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UNION CARBIDE

# Summary of the Radiological Assessment of the Fuel Cycle for a Thorium-Uranium Carbide—Fueled Fast Breeder Reactor

V. J. Tennery
E. S. Bomar
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Contract No. W-7405-eng-26

METALS AND CERAMICS DIVISION
CHEMICAL TECHNOLOGY DIVISION
HEALTH AND ENVIRONMENTAL SAFETY DIVISION

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# SUMMARY OF THE RADIOLOGICAL ASSESSMENT OF THE FUEL CYCLE FOR A THORIUM-URANIUM CARBIDE–FUELED FAST BREEDER REACTOR*

V J Tennery† E S Bomar† W D Bond† H R Meyer§
L E Morse‡ J E Till§,‖ M G Yalcintas§

## ABSTRACT

A large fraction of the potential fuel for nuclear power reactors employing fissionable materials exists as ores of thorium In addition, certain characteristics of a fuel system based on breeding of the fissionable isotope $^{233}U$ from thorium offer the possibility of a greater resistance to the diversion of fissionable material for the fabrication of nuclear weapons This report consolidates into a single source the principal content of two previous reports which assess the radiological environmental impact of mining and milling of thorium ore and of the reprocessing and refabrication of spent LBR thorium uranium carbide fuel

Environmental assessment involves two major steps The first is the selection of source terms that describe the rate and physical and chemical forms of radioactivity released from the required processes, and the second is the calculation of the resulting dose to a maximally exposed individual and the general population near the site Releases from the processing of thorium ores or thorium-based fuels are inferred from process flowsheets and plant containment design Radiological doses are calculated through use of environmental and dosimetric models

Lung is the critical organ for a maximally exposed individual located 1 6 km from a mine and mill site, receiving a dose of 35 3 millirems Radon-220 decay products are the principal contributors, $^{228}Ra$ is second in importance Thyroid receives the highest dose (6 8 millirems from $^{129}I$) for an individual located 1 km from the reprocessing plant, but tritium contributes more than 60% of the dose to total body, lungs, and other organs Cesium-137, $^{14}C$, and $^{232}U$ also make significant contributions to dose Doses resulting from operation of the refabrication plant are generally less than those due to reprocessing and are dominated by release of $^{220}Rn$, lungs receive the largest dose (3 8 millirems)

A population of 15,000, assumed to reside within 80 km of a mine and mill site near the Lemhi Pass, receives a lung dose of 0 7 man rem due primarily to $^{220}Rn$ decay products The reprocessing and refabrication plants are located in a more populous region with an impacted population of 1,000,000 Thyroid receives the highest total dose (60 1 man-rems) from the reprocessing plant releases, due primarily to $^{3}H$ and $^{129}I$ Lungs receive the highest dose (2 6 man-rems) from refabrication plant releases, due to $^{220}Rn$ decay products

A need is established for additional information describing thorium ores and for site-specific meteorology for the mountainous areas in the western U S where large deposits of thorium ores are found

## 1 INTRODUCTION

This report summarizes the information contained in two recent publications which assess the radiological environmental impact resulting from the mining and milling of thorium ore[1] and from the reprocessing and refabrication of thorium-uranium carbide fuel[2] irradiated in a fast breeder reactor (FBR) The characteristics of thorium-uranium nuclear fuel systems are being examined for comparison with uranium-plutonium fuel systems as to their ability to breed fissionable material as well as comparing their resistance to diversion of nuclear material The potential resistance of thorium-uranium fuel materials to increasing

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proliferation of nuclear weapons is a subject of particular interest. Carbide fuels have several potential performance advantages in FBRs compared with oxide fuel, and this work is specifically concerned with carbide fuel in the $^{232}Th/^{233}U$ system.

The amount of thorium used in the United States is small and is currently obtained as a secondary product of the mining of minerals containing titanium and the rare earths. Power generation on a commercial scale using nuclear reactors fueled with thorium-based fuels will require a very large increase in the amount of thorium mined plus the reprocessing and refabrication of the spent fuel to recover the $^{233}U$ bred during irradiation of thorium in the reactor.

The information summarized here extends that resulting from other studies[3, 4, 5] incorporates recently improved dose assessment methodology, and identifies the radioisotopes which are the principal contributors to the radiological environmental impact.

## 2. METHODOLOGY

Prediction of the impact of future ore mining and milling or spent fuel reprocessing operations in the absence of prior, full-scale commercial experience requires the preparation of a model of these operations, using flowsheets and mathematical formulas which permit the calculation of source terms (rates of release of various isotopes). Radiological doses may then be calculated by simulating the resulting interactions with the environment through contact or assimilation by vegetation or animals and, in particular, man.

Flowsheets were prepared to describe the movement of material through the various operations at a mine and mill complex and through reprocessing and refabrication plants. Radioactivity is released from each of these processes in the form of particles resulting from mechanical operations such as blasting or grinding. There is also a release of gases during chemical dissolution of spent fuel. Liquids, containing radioactivity, will also be released from the mill and the fuel refabrication plant. The source terms describing liquid, particulate, and gaseous release rates plus meteorology information were used as input to the AIRDOS-II computer code,[6] which predicts the airborne distribution and deposition of the released radioactivity. Doses received by both the "fence-post man," who is assumed to be 1.6 km from the facility, and by the general population contained within an 80-km radius of the facility are then calculated. The "fence-post man" is a hypothetical individual who lives just beyond the facility exclusion fence. Consumption of only locally grown food products was assumed for this individual; this is a conservative or dose-increasing assumption. Thus the dose calculated for this individual should be the maximum for a member of the general population. Population doses are based on consumption of both local and "imported" food products. The AIRDOS-II computer code, supplemented by the INREM-II computer code, also accounts for the differences in chemical and physical behavior of the various isotopes released, predicts the inhalation and ingestion of these isotopes by individuals at various distances from the facility, incorporates estimates of distributions within and elimination rates from the body, then uses such information to calculate doses to man.

