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Assessment of the Radiological Impact of ²³²U and Daughters in Recycled ²³³U HTGR Fuel

John E. Till

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ENVIRONMENTAL SCIENCES DIVISION

ASSESSMENT OF THE RADIOLOGICAL IMPACT OF ^{232}U AND
DAUGHTERS IN RECYCLED ^{233}U HTGR FUEL

John E. Till

(Environmental Sciences Division
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OAK RIDGE NATIONAL LABORATORY
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ASSESSMENT OF THE RADIOLOGICAL IMPACT OF ^{232}U AND
DAUGHTERS IN RECYCLED ^{235}U HTGR FUEL

John E. Till

ABSTRACT

This paper provides an evaluation of the potential radiation exposures to man resulting from the operational releases of ^{232}U and daughters from a hypothetical HTGR fuel reprocessing plant. These estimated exposures are compared to those from the other radionuclides which could comprise the airborne releases from an HTGR fuel reprocessing plant. A hypothetical source term involving both particulate and volatile releases from an HTGR fuel reprocessing plant (450 metric tons of heavy metal annual capacity) is given assuming radionuclide inventories and decontamination factors from the literature. Radionuclides in the source term are ranked according to the magnitude of dose to total body, bone, and lungs for individuals at a distance of 1.5 miles from the stack. Although ^{232}U and daughters account for only 0.1% of the total particulate activity, the total-body dose from these radionuclides is greater in magnitude than the dose from all other radionuclides except ^{134}Cs , ^{137}Cs , and ^{90}Sr . The dose to the lungs from ^{232}U and daughters is greater than that contributed by any radionuclide other than ^{137}Cs and ^{134}Cs . Only five radionuclides, ^{90}Sr , ^{137}Cs , ^{134}Cs , ^{238}Pu and ^{244}Cm , deliver a greater dose to bone. Uranium-232 and daughters account for approximately 4% of the dose to each organ due to particulates. An analysis of the volatile releases indicates that if the decontamination factor for ^{220}Rn is between 10^3 and 10^4 , the dose commitment from ^{220}Rn

at 1.5 miles will not exceed the dose commitment from the other radionuclides released from the plant.

In view of the high radiotoxicity of the ^{232}U present in the recycled ^{233}U HTGR fuel, a comparison is made of the dose commitment to bone resulting from inhalation or ingestion of 10^{-12} g of either freshly separated HTGR fuel, LMFBR fuel, or LWR fuel. The results of this comparison indicate that the HTGR uranium fuel and LMFBR plutonium fuel are significantly more radiotoxic than LWR uranium fuel, and that the LMFBR fuel is approximately 500 times more radiotoxic than HTGR fuel for inhalation of equivalent masses of each fuel soon after release to the atmosphere. In addition it is shown that the dose commitment to bone from inhaled recycle HTGR uranium fuel increases with time after release. The dose commitment to bone from HTGR fuel may increase by a factor of approximately 7.5 due to buildup of ^{232}U daughters.

INTRODUCTION

This report focuses on three principal areas. First, an assessment is made of the radiological impact of ^{232}U and daughters relative to other radionuclides which are released to the environment from an HTGR fuel reprocessing plant. A hypothetical reprocessing plant is assumed using source terms and decontamination factors from the literature.

The second area discussed in this study reviews the effect of increasing ^{232}U concentrations on dose commitment to bone resulting from inhalation of ^{233}U HTGR fuel. Finally, this report evaluates the impact of ^{232}U daughter buildup in the environment on the dose commitment to

bone resulting from inhalation and ingestion of equal masses of LWR uranium fuel, HTGR uranium fuel, and LMFBR plutonium fuel.

The conclusion summarizes the radiological impact of ^{232}U and daughters in each area; routine releases to the environment, radiotoxicity of recycled HTGR fuel, and potential for long-term dose to man.

PRODUCTION AND DECAY OF ^{232}U IN RECYCLED ^{233}U HTGR FUEL

Uranium-232 is produced in thorium-bred ^{233}U fuel by several interactions (Fig. 1). The most important production mode originates in the ^{230}Th (n,γ) radiative capture reaction which has a cross section of 23.2 b at 2200 m/sec.² The content of ^{230}Th present in fertile ^{232}Th significantly affects the concentration of ^{232}U which is ultimately mixed with ^{233}U , ^{234}U , ^{235}U and ^{236}U in recycled HTGR fuel. Estimates of ^{230}Th in ^{232}Th range between 0.01 ppm in typical thorite-vein ore to 100 ppm, which is an upper limit for raw material thorium for commercial HTGR's.³ These levels of ^{230}Th in ^{232}Th result in ^{232}U concentrations at equilibrium recycle of 360 ppm to 1160 ppm.⁴

The ^{232}U decay chain is a member of the 4n or thorium series. In this decay chain there is no long-lived "stopping" nuclide such as exists in the ^{238}Pu , ^{239}Pu , ^{240}Pu , or ^{241}Pu chains. This property implies that the effective absorbed energy per disintegration, $\Sigma\text{EF}(\text{RBE})n$, of ^{232}U is high when compared to most other radionuclide chains. Figure 2 illustrates $\Sigma\text{EF}(\text{RBE})n$ to the bone for several chains as reported in Publication 2 of the International Commission on Radiological

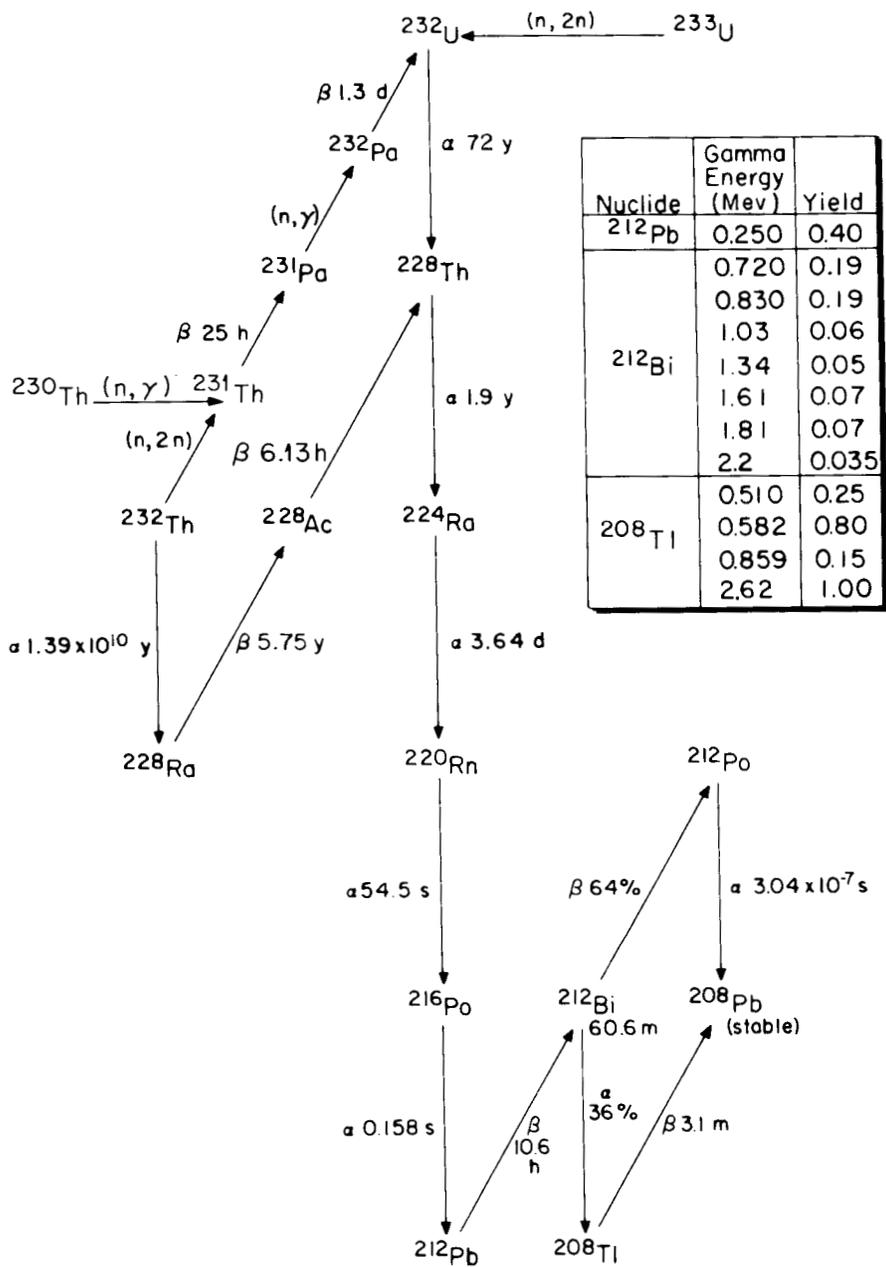


Figure 1. Production and Decay of ^{232}U .

