.

INTRODUCTION

This paper lists a number of variables which determine the duration and cost of holdup of irradiated fuel before and after chemical processing. The costs of decay is composed of three types of charges: (1) the cost of physical storage, (2) the use charge for any fissionable material leased from the AEC, and (3) an inventory cost for working capital tied up in fissionable material bought from the AEC or awaiting sale to a customer. To determine the total cost, the following items must be defined: (1) the storage space required, (2) the cost of the space and its operation, (3) the base charge for holding the fuel material,^{1,2} and (4) how long the material will be held.

If material is leased, the basis for the use charge in most cases will be the value as taken from the AEC. If the material is purchased or awaiting sale, the working capital represented by process inventory should be calculated, based on its form at the particular moment in the processing cycle.

The decay time required depends on the radiation levels which can be tolerated for various refabrication techniques. Firm criteria have not been established to fix these levels. Since power reactors will discharge fuels which are highly irradiated by present standards, either long decay periods or a form of remote refabrication will be necessary. The economics of these alternates have not yet been fully investigated. In the meanwhile,

it is taken as axiomatic in this paper that the cost of fabricating elements will be increased by the presence of any residual radioactivity beyond that found in natural thorium or uranium.

The length of the decay time should be based more upon the biological requirements and tolerances for the type of processing or refabrication necessary than upon arbitrary comparisons with natural uranium. Whereas a long time is required to reduce the fissile or fertile daughter activity to less than normal background for direct metallurgy, the decay period may be much shorter provided semi-remote metallurgy may be performed. The concept of semi-remote metallurgy entails only lucite and/or distance shielding.

The activity levels for semi-remote refabrication are assumed to be 30 mr/hr at 1 ft. This activity level will deliver a dose of 1200 mr per 40-hr week to the hands and 300 mr per 40-hr week to the total body, which is taken to be 2 ft from the source.

REASONS FOR DECAY STORAGE

Fission Product Decay

If fuel elements are stored after discharge from a reactor, their radioactivity decreases with time. Reduction of fission product activity simplifies problems of heat removal during transport or storage. Also, if any process reagents or plant materials of construction are susceptible to radiation damage, sufficient decay reduces or eliminates the difficulty. Required shield thicknesses are reduced by decay. Since 12.8-day Ba^{140} (La^{140}) controls shielding up to about 90 days' decay, considerable savings in shield weight can be made by permitting decay before handling. Finally, processing is simplified if there are fewer species of fission products to be removed from the fuel. For example, 8-day I^{131} can be eliminated from process consideration after about 100 days' decay.

Transmutation Product Decay (Fig. 1)

Neptunium²³⁹ is an intermediate in the production of Pu^{239} , and Pa^{233} is an intermediate in the production of U^{233} . Unless it is economically feasible to lose the unconverted intermediate, it must be either recovered or allowed to decay to the product. The decay can be performed before the initial separation, or a second separation can be made after the waste from the first step has decayed. Since almost a year's decay is required to

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Fig. 1. Transmutation Isotope Production Routes.

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reduce the Pa^{233} content to 0.1 percent of its value at time of discharge, a decay period this long may be ruled out as leading to excessive inventory charges. On the other hand, since only a month is required for comparable Np^{239} decay, it may be practical to allow decay before processing.

Uranium²³⁷ is produced in the irradiation of U^{235} . Similarly, Th^{234} is produced in the irradiation of Th^{232} . Product decay may be required since any undecayed U^{237} will carry through the processing with the recovered uranium and any undecayed Th^{234} will appear in the recovered thorium.

Seventy-year U^{232} is produced from Th^{232} , and decays to 1.9-year Th^{228} and its daughters. This leads to a particularly nasty situation: a short decay period will lead to U^{232} in the U^{233} product and some Th^{228} in the thorium product with subsequent ingrowth of activity into both; a longer decay period will increase the activity in the thorium product which, since it is much larger in bulk than the U^{233} , does not help the situation. Processing after a short decay time to minimize the activity of the thorium product with periodic repurification of the uranium product has been proposed. Since such a scheme will feed appreciable quantities of Pa^{233} and Th^{234} to the process, storage of protactinium-bearing waste and product thorium may be required.