There are a number of potential pathways of exposure to man from radioactive effluents released to the environment; these are illustrated schematically in Fig. 1. Radionuclides enter the environment from a facility via either liquid or atmospheric transport modes. They reach man through one or more collectors, with possible reconcentration by animals, crops, and aquatic biota. Pathways to human exposure include inhalation, ingestion, immersion in air, contamination of land surfaces, and submersion in water. An adaptation of the task group lung model[7] is utilized to describe the dynamics of radionuclide retention within the respiratory tract. A catenary GI tract model based on the transit times recommended by Eve[8] simulates retention in the GI tract. Retention in other organs is represented by multicompartment models consisting of series of decaying exponential terms. Detailed discussion of these models and assumptions is available in a report by Dunning et al.[9]

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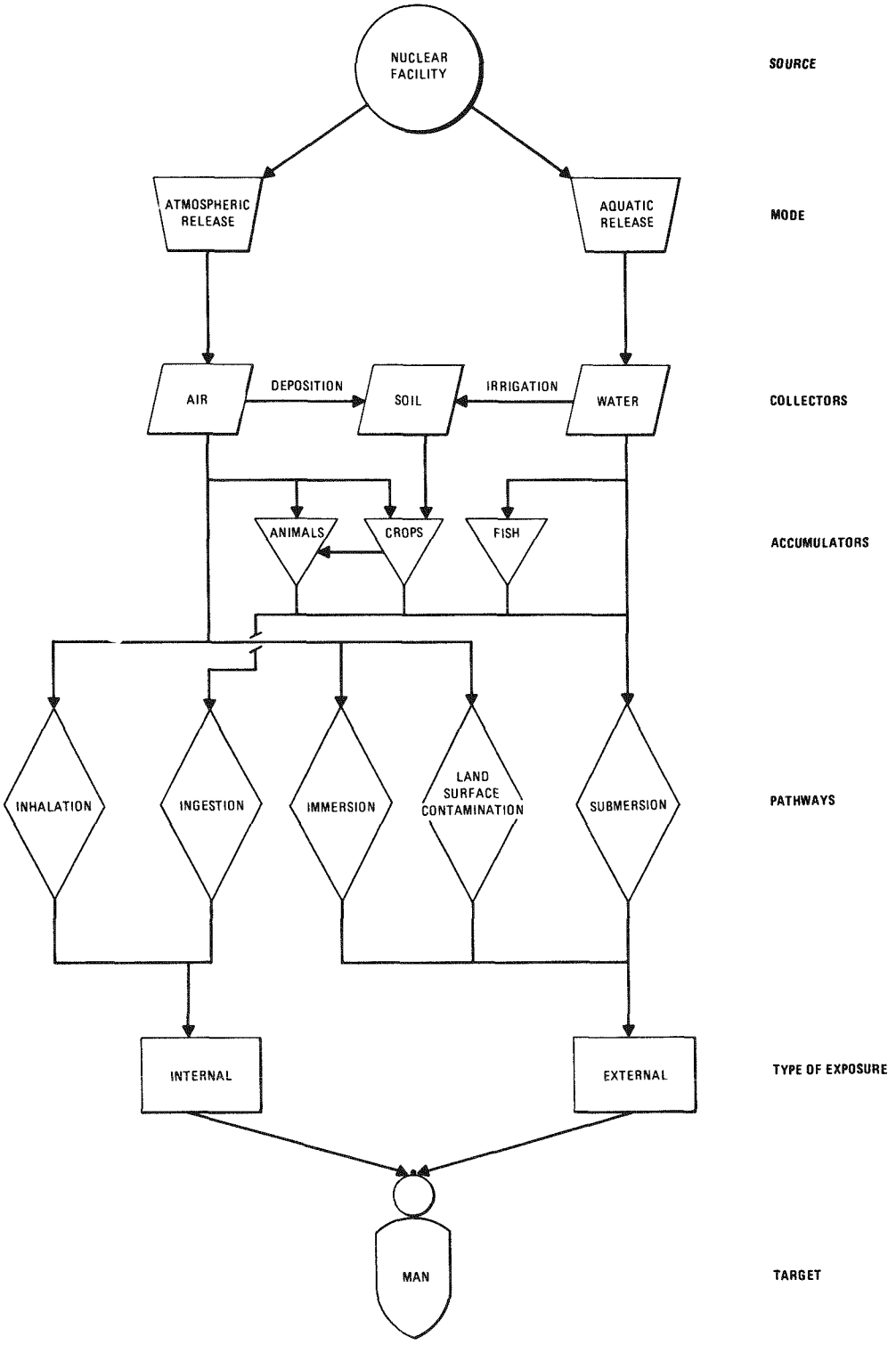


Fig. 1. Exposure pathways to man.

Radiological impact is calculated as the 50-year dose commitment to individuals or populations in millirems or man-rems per year of facility operation. The dose commitment is calculated for a given intake of radionuclide and is defined as the total dose to a specific organ, resulting from a one-year intake or exposure, which will accrue during the remaining lifetime of the individual. Estimates of radiation dose to the total body and to major organs are considered for all pathways of internal exposure and are based on parameters applicable to the average adult. Population dose estimates are the sums of total-body or specific-organ doses to all individuals assumed living within 80 km of the facility.