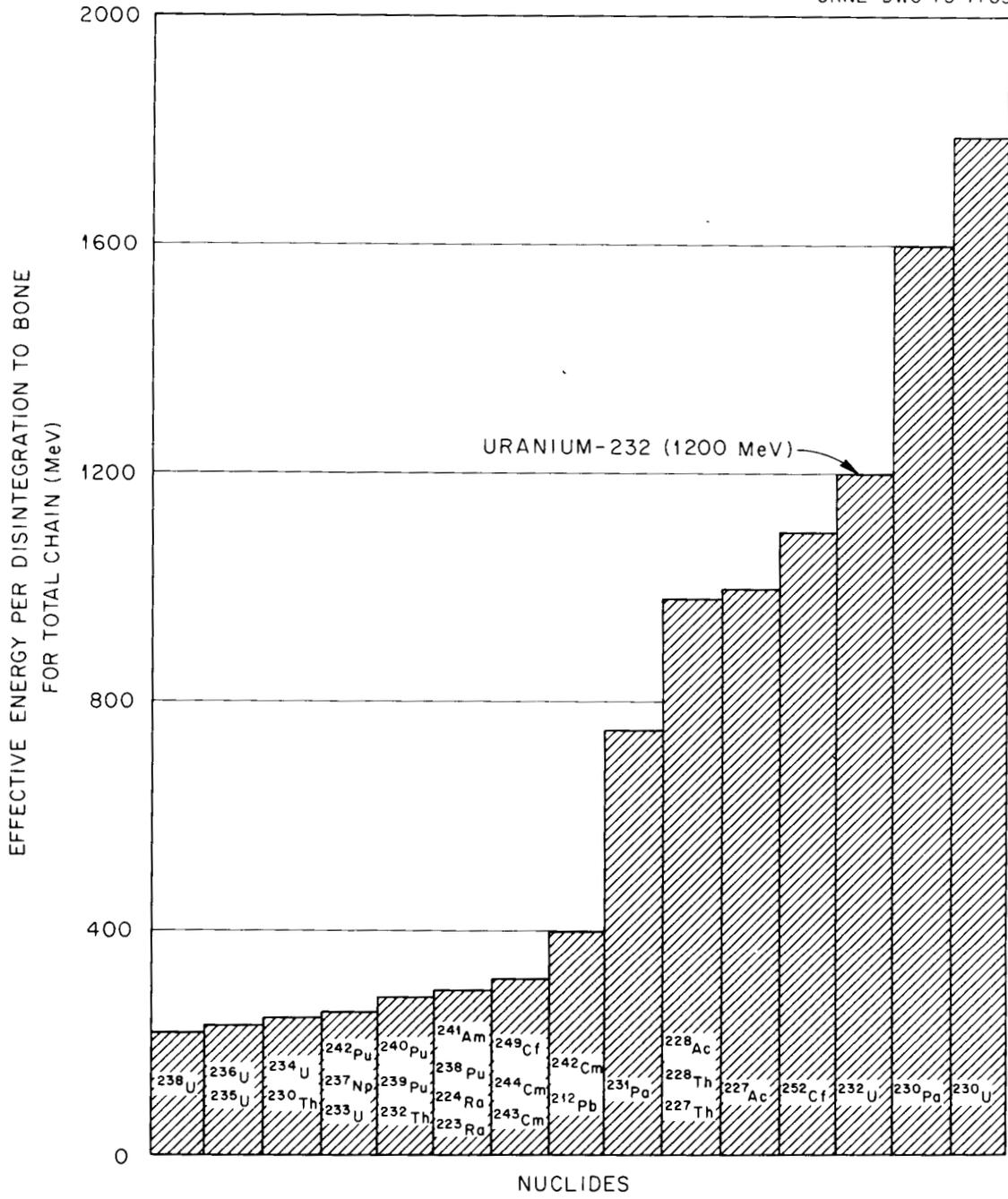


Figure 2. Effective Energy per Disintegration to Bone for Various Radionuclide Chains.

Protection.⁵ The effective absorbed energy per disintegration to bone for ^{232}U is 1200 MeV and is approximately 4 times greater than that for any of the plutonium radionuclide chains. The first two daughters of ^{232}U , ^{228}Th and ^{224}Ra , also result in relatively high values of $\Sigma\text{EF(RBE)n}$. A high value of $\Sigma\text{EF(RBE)n}$ does not necessarily imply that resulting doses from inhalation or ingestion are also high. Other factors such as biological half-time and absorption into the body through the GI tract may also affect dose.

The importance of gamma radiation emitted by the ^{232}U daughters ^{212}Pb , ^{212}Bi , and ^{208}Tl has been reviewed by Arnold⁶ for in-plant operations. Buildup of ^{232}U daughters in ^{233}U fuel following separation at the reprocessing plant creates special handling problems for the next step in the fuel cycle, fabrication into elements. These difficulties can be overcome with two possible alternatives. First, fuel may be fabricated within a short time after separation at the reprocessing plant before significant daughter buildup has occurred. Second, shielding and remote fuel handling may be incorporated into the fabrication facility design.

External exposure may also result from ^{232}U gamma emitting daughters which are released to the environment and deposited on ground surfaces. Exposure via this pathway is evaluated in the next section of this report.

RADIOLOGICAL ASSESSMENT OF ^{232}U AND DAUGHTERS

An evaluation of the radiological impact of ^{232}U and daughters is made for a hypothetical HTGR fuel reprocessing plant. This study

examines the dose from ^{232}U as related to other radionuclides released from the stack of the model plant. This assessment presents doses calculated on the basis of information derived from the literature, therefore use of these data should be limited to a relative indication of radiological impact from ^{232}U . This study is not intended as an absolute assessment of the dose to individuals near an HTGR fuel reprocessing facility.

Analysis of Dose Commitment Factors for ^{232}U and Daughters

Table 1 lists dose commitment factors for total body and bone per μCi of radionuclide inhaled or ingested. These factors were obtained from the INREM computer code.⁷ It is evident from this table that only four radionuclides are of major concern for the inhalation pathway; ^{232}U , ^{228}Th , ^{224}Ra , and ^{212}Pb . Uranium-232, ^{228}Th , and ^{224}Ra are of significant importance via the ingestion pathway. INREM dose factors do not include environmental transport and therefore should not be considered as final indicators of relative potential dose; however, a very small INREM dose factor implies that the resulting exposure will be negligible unless very large quantities of isotopes are present.

The noble gas ^{220}Rn can create a potential effluent treatment problem unique to reprocessing plants which handle recycled ^{233}U fuel. Radon-220 is produced in the reprocessing plant from the decay of ^{224}Ra almost as rapidly as it is removed by the off-gas system. Because ^{220}Rn is a gas with a short half-life (54.5 sec) the most economical

Table 1. Dose Commitment Factors for ^{232}U
and Daughters Calculated for 50 Years
with the INREM Code

Radionuclide	Inhalation Dose Commitment Factor rem/ μCi		Ingestion Dose Commitment Factor rem/ μCi	
	Total Body	Bone	Total Body	Bone
^{232}U	7.3 E 0	1.1 E 2	2.9 E-1	4.1 E 0
^{228}Th	4.2 E 1	1.3 E 3	1.6 E-2	4.9 E-1
^{224}Ra	2.7 E-1	2.1 E 0	2.0 E-1	1.6 E 0
^{216}Po	9.8 E-8	3.6 E-7	2.1 E-8	7.2 E-8
^{212}Pb	1.1 E-2	1.5 E-1	3.0 E-3	3.8 E-2
^{212}Bi	9.5 E-4	1.4 E-3	3.7 E-5	5.4 E-5
^{212}Po	6.2 E-14	3.3 E-13	1.3 E-14	6.7 E-14
^{208}Tl	9.0 E-7	6.3 E-7	8.5 E-7	6.1 E-7

effluent treatment is probably to delay its release to the environment. However some ^{220}Rn gas will still enter the atmosphere.

The dose from inhalation of ^{220}Rn primarily results from the decay of daughters - particularly ^{212}Pb (10.6 hr).^{8,9} The short half-life of ^{220}Rn implies that it will decay shortly after it is released to the environment, therefore most of the radiological impact in the vicinity of the reprocessing plant is a result of inhalation of the ^{212}Pb daughter.