Buildup of Pu^{240} by continued recycle of plutonium may increase gamma and x-ray background by a factor of 4 over that of pure Pu^{239} . It also appears that the Pu^{238} content may have to be limited to keep the alpha-neutron reactions within the allowable neutron background.

Table 1 is a summary of the proposed methods of transmutation product handling.

METHODS OF STORAGE

Storage Before Processing

Before processing, fuels can be stored in some type of basin or vessel or possibly mounted as a mobile radiation source. Fluid fuels may best be stored in some type of vessel. The vessel can then be handled in a manner similar to an assembly of clad metallic elements.

The usual method of storing clad metallic elements is to keep them in a canal filled with water to a sufficient depth to act as an effective

Table 1. Summary of Transmutation Isotope Situations

Np^{239} in Pu^{239}	Allow one month's decay of fuel or recover Np^{239}
U^{237} in U^{235}	Allow 120 days' decay of fuel, or increase refabrication cost
Pa^{233} in U^{233}	Allow one year's decay of fuel or waste, or recover Pa^{233}
U^{232} in U^{233}	Repurify periodically to remove U^{232} daughters
Th^{234} in Th^{232}	Allow one year's decay of fuel or product, or increase refabrication cost
Th^{228} in Th^{232}	Allow less than 60 days' decay of fuel to minimize product activity, or increase refabrication cost

radiation shield. In all cases encountered to date, natural convection in the basin has been sufficient to cool the elements. The storage of Na- or NaK-bonded elements presents a problem because of the danger of reaction with water.

The canal water may become contaminated as a result of cladding ruptures or erosion. Ion exchange is used to keep the water decontaminated of excess activity and clean for good visibility.

Provision must be made in a storage canal to unload elements from a large carrier arriving from the reactors. The unloading facilities, which include a heavy-duty crane, can also be used to load chargers when elements are ready for processing.

Highly enriched U^{235} alloys are subject to restrictions to prevent assembly of a critical mass. The number of elements that may be clustered in any heap or bucket is usually limited and a minimum spacing is kept between clusters. Criticality control can be achieved by storing fuel pieces in baskets suspended in the water from trolley rails. The suspended baskets cannot approach each other by virtue of the construction of the equipment. Light-duty hoists are used to transfer the baskets around the canal area.

Storage After Processing

If elements are processed after a short decay time and direct fabrication is desired, it may be necessary to provide storage to permit decay of Pa^{233} , U^{237} , Th^{228} and Th^{234} . Under present conceptions the materials are stored as solutions in suitably shielded vessels and subsequently withdrawn for recycle or shipment.

The inventory charge for storage of off-standard streams is negligible since very little material is recycled, and it is held up for only a short time. Storage requirements for an 8-hr hold period of off-standard extraction column feed material are 400 gal for a 1-ton/day Purex plant. Such a tank, including the necessary shield walls, costs approximately \$1,200.

FACTORS AFFECTING STORAGE SPACE REQUIRED

Enrichment

As the concentration of fissionable material in an element increases, the number of elements which can be safely stored per unit volume decreases. Extra investment is also required for highly enriched elements to guard

against creating a critical assembly. Against this can be balanced the fact that enriched elements will be shipped in smaller carriers, which can be handled with a lighter crane.

Plant Throughput

Assuming a given type of fuel elements with a fixed decay schedule, the area required for storage will be directly proportional to the plant throughput. In the case of enriched fuels, the cost of auxiliaries to maintain criticality control, such as baskets, will be roughly proportional to the throughput.

For a constant enrichment, over a wide range of plant throughput, the cost of the unloading area and heavy-duty crane will remain fixed. The operating costs, chiefly for personnel and water treatment, should not vary greatly with throughput.