## 3. RADIOCHARACTERISTICS OF THORIUM AND DAUGHTERS

Several characteristics of the $^{232}$Th decay chain, shown in Fig. 2, result in noteworthy effects during and after processing of the thorium in ore or spent fuel. The decay products or daughters of $^{232}$Th are a

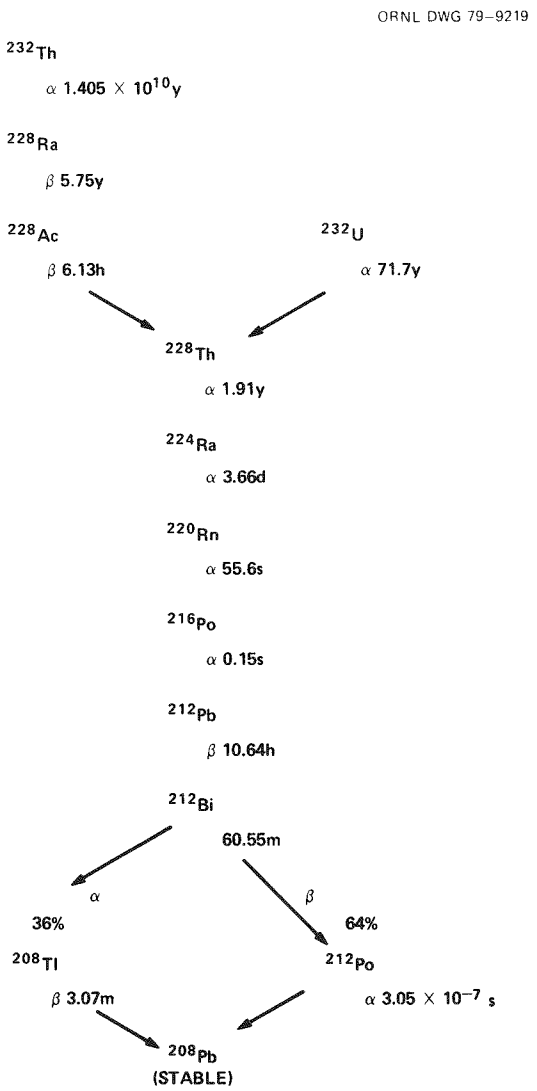


Fig. 2. Decay of $^{232}$Th and $^{232}$U.

mixture of alpha and beta emitters with some accompanying gamma radiation. The half-life of $^{232}Th$ (1.4E 10 years) is extremely long compared with its daughters; therefore, mass concentrations of the daughters in secular equilibrium with $^{232}Th$ are very low. One of the daughters, $^{220}Rn$, is a chemically inert gas with a short half-life (56 sec). Gaseous $^{220}Rn$ is rapidly converted to other isotopes that are solid under normal conditions; therefore, a model which realistically predicts the behavior of any $^{220}Rn$ released to the environment must take this physical change into account.

Examination of the decay chain shows that the radioactivity of thorium chemically separated from its nonthorium daughters will be strongly influenced by the relatively short-lived $^{228}Th$, accompanied by a gradual buildup of $^{228}Ra$. Concurrently, the radioactivity of material from which most of the thorium has been removed will be controlled by the decay of $^{228}Ra$ as its concentration gradually declines to a level sustained by residual $^{232}Th$.

## 4. MINING, MILLING, AND REFINING OF THORIUM

### 4.1 Description of Model Mine and Mill

The locations and types of thorium deposits found in the United States are shown in Fig. 3 and in Tables 1 and 2. A substantial increase in demand for thorium would probably result in mining of the vein deposits located in the western United States. Two sites, one in the Lemhi Pass district of Idaho and Montana and the other in the Wet Mountains of Colorado, were selected for site-specific analysis in this study.