Source Terms for a Model HTGR Fuel Reprocessing Plant

The radiological impact was calculated for ^{232}U and daughters in addition to fission products and transuranium elements released from a model HTGR fuel reprocessing facility. With the exception of ^{14}C , theoretical source terms were computed using curies per metric ton heavy metal (Ci/MTHM) inventories at 150 days after removal from the reactor reported by Blomeke, et al.¹⁰ The source term for ^{14}C was assumed to be 11.1 Ci/MTHM or, with a decontamination factor of 1, 5.0×10^3 Ci/year.¹¹ The model plant reprocesses 450 metric tons of heavy metal annually which is sufficient to accommodate the requirements of approximately 50 HTGR reactors.¹²

The inverse of the decontamination factor gives the fraction of the radionuclide inventory within the reprocessing plant that is released to the environment each year. Decontamination factors for this study are based upon those used in analysis of the Allied-General Nuclear Services fuel reprocessing facility near Barnwell, South

Carolina.¹³ A decontamination factor of 5×10^8 was selected for particulates with the exception of uranium and thorium isotopes. Uranium and thorium were given a decontamination factor of 1×10^8 . These elements receive more handling in the plant and therefore more opportunity exists for their release to the environment.¹⁴ Decontamination factors for volatile elements were as follows: ^3H , 1; ^{14}C , 1; ^{85}Kr , 1; ^{129}I , 20; ^{131}I , 40; and ^{220}Rn , 10^4 .

Radionuclide inventories reported by Blomeke, et al.,¹⁰ do not account for the production of daughters from precursors in the fuel after it is removed from the reactor. Therefore, a nuclide formation rate correction factor must be introduced to compensate for daughters continuously being formed. This is necessary for ^{95}Nb , ^{228}Th , ^{224}Ra , and particularly ^{220}Rn . The correction factor represents the number of curies derived annually for each curie present at any time, i.e., the decay constant in year^{-1} . For example, ^{220}Rn with a 55.6 sec half-life has a decay constant of $3.93 \times 10^5 \text{ year}^{-1}$. This implies 3.93×10^5 curies of ^{220}Rn are produced in 1 year for each curie present at steady state. The nuclide formation rate is expressed as curies formed per curie present at secular equilibrium.

Source terms for the model 450 MTHM/year HTGR reprocessing plant are listed in Tables 2 and 3 for particulates and gases, respectively. Radionuclides contributing less than $5.0 \times 10^{-4}\%$ to the final total body dose from particulates have been omitted.

Uranium-232 content is assumed to be 1200 ppm. This represents an upper limit at equilibrium recycle. Twelve hundred parts per million is approximately 5 times greater than that assumed in the report by

Table 2. Source Term for Particulate Radionuclides Released from a Model HTGR Fuel Reprocessing Plant

Radionuclide	Activity in Fuel 94,271 MWd — Aged 150 days		Nuclide Formation Rate Correction Factor (year ⁻¹)	Decontamination Factor	Release Rate pCi/sec
	Ci/MTHM	Ci/year			
⁸⁹ Sr	3.97 E 5	1.79 E 8	—	5 E 8	1.13 E 4
⁹⁰ Sr	2.89 E 5	1.30 E 8	—	5 E 8	8.24 E 3
⁹¹ Y	5.12 E 5	2.30 E 8	—	5 E 8	1.46 E 4
⁹⁵ Zr	6.54 E 5	2.94 E 8	—	5 E 8	1.87 E 4
⁹⁵ Nb	1.23 E 6	5.54 E 8	7.2 E 0	5 E 8	3.52 E 4
¹⁰³ Ru	8.76 E 4	3.94 E 7	—	5 E 8	2.50 E 3
¹⁰⁶ Ru	1.43 E 5	6.44 E 7	—	5 E 8	4.09 E 3
^{127m} Te	2.23 E 4	1.00 E 7	—	5 E 8	6.37 E 2
^{129m} Te	7.13 E 3	3.21 E 6	—	5 E 8	2.04 E 2
¹³⁴ Cs	6.88 E 5	3.10 E 8	—	5 E 8	1.97 E 4
¹³⁷ Cs	3.02 E 5	1.36 E 8	—	5 E 8	8.62 E 3
¹⁴⁴ Ce	1.78 E 6	8.01 E 8	—	5 E 8	5.07 E 4
⁵⁴ Eu	1.35 E 4	6.08 E 6	—	5 E 8	3.87 E 2
²²⁴ Ra	8.50 E 2	3.83 E 5	7.0 E 1	5 E 8	1.69 E 3
²²⁸ Th	8.50 E 2	3.83 E 5	1.4 E 0	1 E 8	1.65 E 2
²³³ Pa	1.04 E 6	4.68 E 8	—	5 E 8	2.97 E 4
²³² U	1.43 E 3	6.44 E 5	—	1 E 8	2.04 E 2
²³³ U	2.21 E 2	9.95 E 4	—	1 E 8	3.15 E 1
²³⁴ U	6.18 E 1	2.78 E 4	—	1 E 8	8.81 E 0
²³⁸ Pu	1.88 E 4	8.46 E 6	—	5 E 8	3.56 E 2
²³⁹ Pu	1.50 E 1	6.75 E 3	—	5 E 8	4.28 E-1
²⁴⁰ Pu	3.18 E 1	1.43 E 4	—	5 E 8	9.07 E-1
²⁴¹ Pu	1.07 E 4	4.82 E 6	—	5 E 8	3.05 E 2
²⁴¹ Am	1.79 E 1	8.06 E 3	—	5 E 8	5.10 E-1
²⁴³ Am	7.28 E 0	3.28 E 3	—	5 E 8	2.08 E-1
²⁴² Cm	2.17 E 3	9.77 E 5	—	5 E 8	6.18 E 1
²⁴⁴ Cm	1.64 E 3	7.38 E 5	—	5 E 8	4.69 E 1

Table 3. Source Term for Volatile Radionuclides Released from a Model HTGR Fuel Reprocessing Plant

Radionuclide	Activity in Fuel 94,271 MWd — Aged 150 days		Nuclide Formation Rate Correction Factor (year ⁻¹)	Decontamination Factor	Release Rate pCi/sec
	Ci/MTHM	Ci/year			
³ H	4.18 E 3	1.88 E 6	—	1.0 E 0	5.98 E 10
⁸⁵ Kr	6.11 E 4	2.75 E 7	—	1.0 E 0	8.73 E 11
¹²⁹ I	1.25 E-1	5.63 E 1	—	2.0 E 1	8.95 E 4
¹³¹ I	3.92 E 0	1.76 E 3	—	4.0 E 1	1.40 E 6
¹⁴ C	1.11 E 1	5.00 E 3	—	1.0 E 0	1.59 E 8
²²⁰ Rn	1.73 E 2	7.79 E 4	3.93 E 5	1.0 E 4	9.71 E 10

Blomeke. Uranium-232, ^{228}Th , and ^{224}Ra account for approximately 0.1% of the total particulate activity released. The ^{232}U daughter ^{220}Rn represents 9.4% of the total activity from volatile elements assuming a decontamination factor for ^{220}Rn of 10^4 .

Assessment of Dose at 1.5 Miles

Maximum individual doses at 1.5 miles from the stack of the model fuel reprocessing plant were calculated with the AIRDOS computer code.¹⁵ Meteorological data for the midwestern site used by Finney, et al.,¹⁶ was assumed. Stack parameters consisted of the following: height, 100 m; diameter, 3.43 m; and effluent velocity, 16.24 m/sec. AIRDOS is used to compute the 50 year dose commitment via all pathways of exposure for each radionuclide released.

Table 4 lists dose commitments in mrem to total body, bone, lungs, and kidneys for particulates. Doses are shown in decreasing order of magnitude. The only significant source for the particulates ^{228}Th and ^{224}Ra is the decay of ^{232}U ; consequently, these three radionuclides are combined into a single dose. This dose is listed in Table 4 as $^{232}\text{U}^*$. The dose contribution from ^{232}U particulate daughters other than ^{228}Th and ^{224}Ra is insignificant.