Element Geometry

For natural uranium where criticality hazards are not considered for storage, the sole effect of fuel element geometry is to change the weight that can be stored per unit volume. In general, shorter and fatter fuel elements, as well as any having extended surfaces or coolant passages, will cost more to store per unit weight than long, thin, solid elements.

In the case of enriched uranium alloy rods there will be a spacing corresponding to a uranium-water assembly optimum for criticality. This spacing will not correspond to the most dense packing of fuel, since the water-moderated critical mass will be lower than the water-reflected one. A greater number of rods can therefore be stored tightly packed than that corresponding to the minimum possible critical mass.

Alloy Composition

Any dilution of natural uranium by alloying constituents will have the same effect on storage cost as voids in the element, namely, increased cost per unit weight of uranium stored.

For highly enriched alloys, the situation is more complex. A safe procedure would be to assume that as the percent uranium in the alloy changes, the same mass of uranium may be stored in any one container. Therefore, more dilute alloys cost more to store than more concentrated alloys. Against this may be balanced the fact that in a more dilute alloy more poison will be present and more neutron leakage will occur, and

therefore it might be permissible to store a larger mass of uranium per container. The particular range of moderator-to-uranium ratio will also have a bearing on the effect of alloy composition.

FACTORS AFFECTING DECAY TIME REQUIRED

The type of reactor, the method of irradiation, method of fuel recycle, and refabrication techniques influence the amount of decay time required and thus the cost. These variables will be illustrated by the U^{237} problem associated with natural or enriched uranium. A reactor which uses depleted uranium in the blanket and highly enriched uranium or plutonium in the core requires different decay times than one using slightly enriched uranium. A similar situation exists for thorium breeder reactors.

Reactor Type

The concentration of U^{237} as a function of specific power, flux, and irradiation time (starting with $N_{26} = 0$) may be estimated by the following equation:

$$N_{27} = 8.84 \times 10^4 P \phi \left[\lambda_{27} t - (1 - e^{-\lambda_{27} t}) \right]$$

This equation was obtained by combining the following relations:

$$P = \phi \frac{W A E}{235} \frac{\sigma_f(25)}{3.38 \times 10^{16}}$$

and

$$N_{27} = \phi^2 \frac{W A E}{235} \frac{\sigma_c(25) \sigma_c(26)}{\lambda_{27}^2} \left[\lambda_{27} t - (1 - e^{-\lambda_{27} t}) \right]$$

where P = specific power, Mw/kg U

ϕ = neutron flux, n/cm²/sec

t = irradiation time, sec

λ_{27} = decay constant of U^{237} , 1.19×10^{-6} sec⁻¹

N_{25}, N_{26}, N_{27} = $U^{235}, U^{236}, U^{237}$ concentration, atoms/kg U

W = wt of uranium, g

A = Avogadro number = 6.023×10^{23}

E = fuel enrichment, wt% U^{235}

$\sigma_f(25), \sigma_c(25), \sigma_c(26)$ = respective fission or capture cross-sections

3.38×10^{16} = fissions/Mw/sec

For long irradiations, the activity of U^{237} after the first irradiation cycle is directly proportional to the product of flux, specific power, and irradiation time. Fuel material irradiated at a flux of 1.5×10^{13} will require 15 days' less decay time than that irradiated at 7.5×10^{13} for the same time and at the same specific power. In addition, for each factor of 2 that a fuel is lower in enrichment (for the same flux and specific power) 6.7 days' less decay time will be required for U^{237} .

About 30 days' less decay time is therefore required for a fertile blanket ($0.3\% U^{235}$, $\phi = 1.5 \times 10^{13}$ n/cm²/sec) than for an internal converter fuel ($2\% U^{235}$, $\phi = 7.5 \times 10^{13}$ n/cm²/sec) irradiated to the same level. The decay time required for the highly enriched core is considerably longer than that for either the blanket or the internal converter, but less material will be stored (see comparison in Table 2).