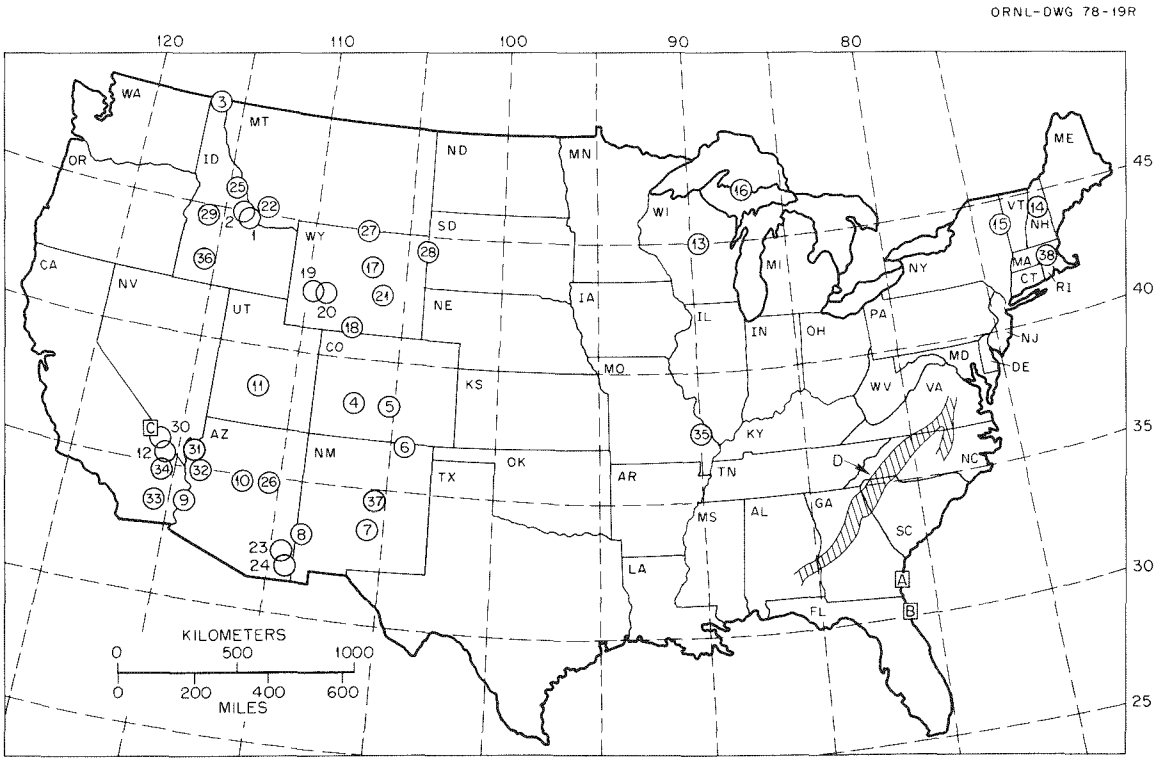


**Fig. 3. Thorium resources in the United States.** (Also see Tables 1 and 2 for legend.)

**Table 1. Vein thorium deposits – United States**

| Location (see map in Fig. 3) | County, state | Latitude (°N) | Longitude (°W) | Number of samples | Max vein length (m) | Max vein thickness (m) | Thorium content (%) | Population within 80 km (1970) | Ref. |
|---|---|---|---|---|---|---|---|---|---|
| 1. Lemhi Pass | Lemhi, ID<br>Beaver, MT | 44.93 | 113.5 | 200+ | $1.2 \times 10^3$ | 9 | 0.001–16.3 | 14,242 | 2–5 |
| 2. Diamond Creek | Lemhi, ID | 45 | 114 | 9 | $1.7 \times 10^2$ | 7.5 | 0.02–1.71 | 7,364 | 3 |
| 3. Hall Mt. | Boundary, ID | 48.99 | 116.38 | 14 | $2.1 \times 10^2$ | 4 | 0.01–21 | 15,359 | 6 |
| 4. Powderhorn | Gunnison, CO | 38.25 | 107 | 200+ | $1.1 \times 10^3$ | 5.5 | 0.01–4.3 | 32,192 | 7 |
| 5. Wet Mts. | Custer, CO | 38.25 | 105.35 | 400+ | $1.5 \times 10^3$ | 15 | 0.02–12.5 | 262,144 | 8,9 |
| 6. Laughlin Pk. | Colfax, NM | 36.75 | 104.25 | 10 | $2.4 \times 10^2$ | 6.1 | 0.05–0.82 | 27,615 | 2 |
| 7. Capitan Mts. | Lincoln, NM | 33.5 | 105.78 | 12 | 46 | 2.4 | 0.01–1.12 | 47,668 | 10 |
| 8. Gold Hill | Grant, NM | 33 | 109 | 2 | 12 | | 0.05–0.72 | 39,900 | 2 |
| 9. Quartzite | Yuma, AZ | 33.75 | 114.25 | 2 | 15 | 2.4 | 0.027–0.27 | 22,613 | 2 |
| 10. Cottonwood | Yavapai, AZ | 34.75 | 112 | 1 | 30 | 18 | 0.013–0.91 | 64,769 | 2 |
| 11. Monroe Canyon | Seiver, UT | 38.58 | 112 | 1 | 7.6 | 15 | 0.18–0.29 | 19,868 | 2 |
| 12. Mountain Pass | San Bernardino, CA | 35.46 | 115.5 | 18 | $4.9 \times 10^2$ | 3 | 0.02–4.9 | 30,321 | 11 |
| 13. Wausau | Marathon, WI | 45 | 89.5 | 20 | $4.6 \times 10^2$ | 0.5 | | 338,408 | 12 |

Table 2. Other thorium deposits – United States

| Location (see map in Fig 3) | County, state | Latitude (°N) | Longitude (°W) | Thorium content (ppm) | Population within 80 km (1970) | Ref |
|---|---|---|---|---|---|---|
| 14 Conway | Conway, NH | 44 00 | 71 16 | 64 | | 29 |
| 15 Mineville | Essex, NY | 44 16 | 73 58 | 100–3800 | | 30 |
| 16 Palmer | Marquette, MI | 46 5 | 87 5 | 50,000 | | 31 |
| 17 Owl Creek | Hot Spring, WY | 43 48 | 105 50 | 134 | 30,292 | 13,14 |
| 18 Rawlings uplift | Carbon, WY | 41 78 | 107 13 | 146 | 13,201 | 13,14 |
| 19 Wind River | Fremont, WY | 43 5 | 109 5 | 366 | 31,648 | 13,14 |
| 20 Wind River | Fremont, WY | 43 5 | 109 6 | 66 | 31,648 | 13,14 |
| 21 Seminoe | Natrona, WY | 42 47 | 106 75 | 194–273 | 51,995 | 13,14 |
| 22 Deer Creek | MT | 45 2 | 112 5 | | | 30 |
| 23 Blue Mt | Greenlee, AZ | 32 55 | 109 20 | 40 | 30,481 | 13,15 |
| 24 Dos Cabesas | Cochise, AZ | 32 2 | 109 42 | 19 | 30,098 | 13,15 |
| 25 Mineral Hill | Lemhi, ID | 45 6 | 114 9 | | | 13,15 |
| 26 Diamond Rim | Gila, AZ | 34 25 | 111 08 | 24 | 10,416 | 13,15 |
| 27 Little Big Horn | Big Horn, WY | 44 66 | 106,95 | | | |
| 28 Bear Lodge | Crook, WY | 44 5 | 104 33 | 400–2500 | | 30 |
| 29 Idaho | Idaho, ID | 46 | 115 | 200 | | 32 |
| 30 McCullough Mt | Clark, NV | 36 | 116 | 55–283 | 145,059 | 13,15 |
| 31 Black Mts | Mohave, AZ | 35 5 | 114 5 | 180–253 | 137,958 | 13,15 |
| 32 S Peacock Mts | Mohave, AZ | 35 | 114 | 37–153 | 137,958 | 13,15 |
| 33 Big Maria Mts | Riverside, CA | 33 5 | 116 | 29–146 | 136,470 | 13,15 |
| 34 Marble Mts | San Bernardino, CA | 35 | 116 | 75–148 | 136,470 | 13,15 |
| 35 St Francois Mts | St Francois, MO | 37 5 | 90 | 47 | 320,378 | 13,17 |
| 36 Idaho Batholith | Boise, ID | 44 0 | 115 90 | 100 | | 32 |
| 37 Gallinas | Lincoln, NM | 34 15 | 105 63 | | | 33 |
| 38 Worcester | Worcester, MA | 42 25 | 71 75 | 300 | | 30 |
| A Georgia | Charlton, GA | 32 | 81 6 | <1000 | | 34 |
| B Florida | Nassau, FL | 30 2 | 81 3 | <1000 | | 34 |
| C California | San Bernardino, CA | 36 | 117 | 200–5000 | | 30 |
| D Piedmont District | VA, NC, SC, AL | 32–38 | 78–87 | [5 67%] | | 30 |