Uranium-232* contributes 10% of the dose to the lungs, 6% of the dose to the kidneys, 4% of the dose to the bone, and 0.4% of the dose to the total body. Although $^{232}\text{U}^*$ ranks high in Table 4 (3rd for lungs, 4th for total body and kidneys, 6th for bone), it is concluded that effluent control sufficient to keep doses from Sr and Cs at acceptable levels also provides satisfactory treatment for ^{232}U and particulate

Table 4. Ranking of Significant Particulate Radionuclides by Annual Dose to Reference Organs at 1.5 Miles from Stack

Total Body		Bone		Lungs		Kidneys	
Radionuclide	Dose (mrem)						
¹³⁴ Cs	1.4 E 0	⁹⁰ Sr	9.8 E 0	¹³⁷ Cs	7.0 E-1	¹³⁷ Cs	8.2 E-1
¹³⁷ Cs	9.3 E-1	¹³⁷ Cs	1.3 E 0	¹³⁴ Cs	6.2 E-1	²⁴⁴ Cm	3.0 E-1
⁹⁰ Sr	2.0 E-1	¹³⁴ Cs	1.2 E 0	²³² U*	2.0 E-1	⁹⁰ Sr	2.0 E-1
²³² U*	1.1 E-1	²³⁸ Pu	9.0 E-1	⁹⁰ Sr	2.0 E-1	²³² U*	1.0 E-1
¹⁰⁶ Ru	7.7 E-2	²⁴⁴ Cm	9.0 E-1	²⁴⁴ Cm	5.4 E-2	¹³⁴ Cs	9.9 E-2
²⁴⁴ Cm	5.4 E-2	²³² U*	6.0 E-1	¹⁵⁴ Eu	4.2 E-2	²³⁸ Pu	9.3 E-2
¹⁵⁴ Eu	4.5 E-2	⁸⁹ Sr	4.0 E-1	¹⁴⁴ Ce	3.5 E-2	¹⁰⁶ Ru	4.7 E-2
⁹⁵ Zr	3.1 E-2	¹⁰⁶ Ru	1.2 E-1	⁹⁵ Zr	3.0 E-2	¹⁵⁴ Eu	3.7 E-2
²³⁸ Pu	2.2 E-2	²³³ Pa	7.5 E-2	²³⁸ Pu	2.9 E-2	⁹⁵ Zr	2.8 E-2
²³³ Pa	2.0 E-2	¹⁵⁴ Eu	4.6 E-2	²³³ Pa	2.0 E-2	¹⁴⁴ Ce	1.8 E-2
⁹⁵ Nb	1.6 E-2	¹⁴⁴ Ce	3.9 E-2	⁹⁵ Nb	1.6 E-2	⁹⁵ Nb	1.5 E-2
¹⁴⁴ Ce	1.5 E-2	⁹⁵ Zr	3.1 E-2	⁸⁹ Sr	1.1 E-2	⁸⁹ Sr	1.0 E-2
⁸⁹ Sr	1.0 E-2	²⁴¹ Pu	1.7 E-2	¹⁰⁶ Ru	9.5 E-3	²³³ Pa	7.3 E-3
¹⁰³ Ru	1.0 E-3	⁹⁵ Nb	1.6 E-2	⁹¹ Y	1.4 E-3	^{127m} Te	4.7 E-3
²⁴¹ Pu	3.5 E-4	²⁴⁰ Pu	2.5 E-3	²⁴² Cm	1.1 E-3	¹⁰³ Ru	2.0 E-3
^{127m} Te	2.5 E-4	²³³ U	2.3 E-3	¹⁰³ Ru	1.0 E-3	^{129m} Te	1.7 E-3
²³³ U	1.8 E-4	⁹¹ Y	2.2 E-3	²³³ U	8.9 E-4	²⁴¹ Pu	1.7 E-3
^{127m} Te	9.6 E-5	^{127m} Te	1.7 E-3	^{127m} Te	2.8 E-4	²³³ U	5.6 E-4
⁹¹ Y	9.6 E-5	²⁴² Cm	1.4 E-3	²³⁴ U	2.3 E-4	²⁴⁰ Pu	2.6 E-4
²⁴² Cm	9.2 E-5	²³⁹ Pu	1.2 E-3	^{129m} Te	1.1 E-4	²⁴¹ Am	2.3 E-4
²⁴⁰ Pu	6.2 E-5	²³⁴ U	6.1 E-4	²⁴⁰ Pu	6.9 E-5	²³⁴ U	1.4 E-4
²³⁴ U	4.4 E-5	²⁴¹ Am	4.8 E-4	²⁴¹ Pu	4.1 E-5	²³⁹ Pu	1.2 E-4
²⁴¹ Am	3.7 E-5	^{129m} Te	3.6 E-4	²³⁹ Pu	3.3 E-5	²⁴³ Am	9.8 E-5
²³⁹ Pu	2.9 E-5	²⁴³ Am	2.1 E-4	²⁴¹ Am	1.9 E-5	⁹¹ Y	8.5 E-5
²⁴³ Am	2.3 E-5	¹⁰³ Ru	1.4 E-4	²⁴³ Am	1.3 E-5	²⁴² Cm	4.2 E-5
Total	2.93 E 0		1.59 E 1		1.97 E 0		1.79 E 0

NOTE: ²³²U* is the combined dose from ²³²U, ²²⁸Th, and ²²⁴Ra. Each of these radionuclides has a source term listed in Table 2; however, the only significant mechanism for the production of ²²⁸Th and ²²⁴Ra is from the decay of ²³²U. Since the purpose of this study was to evaluate the radiological impact of ²³²U and daughters in ²³³U fuel, the doses from ²³²U, ²²⁸Th, and ²²⁴Ra are combined to give a single dose.

daughters. However, the high position of $^{232}\text{U}^*$ in Table 4 indicates potential significance for exposure from these particulates. The position of $^{232}\text{U}^*$ also suggests that a more thorough evaluation of these radionuclides, especially long-term effects, may be necessary.

A breakdown of percentage of the dose from each component in $^{232}\text{U}^*$ is given in Table 5. The dose to total body and kidneys is due almost entirely to exposure from ^{232}U (50%) and ^{224}Ra (45%). Radium-224 contributes 64% of the dose to bone; ^{232}U , 20%; and ^{228}Th , 16%. The dose to lung is more evenly divided between ^{232}U (35.3%), ^{228}Th (37.5%) and ^{224}Ra (27.2%).

The radiological impact for volatile radionuclides at 1.5 miles is summarized in Table 6. Doses are listed in decreasing order of magnitude. Radon-220 accounts for only 0.3% of the dose to total body, 2% of the dose to the bone, and 3% of the dose to the lungs. In each case, more than 99.99% of the ^{220}Rn dose results from inhalation of the daughter, ^{212}Pb .

The effect of varying the decontamination factor for ^{220}Rn vs dose commitment from all radionuclides to total body and bone at 1.5 miles is demonstrated in Fig. 3. This graph shows that release of ^{220}Rn without effluent treatment would be unacceptable. Figure 3 also points out that if doses, as calculated in this report, from particulates and volatiles other than ^{220}Rn are acceptable, then a decontamination factor of approximately 10^3 would be optimum. Increasing the decontamination factor for ^{220}Rn above 10^3 has little effect on reducing the dose.

Table 7 lists the percent exposure via each pathway from ^{232}U , ^{228}Th , ^{224}Ra , ^{220}Rn , and ^{212}Pb . Surface exposure accounts for 91%,

Table 5. Percent of $^{232}\text{U}^*$ Dose from ^{232}U ,
 ^{228}Th , and ^{224}Ra to Reference Organ

Radionuclide	Percent of $^{232}\text{U}^*$ Dose (%)			
	Total Body	Bone	Lungs	Kidneys
^{232}U	50	20	35	45
^{228}Th	5	16	38	9
^{224}Ra	45	64	27	46

Table 6. Ranking of Significant Volatile Radionuclides by Annual Dose to Reference Organs at 1.5 Miles from Stack

Total Body		Bone		Lungs	
Radionuclide	Dose (mrem)	Radionuclide	Dose (mrem)	Radionuclide	Dose (mrem)
³ H	1.6 E 1	¹⁴ C	1.6 E 1	³ H	1.6 E 1
¹⁴ C	1.2 E 1	³ H	1.1 E 1	⁸⁵ Kr	1.6 E 1
⁸⁵ Kr	7.7 E 0	⁸⁵ Kr	9.4 E 0	¹⁴ C	5.2 E 0
¹²⁹ I	2.2 E 0	¹²⁹ I	2.8 E 0	²²⁰ Rn	2.1 E 0
¹³¹ I	7.2 E-1	²²⁰ Rn	2.1 E 0	¹²⁹ I	1.4 E 0
²²⁰ Rn	2.8 E-1	¹³¹ I	8.3 E-1	¹³¹ I	6.0 E-1
Total	<u>3.92 E 1</u>		<u>4.21 E 1</u>		<u>4.13 E 1</u>