Recycle of a Pu^{239} - U^{238} fuel, such as that used in a fast breeder reactor, can be accomplished after only 20-30 days for neptunium decay. The Pu^{239} is separated from the U^{238} and fission products and then blended with fresh U^{238} before refabrication. If the Purex process is used the separated U^{238} , as 2 M uranyl nitrate, would be held back for 120 days for U^{237} decay. The tank charges for holdup of this U^{238} fraction would be approximately \$120 per ton of U^{238} processed, with the additional inventory charges being approximately \$40 per ton of U^{238} processed. Thus it is possible to minimize both the charge due to plutonium holdup and the increased cost of remote or semi-remote refabrication with only a slight increase in the storage and inventory costs for U^{238} holdup.

Mode of Irradiation

The decay time of reactor-discharged material depends upon the method of irradiation, whether continuous (homogeneous type of operation) or batch, and, if batch, upon the number of irradiation cycles. For any batch cycle, before (effectively) an infinite number of cycles has been accumulated, the concentration of transmutation isotopes of fertile and fissile materials is lower than that for the continuous irradiation at steady state. For an infinite number of batch irradiations the transmutation product activities are the same as for the continuous case. Fission product activities are always lower in continuously discharged than in batch-discharged material.

The principal transmutation product activity in U^{235} or natural uranium is U^{237} . The difference between the U^{237} produced during a batch and a continuous irradiation depends on the U^{236} content. In batch irradiation the U^{236} content depends on both the degree of its removal in a diffusion plant and the number of times the material has been recycled. If no U^{236} removal, as in a diffusion plant, occurs, the activity of U^{237} after any cycle of batch irradiation is $N_{26}/N_{25} \times \sigma_c(26)/\sigma_c(25)$ times the U^{237} activity resulting from continuous irradiation. For an infinite number of batch cycles this ratio is 1/1, but, for example, after 16 cycles at a flux-time of 10^{21} n/cm² per cycle, the ratio is $\sigma_c(26)/\sigma_c(25)$, since N_{26}/N_{25} is 1/1.

Method of Recycle

The effect of recycle of both fertile and fissionable material through a diffusion plant such as would be the case in a one-region slightly enriched reactor can be illustrated by Fig. 2 and the following analysis. After infinite recycle of slightly enriched uranium with the burned U^{235} replaced in each pass, the U^{236}/U^{235} atom ratio charged to the reactor will be approximately 7.5/1. However, if the reprocessed fuel is recycled through the gaseous diffusion plant after each reprocessing cycle with 25% removal of U^{236} per pass, then this ratio is only 0.265/1 at a ϕt of 10^{21} n/cm² and 0.636/1 at a ϕt of 4×10^{21} n/cm². The effect of the decreased U^{236}/U^{235} ratio is illustrated in Fig. 2, which indicates the decay time necessary to reduce U^{237} radioactivity to 10^9 d/m/kg uranium. For example: with a ϕt of 10^{21} n/cm² per cycle at a flux of 5×10^{13} using 1.2% enriched fuel, the decay time required for diffusion plant-recycled fuel would be 80 days as compared to 120 days for fuel which has the equilibrium U^{236}/U^{235} ratio.

Refabrication Techniques

The permissible activity levels in natural or enriched uranium are set by the U^{237} content. For direct refabrication, the activity of U^{237} must have decayed to 1×10^9 d/m/kg U which is only 3 mr/hr at direct contact, whereas semi-remote fabrication may be performed if the activity is less than 4×10^{12} d/m/kg U. The decay time may therefore be shortened by 80 days if semi-remote fabrication is used. Using examples of 150 days for enriched uranium to decay to 1×10^9 d/m/kg (corresponding to $\phi t = 10^{21}$ n/cm², $\phi = 10^{14}$, 16 cycles), semi-remote fabrication can thus save over 50 percent of the inventory and storage charges.