Vein locations are found by using sensitive instruments to detect the radioactivity of thorium daughters Bulldozers have been used to remove the overburden and expose the veins An aerial view of several prospects in the Lemhi Pass district is shown in Fig 4

The principal components of a mine and mill complex include open-pit mine, ore storage pile, mill, tailings impoundment, and thorium refinery The ore pile will contain a 60-day reserve to allow operation of the mill at its 1600-Mg/day capacity during those winter months when the weather stops mining activities at the higher elevations Several characteristics of the mine, mill, and refinery are listed in Table 3

After removing the overburden at the mine site, the exposed vein(s) of thorium-bearing ore would be broken up by bulldozers, dozer-rippers, and selective blasting Front-end loaders are used to load trucks for transport to the ore storage pile at the mill The sequence for processing ore at the mill and recovery of the thorium is shown in Fig 5 The flowsheet is based on experiments performed by the U S Bureau of Mines [10, 11] The crude $Th(CO_3)_2$ product from the mill is purified to produce reactor-grade $Th(NO_3)_4 \cdot 4H_2O$, using a flowsheet[12] developed for processing irradiated $ThO_2$ and described in Fig 6 Mass and volumetric flow rates for key process streams in the mill and refinery are given in Table 4 The leached and washed solid wastes from the mill are transferred as a slurry to a tailings pond where the solids separate out and are eventually allowed to dry About 9% of the thorium originally contained in the ore and essentially all of the daughter activity are deposited in the tailings pond Some $^{220}Rn$ gas is released at the mine and


**Fig. 4. Lemhi Pass exploratory trenches.**

**Table 3 Characteristics of the open pit thorium mine and the model thorium mill and refinery**

| | | |
|---|---|---|
| **Mine** | | |
| Approximate total area ($m^2$) | | 4 9E4[a] |
| Exposed thorium bearing vein(s) ($m^2$) | | 1 2E4 |
| Ore production (Mg/day) | | 1600 |
| Average thorium content (% $ThO_2$ equivalent) | | 0 5 |
| Water drainage ($m^3$/day) | | 6 8E2 |
| Average depth (m) | | 23 |
| **Mill and refinery** | | |
| Ore capacity (Mg/day) | | 1600 |
| Days of operation annually | | 300 |
| Thorium recovery efficiency (%) | | |
| Mill | | 91 |
| Refinery | | 99 5 |
| $Th(NO_3)_4$ 4H O production (Gg/year) | | 4 5 |
| Water required ($m^3$/day) | | 2 4F3 |
| Ore pile (m) | | 100 × 32 × 15 |
| (Gg) | | 96 |
| Air discharge from complex ($m^3$/sec) | | 11 3 |
| Filter losses (%) | | |
| Crusher dust | | 0 7 |
| $Th(NO_3)_4$ $4H_2O$ product line (bags plus HFPA) | | 0 05 |
| **Tailings impoundment** | | |
| | **Montana** | **Colorado** |
| Average area during 20 year mill life ($m^2$) | | |
| Dry beach | 4E3 | 36E3 |
| Pond | 5 7E5 | 4 9E5 |
| Average area exposed during post mill life ($m^2$) | | |
| Dry beach | 4E3 | 4E3 |
| Pond | 3 8E5 | 2 7E5 |