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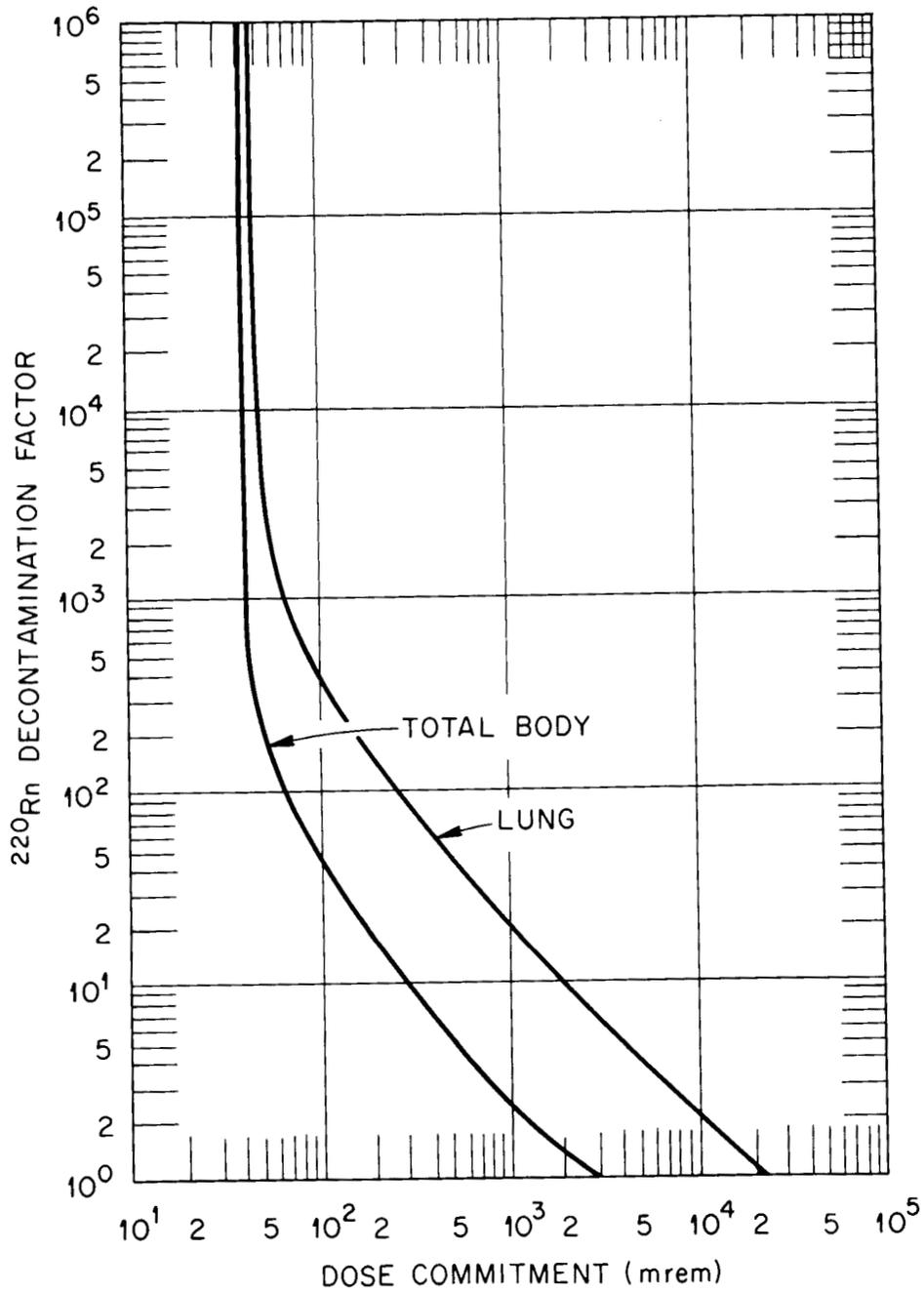


Figure 3. Dose Commitment to Total Body and Lung at 1.5 Miles from the Stack of a Model HTGR Fuel Reprocessing Plant vs Increasing ^{220}Rn Decontamination Factor.

Table 7. Percent of Dose Via Each Pathway for
 ^{232}U , ^{228}Th , ^{224}Ra , ^{220}Rn , and ^{212}Pb

Radionuclide	Pathway	Reference Organ		
		Total Body	Bone	Lungs
^{232}U	Inhalation	1.2	7.6	28.3
	Submersion in air	0.0	0.0	0.0
	Surface	91.0	43.5	65.8
	Ingestion	7.8	48.9	5.9
	Swimming	0.0	0.0	0.0
^{228}Th	Inhalation	51.0	93.3	96.4
	Submersion in air	0.0	0.0	0.0
	Surface	47.0	3.0	3.4
	Ingestion	2.0	3.6	0.2
	Swimming	0.0	0.0	0.0
^{224}Ra	Inhalation	0.4	0.4	12.0
	Submersion in air	0.0	0.0	0.0
	Surface	0.3	0.0	0.3
	Ingestion	99.3	99.6	87.7
	Swimming	0.0	0.0	0.0
^{220}Rn	Inhalation	100.0	0.0	99.8
	Submersion in air	0.0	100.0	0.2
	Surface	0.0	0.0	0.0
	Ingestion	0.0	0.0	0.0
	Swimming	0.0	0.0	0.0
^{212}Pb	Inhalation	23.0	43.0	90.9
	Submersion in air	0.3	0.1	0.0
	Surface	49.2	10.5	5.5
	Ingestion	27.5	46.4	3.6
	Swimming	0.0	0.0	0.0

44%, and 66% of the dose from ^{232}U to total body, bone, and lungs, respectively. Inhalation is the critical pathway for ^{228}Th . Ingestion is the primary exposure mode for ^{224}Ra .

EFFECT OF INCREASING ^{232}U CONCENTRATION ON DOSE COMMITMENT TO BONE

Figure 4 shows the effect of increasing the concentration of ^{232}U in recycled ^{233}U HTGR fuel on dose commitment to bone. The dose commitment is calculated assuming 10^{-12} g of fuel is inhaled. The lower curve is dose commitment for HTGR fuel at 90 days following separation. The upper curve is dose commitment for HTGR fuel at 10 years following separation. The broken horizontal line near the top of the graphs indicates the dose commitment from inhalation of 10^{-12} g of ^{233}U fuel with no ^{232}U . The vertical broken line marks the maximum anticipated ^{232}U concentration of 1200 ppm at equilibrium recycle.

Increasing the concentration of ^{232}U from 0 ppm to 1200 ppm causes dose commitment to bone to increase by a factor of approximately 35 for 90 day-old fuel and a factor of approximately 185 for fuel with ^{232}U and daughters at equilibrium. Two basic conclusions may be drawn from this figure. First, as buildup to equilibrium recycle develops and concentration of ^{232}U become greater, overall radiotoxicity of ^{233}U HTGR fuel increases significantly. Second, the buildup of ^{232}U daughters in ^{233}U fuel also increases fuel toxicity significantly for a given concentration of ^{232}U .