Fig. 2. Decay Time Required for U^{237} to Decay to 10^9 d/m/kg Uranium. Infinite recycle; $\phi t = 10^{21}$ n/cm^2 ; batch irradiation, 1.2% U^{235} charged.

If it is desired to re-enrich uranium in a diffusion plant, inventory charges will accrue for the period necessary for U^{237} to decay to background. Alternately, the burnup can be replaced with fully enriched uranium and semi-remote fabrication used after a shorter cooling period (perhaps half as long). Since the value per unit of contained U^{235} increases with enrichment, it is necessary to balance the increased U^{235} and semi-remote fabrication costs of the second scheme against the higher inventory charges of the first.

HYPOTHETICAL EXAMPLES OF DECAY PROBLEMS

Hypothetical examples of two reactor situations and the effect of decay and burnup upon inventory costs and refabrication techniques are given in Table 2.

These conditions, although arbitrary, give some indication of the enrichment, irradiation, and decay problems associated with reactor fuels. The two-region reactor offers a somewhat different situation with regard to fuel inventory and storage costs since the inventory charge for decay of the blanket is negligible. If the blanket decay inventory is neglected it is necessary to balance the core decay charges against the slightly enriched fuel in order to compare reactors. In order to have a core decay inventory charge equivalent to that for the single-region reactor, it is necessary to have 31% burnup of U^{235} with the full 150-day decay time. The decay time and U^{237} activity associated with various burnups in order to maintain an annual inventory charge for the core equal to that for the one-region reactor are indicated in Table 3.

Charges due to plutonium holdup may be reduced by processing after 2-3 weeks' decay and holding the uranium product for U^{237} decay. The uranium product may be stored as uranyl nitrate solution containing 400 gm of uranium per liter.

THORIUM AND U^{233}

The decay of thorium brings up several additional problems. The activity of Th^{234} takes on the order of a year to decay to 10^9 d/m/kg Th; and the activity of Th^{228} which builds up during the pre-Thorex decay time decays with a 1.9-year half-life. Thorium²²⁸ and its daughter activities will make the thorium product very radioactive---in fact, much more radioactive than natural thorium. However, the activity of infinitely recycled

Table 2. Comparison of Two Reactor Types

	<u>One-Region Reactor</u>	<u>Two-Region Reactor</u>
Enrichment	2%	90% core, depleted blanket
Irradiation level	3000 Mwd/t	3000 Mwd/t (equivalent)
Irradiation time	50 days	250 days for blanket, 42 days for core
Uranium loading	10 tons	50 tons for blanket, 85 kg for core
Flux (thermal)	7.5×10^{13} n/cm ² /sec	1.5×10^{14} n/cm ² /sec for core 1.5×10^{13} n/cm ² /sec for blanket
Specific power	60 Mw/ton	6 Mw/kg U ²³⁵ , 1.8 Mw/ton depleted U
Decay time	123 days	150 days for core, 107 days for blanket

Table 3. Effects of Burnup and Decay Time on the Inventory Cost for U^{237}
Decay and U^{237} Activity^a

Basis: ~ 90% U^{235} ; inventory cost based on value of fuel after burnup

Burnup of U^{235} (%)	Decay Time Necessary to Limit Core Inventory Cost to Same as that for One-Region Reactor	U^{237} Activity (d/m/kg U^{235})	Radiation Level at 1 ft from U^{237} from 1 kg U^{235} (mr/hr)
5	20	2.5×10^{14}	2300
10	44	3.6×10^{13}	330
20	91	4.0×10^{11}	3.7
30	139	3.1×10^9	0.03
31	150	1×10^9	0.01
40	150		
50	150		
60	150		
80	150		

^aThe surface activity of a metal sphere weighing 1 kg with a U^{237} activity of 3.6×10^{13} d/m/kg = 110 mr/hr. Solutions of U^{235} containing 1 kg of U^{235} at a concentration of 450 g U^{235} per liter would have surface activities 100 times that given for pure metal (based on standard containers).