[a] 4 9E4 4 9 × $10^4$

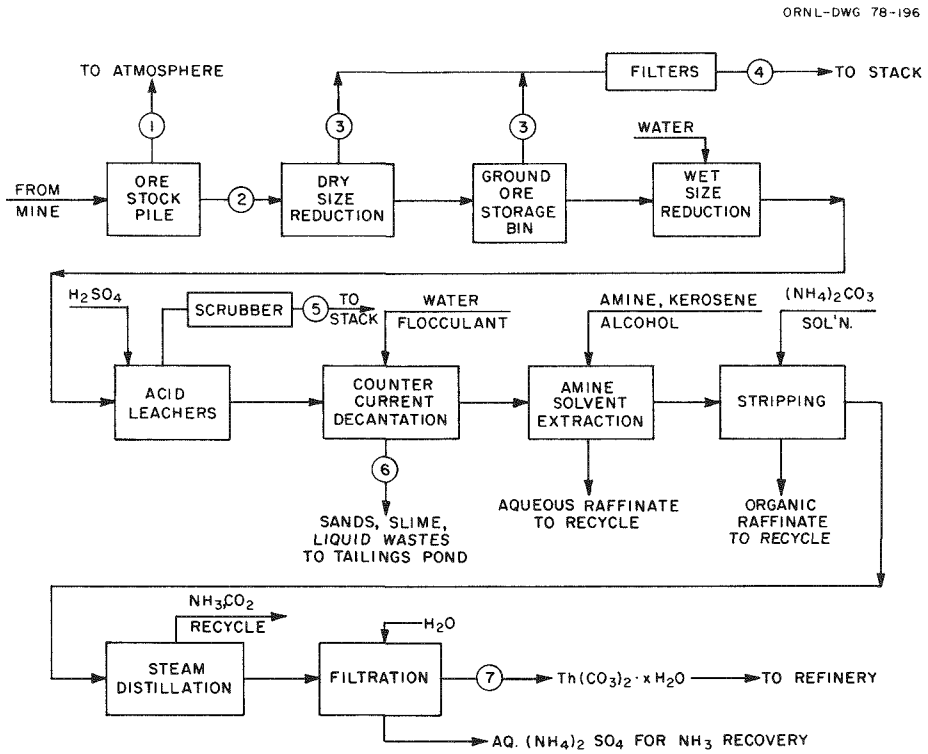


Fig. 5. Conceptual thorium milling, flow diagram.

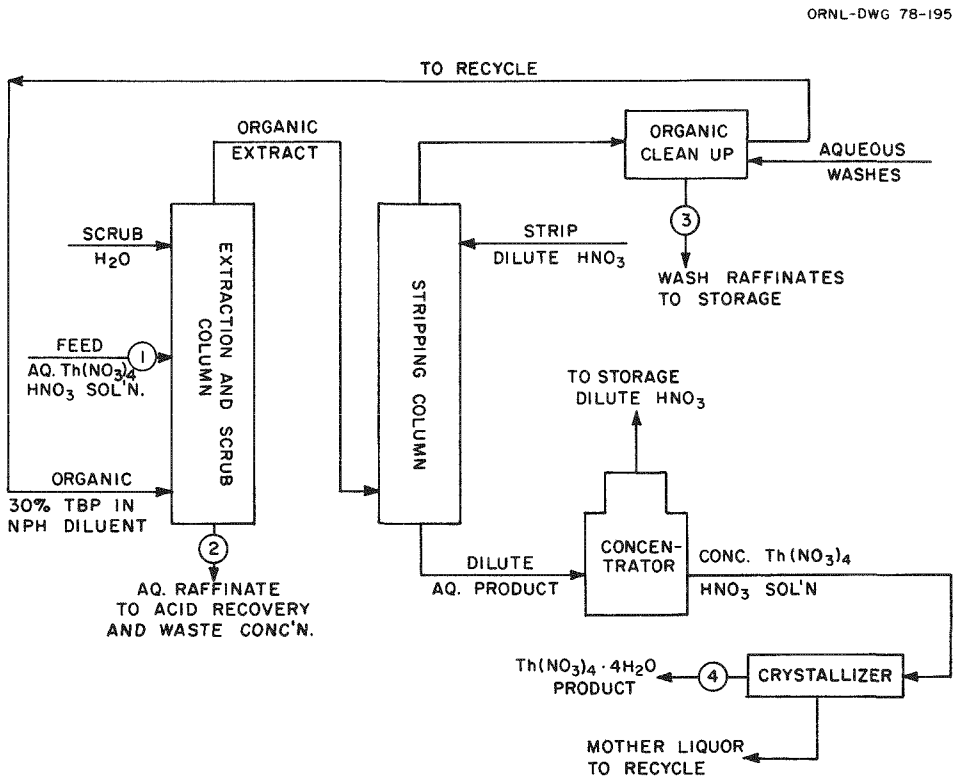


Fig. 6. Conceptual thorium refining, flow diagram.

**Table 4. Mass and volume flow rates for principal process streams of the model mill and refinery**

| | Mill | | | Refinery | | |
|---|---|---|---|---|---|---|
| | Feed[a] | Product | Tailings | Feed | ThNT product | Aqueous wastes |
| Thorium | | | | | | |
| kg/sec | 8.14E-2[b] | 7.41E-2 | 7.33E-3 | 7.41E-2 | 7.37E-2 | 3.70E-3 |
| (kg/day) | (7.03E+3) | (6.40E+3) | (6.33E+2) | (6.40E+3) | (6.37E+3) | (3.20E+2) |
| Volume[c] | | | | | | |
| $m^3$/sec | 3.47E-2 | *d* | 3.47E-2 | 4.06E-4 | *d* | 3.11E-3 |
| ($ft^3$/day) | (1.06E+5) | *d* | (1.06E+5) | (1.24E+3) | *d* | (9.50E+3) |

[a]Feed rate of ore is 18.5 kg/sec (1.6 Gg/day).
[b]Read as $8.14 \times 10^{-2}$.
[c]Includes both process liquids and solids.
[d]Not determined.

during crushing and acid leaching of the ore in the mill; $^{220}Rn$ is continuously regenerated from decay of $^{224}Ra$.