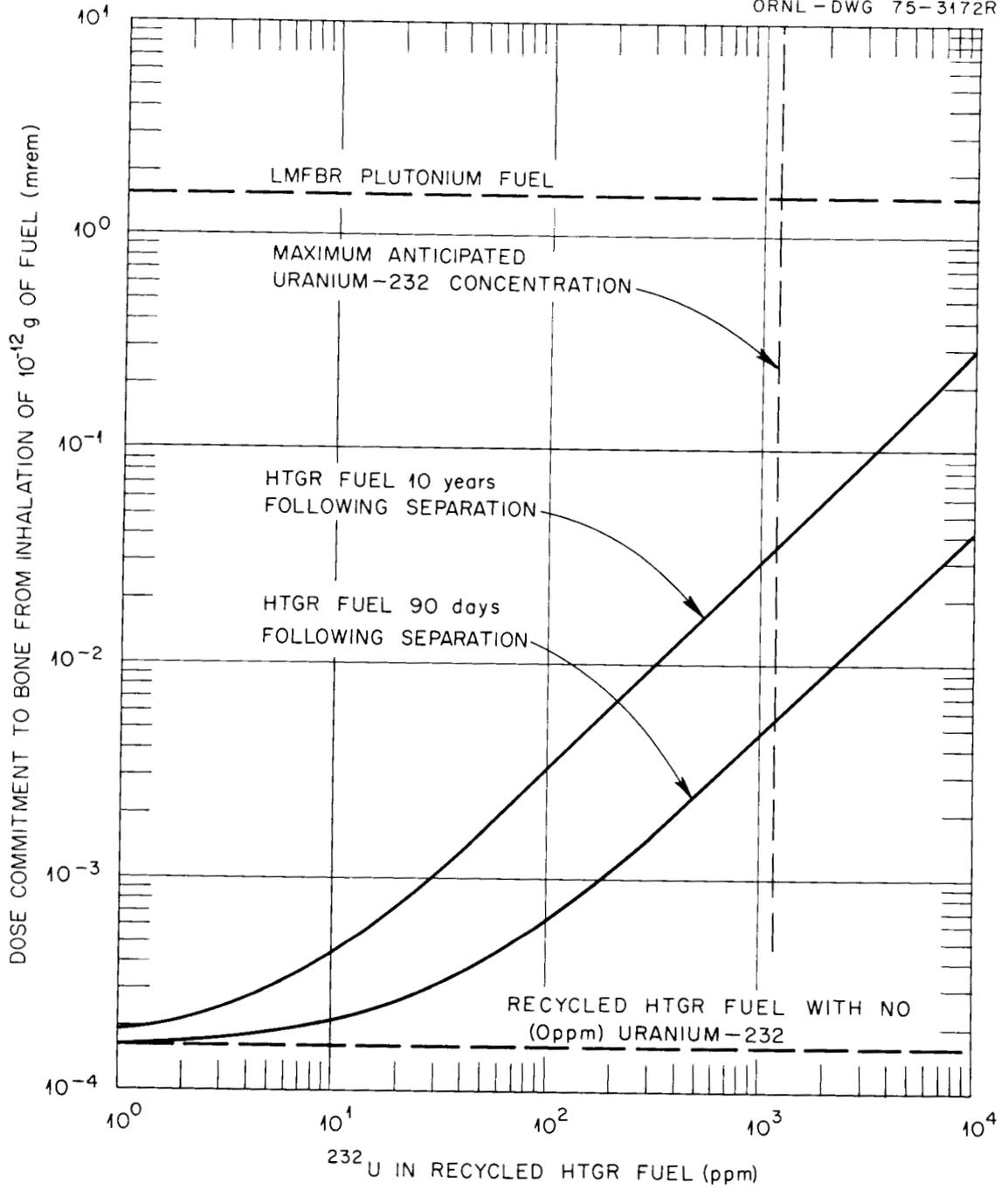


Figure 4. Effect of ^{232}U Concentration on Dose Commitment to Bone.

COMPARISON OF THE LONG-TERM RADIOACTIVE DECAY
AND DOSE COMMITMENTS FROM HTGR URANIUM, LWR
URANIUM, AND LMFBR PLUTONIUM

Buildup and Decay of Activity in HTGR
Uranium Fuel and LMFBR Plutonium Fuel

This section analyzes the radioactive decay of isotopes in freshly separated ^{233}U HTGR fuel and LMFBR plutonium fuel. The purpose is to evaluate the persistence of each fuel in the environment, to determine the effect of ^{232}U and ^{233}U daughter buildup, and to investigate the dose commitment resulting from inhalation or ingestion of equivalent masses of each fuel long after the fuel is released to the atmosphere.

Figure 5 illustrates the specific activity of recycled ^{233}U HTGR fuel and LMFBR plutonium fuel vs time after each fuel is released to the environment. Isotopic mixtures in the fuel are the same as used by Till.¹ Ninety-six percent of the initial LMFBR plutonium fuel activity results from ^{241}Pu , a beta emitter which is not as hazardous from a biological standpoint, as plutonium isotopes which decay by alpha emission. During the first 100 years, decay of ^{241}Pu ($T_{1/2} = 15$ years) causes the initial rapid decrease in LMFBR fuel activity. At this time total activity is divided between ^{238}Pu (24%), ^{239}Pu (10%), ^{240}Pu (14%), and ^{241}Pu (51%). Although the half-life of ^{241}Pu is relatively short, the longer half-lives of the daughters ^{241}Am and ^{237}Np imply that the environmental impact of ^{241}Pu will be present long after the parent has decayed. The activity of LMFBR plutonium between 1000 years and 10,000 years is due almost entirely to ^{239}Pu , ^{240}Pu , and daughters of ^{241}Pu .

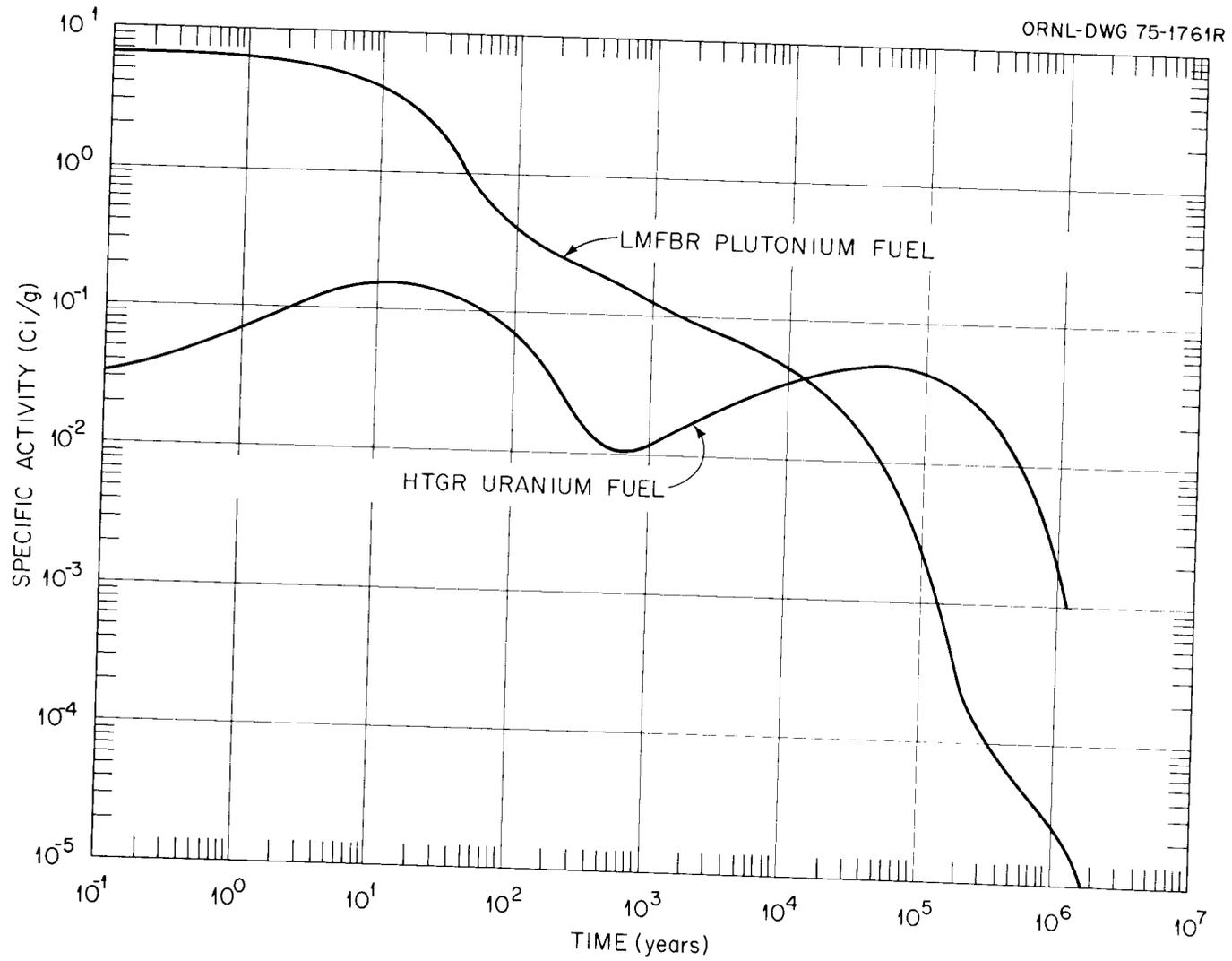


Figure 5. Buildup and Decay of Specific Activity in HTGR Uranium Fuel and LMFBR Plutonium Fuel.

The bimodal shape of the curve for ^{233}U fuel results from build-up and decay of daughters from several uranium isotopes. Table 8 lists the activity of ^{232}U daughters in 1 g of ^{233}U fuel at three intervals following separation. The ^{232}U is assumed to be present initially at 1000 ppm. The total activity of ^{233}U HTGR fuel at 90 days following separation may be broken down as follows: ^{232}U + daughters, 82.1%; ^{233}U , 14.2%; and ^{234}U , 3.7%. Uranium-232 reaches equilibrium with its daughters at approximately 10 years.

The buildup of activity caused by in-growth of ^{232}U daughters accounts for the first peak in the ^{232}U fuel curve. After approximately 300 years, activity from ^{233}U and daughters is greater than that from ^{232}U . The second peak of the curve is due to the buildup of daughters from ^{233}U and ^{234}U .