Fission product activity with 90% removal = 120-180 watts/kg U^{235} = 5-8 $\times 10^{16}$ d/m/kg U^{235} .

thorium may be kept to less than 5 times that of natural thorium by stringent limitations on reactor flux, irradiation level, and decay time. The irradiation level must be below 4000 g/t* and the reactor flux above 3×10^{13} n/cm²/sec in order to meet these specifications with any reasonable decay times (>10 days).³ Higher irradiation levels will require semi-remote refabrication of the thorium products.

The specifications for semi-remote handling based on 30 mr/hr at 1 ft will allow the total radioactivity to grow to 20 times that of natural thorium. This will increase the allowable irradiation level to 8000 g/t at 3×10^{13} n/cm²/sec flux and zero decay time. The maximum permissible pre-Thorex decay time at 6×10^{13} n/cm²/sec flux and 8000 g/t would be 90 days. Assuming that the Th²²⁸ chain contributes 75% of the total allowable activity, semi-remote handling will also allow 150 days' less decay time for the Th²³⁴ decay. Thus, the thorium inventory charges may be decreased by a factor of 2 by using semi-remote refabrication methods.

The problem with U²³³ is different since the pre-Thorex decay time has no effect upon the activity. The only decay time required is the total time for Pa²³³ decay, which may be held back in waste catch tanks. The activity of Th²²⁸ and its daughters in stored U²³³ builds up with a half-life of 1.9 years and reaches a maximum in 10 years. Thus U²³³ products will have to be repurified remotely. A kilogram of U²³³ metal (spherical) recovered from 4000 g/t material would read 300 mr/hr at 1 ft after 3.5 months' storage. This activity level will allow only 4 hr per week working time when using the limits of 1 ft distance for hands and 2 ft for total body. In order to maintain a 40-hr/week working time for semi-remote fabrication, the distance from source to hands must be set and maintained at 3.5 ft and the total body at 5-1/3 ft. Twelve hours' semi-remote operation working time per week may be allowed if the U²³³ product has aged only 30 days since repurification and fabrication is performed at a distance of 1 ft.

The method of irradiation has no effect on the activity of Th²³⁴ in thorium irradiation if it is assumed that thorium will not be returned to a reactor until after the Th²³⁴ activity has decayed at least 100-fold.

*The term g/t for thorium irradiation corresponds to gms Mass-233 per metric for thorium.

CONCLUSIONS

The following points may be concluded from this discussion:

1. The length of the decay time (in some cases both pre-processing and post-processing) determine the type of refabrication techniques which must be employed. (Semi-remote refabrication techniques will reduce the time required for decay by at least a factor of 2.)
2. It may be more economical to store slightly enriched uranium until U^{237} has decayed to background and re-enrich in the gaseous diffusion plant than to replace the U^{235} fraction burned up with highly enriched U^{235} , shorten the decay period, and use semi-remote refabrication.
3. It is possible to minimize both the plutonium inventory charge and the cost of refabrication of fast breeder reactor Pu^{239} - U^{238} fuel elements by processing after 20-30 days' decay. The Pu^{239} fraction would then be blended with fresh U^{238} before refabrication and the separated U^{238} fraction stored as a solution to allow U^{237} to decay.
4. The permissible irradiation level for thorium may be increased by a factor of 2 and the total decay time for thorium decreased by a factor of 2 if semi-remote fabrication techniques are used.
5. Removal of U^{236} in the gaseous diffusion plant will reduce the time required for U^{237} to decay to natural background.
6. For the same plutonium production rate, the total inventory and storage costs are greater for a two-region reactor than for a one-region reactor unless U^{235} burnup of the core fuel in excess of 31% is obtained.
7. The combined costs of decay, enriching, recycle, chemical processing, and refabrication must be considered to optimize the fuel cycle.

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