## 4.2 Source Terms from Mining, Milling, and Refining

### 4.2.1 Mining

Radioactivity will be released to the environment as gas and dust as a result of mining operations. As in ERDA-1541 it is assumed that 3 acres of thorium-bearing ore containing the equivalent of 0.5% $ThO_2$ are exposed in one or more pits being worked to supply 1600 Mg/day of ore to the mill.[13] In the absence of information on release of $^{220}Rn$ from thorium ore, a mechanism similar to that for release of $^{222}Rn$ from uranium ore was assumed.[13, 14] An expression for $^{220}Rn$ flux, $J_0$, was developed:

$$J_0 = 455\rho(k/p)^{1/2}, \quad (1)$$

where

$\rho$ = bulk density of the ore, $kg/m^3$,

$k$ = effective diffusion coefficient for $^{220}Rn$ in ore, m/sec,

$p$ = void fraction in ore.

A flux rate was calculated for $^{220}Rn$ of 1.32 $kBq/m^2 \cdot sec$ or a total of 16 MBq/sec (0.43 mCi/sec) from 3700 $m^2$ (3 acres) of exposed ore.[1]

A number of activities at the mine will inject dust into the air. These include dozing, selective blasting, loading ore into trucks, and movement of vehicles. Information on which to make a quantitative determination of the amount of fugitive dust generated is limited. The dust raised by vehicular traffic was calculated using the results of a study done for the Environmental Protection Agency.[15] The amount of dust generated by other mining activities was also estimated. The expected dampness of the ore plus routine sprin-

kling of the mine area with water controls the amount of dust released The estimated radioactivity contained in the fugitive dust is shown in Table 5

### 4 2 2 Milling and refining

Loading of ore and movement from the ore storage pile to the mill will generate airborne particulate contamination The method used for calculation of the amount of dust suspended was as above [15] The amount of radioactivity contained in dust from ore handling at the mill is shown in Table 6

**Table 5. Radioactivity contained in dust generated by mining operations**

| Isotope | Radioactivity [Bq (pCi)][a] | |
|---|---|---|
| | Per g of ore | Per 13 kg (29 lb) dust[b]/day |
| $^{232}$Th | 18 (4 8E2)[c] | 1 2E5 (3 2E6) |
| $^{228}$Ra | 18 (4 8E2) | 1 2E5 (3 2E6) |
| $^{228}$Ac | 18 (4 8E2) | 1 2E5 (3 2E6) |
| $^{228}$Th | 18 (4 8E2) | 1 2E5 (3 2E6) |
| $^{224}$Ra | 18 (4 8E2) | 1 2E5 (3 2E6) |
| $^{220}$Rn | 18 (4 8E2) | 1 2E5 (3 2E6) |
| $^{216}$Po | 18 (4 8E2) | 1 2E5 (3 2E6) |
| $^{212}$Pb | 18 (4 8E2) | 1 2E5 (3 2E6) |
| $^{212}$Bi | 18 (4 8E2) | 1 2E5 (3 2E6) |
| $^{212}$Po | 11 (3 1E2) | 7 5E4 (2 0E6) |
| $^{208}$Tl | 6 (1 7E2) | 4 2E4 (1 1E6) |

[a] 1 Bq = 27 pCi

[b] Concentration of Th in fugitive dust is diluted by dust containing no Th to about half that of the ore

[c] 4 8E2 = 4 8 × $10^2$

**Table 6 Estimated source terms for operation and closing of the model mill**

| Nuclide | Source term (Bq/sec)[a] | | | | | |
|---|---|---|---|---|---|---|
| | Ore handling | Mill + refinery | Tailings beach and pond | | Covering dry tailings | |
| | | | Montana | Colorado | Montana | Colorado |
| $^{232}$Th | 0 52 | 0 18 | 0 037 | 0 33 | 0 37 | 0 37 |
| $^{228}$Ra | 0 52 | 0 18 | 0 33 | 3 0 | 3 3 | 3 3 |
| $^{228}$Ac | 0 52 | 0 18 | 0 33 | 3 0 | 3 3 | 3 3 |
| $^{228}$Th | 0 52 | 0 18 | 0 26 | 2 2 | 2 4 | 2 4 |
| $^{224}$Ra | 0 52 | 0 18 | 0 26 | 2 2 | 2 4 | 2 4 |
| $^{216}$Po | 0 52 | 0 18 | 0 26 | 2 2 | 2 4 | 2 4 |
| $^{212}$Bi | 0 52 | 0 18 | 0 26 | 2 2 | 2 4 | 2 4 |
| $^{212}$Pb | 0 52 | 0 18 | 0 26 | 2 2 | 2 4 | 2 4 |
| $^{208}$Tl | 0 18 | 0 07 | 0 09 | 0 8 | 0 8 | 0 8 |
| $^{212}$Po | 0 33 | 0 11 | 0 18 | 1 4 | 1 6 | 1 6 |
| $^{220}$Rn | 1 5E7[b] | 3 6E7 | 1 2E7 | 1 6E7 | 1 1E7 | 9 3E6 |

[a] 1 Bq/sec = 27 pCi/sec

[b] 1 5E7 = 1 5 × $10^7$

Mechanical conditioning of the ore within the mill, such as crushing and grinding, and packaging of the refined $Th(NO_3)_4 \cdot 4H_2O$ product will introduce dust into the process ventilation system. A similarity of behavior of thorium materials to uranium ore and uranium yellow cake was assumed for calculation of particulate releases from the mill and refinery.[16, 17] If 0.008% of the ore processed becomes entrained in the process off-gas[3] and is carried to bag filters with a removal efficiency of 99.3%, then 5.6E-7 of the total ore treated will be released to the environment. Concurrently, an estimated 1.25% of the $Th(NO_3)_4 \cdot 4H_2O$ product becomes airborne during packaging and is carried to the filters.[16] The combined efficiency of the bag filters (99.3%) and the high-efficiency particulate air (HEPA) filters (99.95) allows only 4.4E-8 of the $Th(NO_3)_4 \cdot 4H_2O$ processed to escape to the environment. The radioactivity associated with the $^{232}Th$ and its daughters contained in dust released from the mill and refinery is given in Table 6. Wind blowing on the ore storage pile will also contribute to particulate release, but this term was not calculated because an adequate model for this process was not available.