Interpretation of Fig. 5 results in two conclusions. First, assessment of radiological impact from ^{233}U fuel containing ^{232}U must consider the buildup of daughter radionuclides after the uranium parent enters the environment. The second conclusion is that ^{233}U HTGR fuel, if released to the atmosphere, will persist in the environment longer than LMFBR plutonium. Figure 5 suggests that further research should focus on the long-term implication of HTGR fuel after equilibrium is reached with ^{232}U , ^{233}U , and ^{234}U daughters. It is also important to note that the specific activity of LMFBR fuel is approximately 200 times greater than the specific activity of HTGR fuel for the interval soon after separation. However, greater specific activity above does not necessarily imply that LMFBR fuel is more hazardous.

Table 8. Activity of ^{232}U Daughters in 1 g of ^{233}U Fuel at Three Intervals Following Separation

Isotope	Activity (Ci)		
	90 Days	1 Year	10 Years
^{232}U	2.14×10^{-2}	2.12×10^{-2}	1.94×10^{-2}
^{228}Th	1.83×10^{-3}	6.51×10^{-3}	1.94×10^{-2}
^{224}Ra	1.73×10^{-3}	6.43×10^{-3}	1.94×10^{-2}
^{220}Rn	1.73×10^{-3}	6.43×10^{-3}	1.94×10^{-2}
^{216}Po	1.73×10^{-3}	6.43×10^{-3}	1.94×10^{-2}
^{212}Pb	1.73×10^{-3}	6.43×10^{-3}	1.94×10^{-2}
^{212}Bi	1.73×10^{-3}	6.43×10^{-3}	1.94×10^{-2}
^{212}Po	1.11×10^{-3}	4.11×10^{-3}	1.24×10^{-2}
^{208}Tl	6.62×10^{-4}	2.32×10^{-3}	7.00×10^{-3}

Comparison of Dose Commitment from HTGR Uranium
Fuel, LWR Uranium Fuel, and LMFBR Plutonium Fuel

A comparative analysis was performed on dose commitment to bone resulting from inhalation or ingestion of 10^{-12} g of HTGR uranium fuel, LWR uranium fuel, and LMFBR plutonium fuel. This analysis evaluates freshly separated fuel and does not consider fission products, activation products, transuranium radionuclides which have been produced, or the environmental transport of each isotope. Therefore, the resulting analysis should be regarded as estimates of fuel radiotoxicity. The data, however, indicate relative containment requirements for fuel fabrication facilities handling HTGR, LWR, or LMFBR fuel.

Figure 6 demonstrates dose commitment to bone from inhalation of 10^{-12} g of recycled fuel vs time after the fuel is released to the environment. Inhalation long after release most likely results from resuspension of radioactive materials deposited on terrestrial surfaces. Two aspects of the curve are important. First, the dose commitment to bone from LMFBR fuel is approximately 5×10^2 times greater than HTGR fuel and 3×10^7 times greater than LWR fuel when the comparison is made soon after release to the atmosphere. Second, the buildup of ^{232}U daughters increases the dose commitment from HTGR fuel by a factor of approximately 7.5 over the dose commitment at 10^{-1} year.

Dose commitment to bone from ingestion of 10^{-12} g of recycled fuel vs time after the fuel is released to the environment is shown in Fig. 7. Although plutonium and uranium are not readily taken up through the roots of vegetation, ingestion may prove to be the critical pathway in analysis of long-term exposures. Ingestion of an equivalent mass of

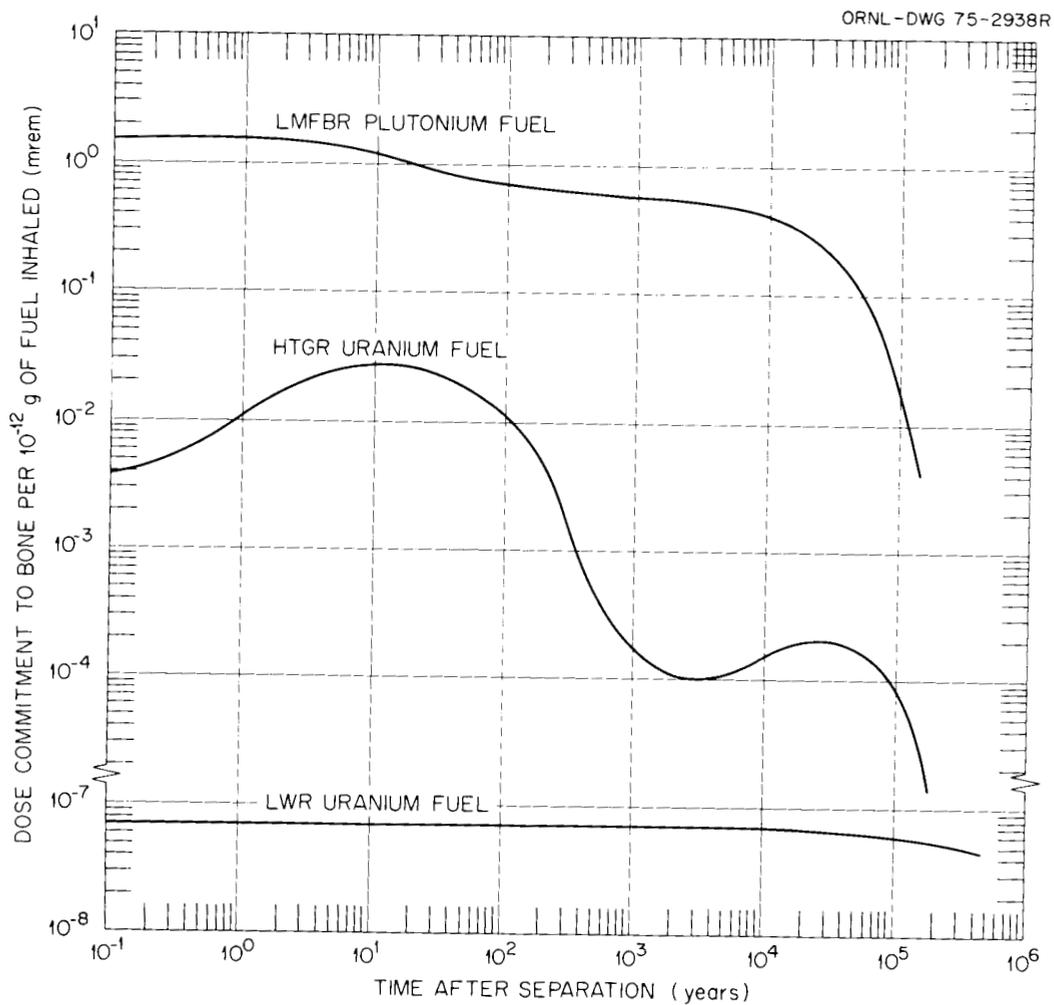


Figure 6. Dose Commitment to Bone vs Time for Inhaled LMFBR Plutonium Fuel, HTGR Uranium Fuel, and LWR Uranium Fuel.

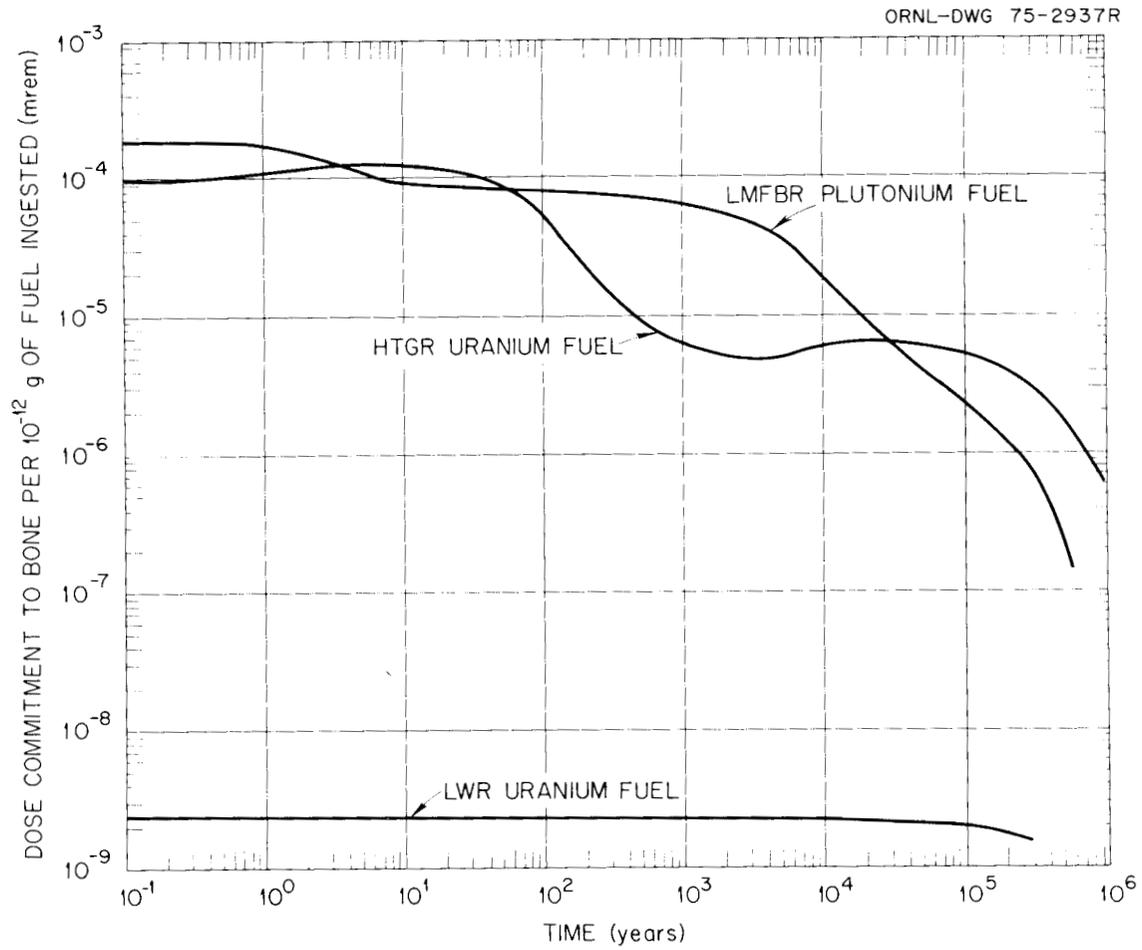


Figure 7. Dose Commitment to Bone vs Time for Ingested LMFBR Plutonium Fuel, HTGR Uranium Fuel, and LWR Uranium Fuel.

HTGR fuel and LMFBR fuel may result in similar radiological impact. The dose commitment from 10^{-12} g of LWR uranium is substantially less than either HTGR fuel or LMFBR fuel. The environmental transport of uranium, plutonium, and daughters is important in completing this assessment. Research projects to provide additional information on uptake and transport through the food chain will provide opportunity for a more in-depth study.

The data in Figs. 6 and 7 are summarized in Fig. 8 using LMFBR and HTGR fuel dose ratios. The dose ratio is calculated by dividing dose from inhalation or ingestion of 10^{-12} g of LMFBR plutonium fuel by the dose from inhalation or ingestion of an equivalent mass of HTGR uranium fuel. A dose ratio of 1 indicates equal radiotoxicity. Uranium fuel for light water reactors is not included in Fig. 8 because the toxicity relative to LMFBR plutonium and HTGR uranium is very low. The minimum inhalation dose ratio is 39; this occurs at approximately 40 years. If equivalent masses of LMFBR plutonium fuel and HTGR uranium fuel are inhaled at 40 years after they are released to the environment, the resulting dose from LMFBR fuel is 39 times greater than the dose from HTGR fuel.

SUMMARY AND CONCLUSIONS

The radiological assessment presented in this paper leads to several conclusions. As specified in this analysis, ^{232}U and daughters released to the environment as particulates from a nuclear fuel reprocessing plant do not impose significant health hazard in terms of dose to man. The high position of the dose from ^{232}U and daughters relative to other

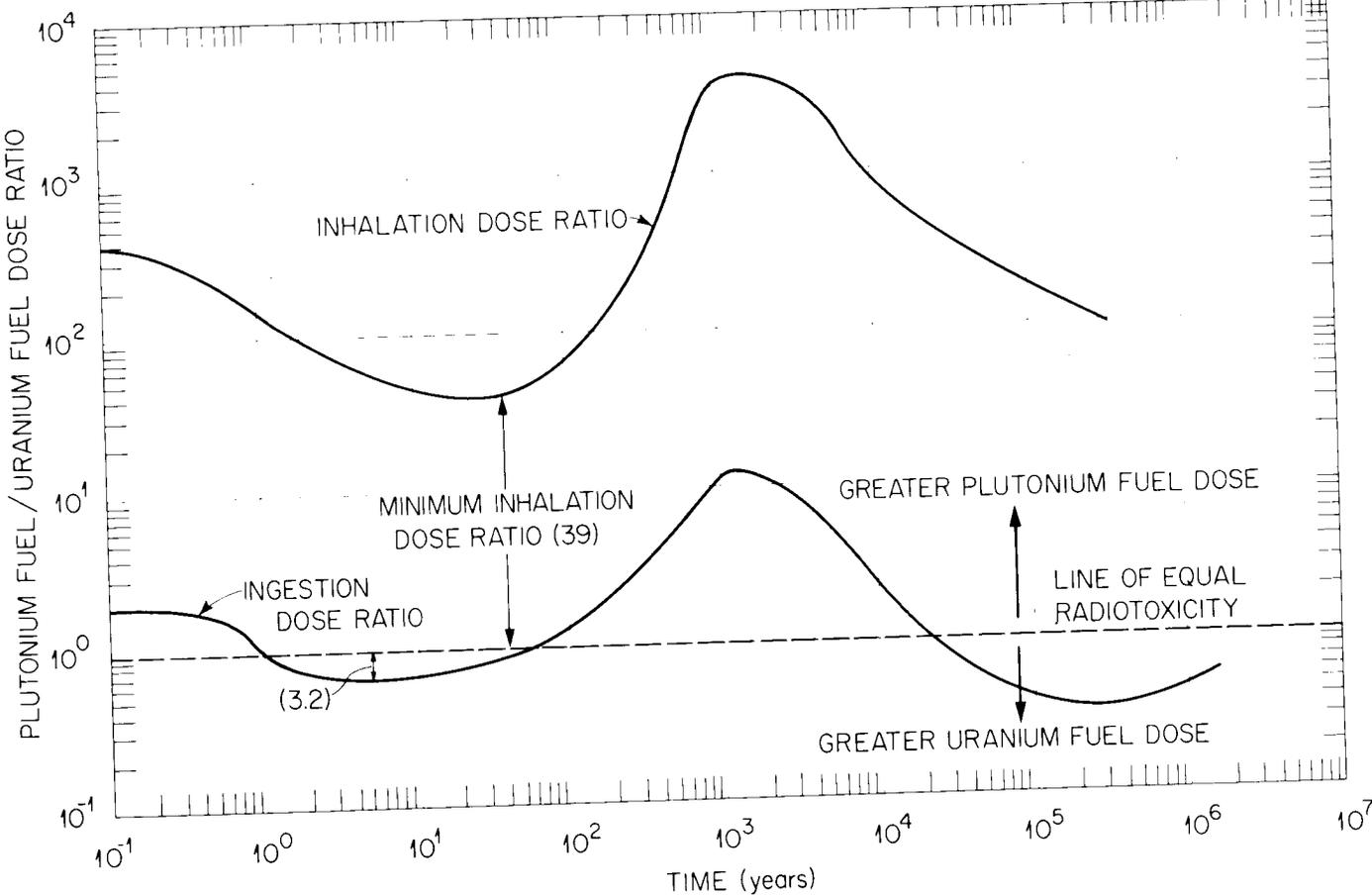


Figure 8. LMFBR Plutonium Fuel/HTGR Uranium Fuel Dose Ratios for Ingestion and Inhalation of 10-12 g of Fuel.

particulates indicates that more research may be needed to understand fully the metabolic and environmental behavior of high specific activity uranium. Release of ^{220}Rn must be delayed to permit decay before the radionuclide enters the atmosphere; an adequate ^{220}Rn decontamination factor falls within the range of 10^3 and 10^4 .

Analysis of increasing ^{232}U concentrations in ^{233}U HTGR fuel reveals that ^{232}U will substantially alter the toxicity of the fuel. Also, for a given level of ^{232}U , toxicity becomes greater with time due to daughter buildup. This greater toxicity must be considered in the design of a fuel handling facility.

Comparison of the dose commitment to bone resulting from inhalation or ingestion of equal masses of LMFBR fuel, HTGR fuel, and LWR fuel leads to several conclusions. The inhalation hazard of HTGR fuel is significantly greater than the ingestion hazard. The inhalation hazard for LMFBR fuel relative to HTGR fuel is greater by a factor of 400 for periods soon after release to the environment, decreases to a value of 39 at 40 years, and increases to approximately 4000 at 100 years, after which a slow decrease is observed. It is noted, however, that inhalation doses calculated long after the fuel has entered the environment would likely be very small.

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