Radon-220 released from ore in storage and during the acid leaching treatment also contributes to the airborne radioactivity. Release of $^{220}Rn$ from the ore storage pile was calculated assuming a surface area of 7160 $m^2$ and a value for $kp^{-1}$ = 5E-6 $m^2$/sec (comparable to coarse sand) in the equation given in Sect. 4.2.1, yielding a rate of 14.6 MBq/sec. Radon-220 is continuously generated from $^{224}Ra$ decay and is released to the off-gas system during the 12-hr acid leaching of the ore. This contribution of $^{220}Rn$, 36 MBq/sec, was calculated from the equation

$$^{220}Rn = C_{Rn} \cdot \lambda_{220} \cdot t \cdot w \cdot f, \qquad (2)$$

where

$C_{Rn}$ = concentration of $^{220}Rn$ in secular equilibrium, 17.7 kBq/kg ore,

$\lambda_{220}$ = decay constant for $^{220}Rn$, 0.0125/sec,

$t$ = leaching time, 4.32E4 sec,

$w$ = mass of ore processed per day, 1600 Mg (18.5 kg/sec),

$f$ = factor correcting for decay of $^{220}Rn$ between generation and release, 0.2 (ERDA, 1976).

Technology is available to increase the holdup time of the $^{220}Rn$ and thus allow decay to any level justified by a cost-benefit analysis.

The tailings impoundment is also a source of airborne contamination in the form of particles and gas during and after mill operation. Wind acting on exposed tailings solids will pick up small particles, while $^{224}Ra$ dissolved in the liquid portions of the tailings and contained in exposed solids will provide a flux of $^{220}Rn$ gas. Very little of the rapidly decaying $^{220}Rn$ ($T_{1/2}$ = 55.6 sec) released from solids covered by a shallow layer of water will reach the atmosphere. Radon-220 flux from the exposed tailings and the pond surface were calculated using Eq. (1), with the following results:

$J_0$ (exposed tailings) = 1.67 kBq/$m^2$·sec,

$J_0$ (pond) = 19.3 Bq/$m^2$·sec.

Particles will be blown from any exposed tailings beach. During mill operation the tailings will be kept wet or treated chemically to minimize the blowing of particles, but these precautions are not completely

effective The flux for tailings solids which become suspended by wind was calculated using a model based on the phenomenon of saltation,[18] that is, the suspension of small particles (<80 $\mu$m) following impact by larger particles blown across the beach surface The total amount of suspended particles depends on the surface area of tailings beach exposed to wind action during and after the operating life of the mill

A model was selected[11] that accounted for an assumed geometry for the tailings impoundment, the rates at which tailings solids and liquid are supplied, wind velocities, and the net rates of evaporation at the Lemhi Pass and Wet Mountains sites The area of tailings beach and pond during and after mill life was used to determine the $^{220}$Rn source terms given in Table 7 and the particulate radioactivity shown in Table 6

**Table 7 Source terms for $^{220}$Rn during mill operating life and during final evaporation of the pond and covering of the dry tailings after the mill is closed**

| Hypothetical location | Area of dry tailings ($m^2$ X E2) | Area of pond ($m^2$ X E4) | Source term (Bq/sec X E5)[a] | | |
|---|---|---|---|---|---|
| | | | Dry tailings | Pond | Total |
| Montana | | | | | |
| Mill operating life | 4[b] | 57 | 7 | 110 | 117 |
| Evaporation of pond and covering of tailings | 40 | 38 | 40[c] | 73 | 113 |
| Colorado | | | | | |
| Mill operating life | 36 | 49 | 60 | 95 | 155 |
| Evaporation ot pond and covering of tailings | 40 | 27 | 40[c] | 52 | 92 |

[a] 1 Bq/sec = 27 pCi/sec
[b] Read as 4 X $10^2$ $m^2$
[c] A factor of 0 6 was used to correct for decay of $^{224}$Ra parent

The requirements for long-term stabilization of the tailings from a thorium mill are much less demanding than those for a uranium mill In principle, 0 3 m of earth cover will reduce the $^{220}$Rn released to 2E-8 of that emanated and, concurrently, essentially eliminate airborne radioactive particles Since the longest-lived daughter in the $^{232}$Th decay chain is $^{228}$Ra ($t_{1/2}$ = 5 75 years), after about 60 years the tailings impoundment will be equivalent to an ore body containing about 0 05% thorium oxide It would take much longer for uranium tailings to decay to a comparable level because of the longer-lived $^{226}$Ra ($t_{1/2}$ = 1622 years)

### 4.3 Radiological Impact

Population data for dose calculations were obtained from the "Reactor Site Population Analysis"[19] computer code available at Oak Ridge National Laboratory Output from this code is based on the U.S 1970 census. The best available meteorological data for both the Lemhi Pass and the Wet Mountains sites were obtained from the National Oceanic and Atmospheric Administration for a pair of first-order weather stations located near each site Meteorological towers are being installed at potential mine and mill sites in the Lemhi Pass area to obtain additional site-specific weather data for use in later studies The following discussions of radiological impact focuses on the Lemhi Pass area

### 4.3.1 Maximum individual doses

The maximum individual doses at 1.6 km from the mill site are shown in Table 8. Lung is the critical organ, receiving a dose commitment of 35.3 millirems, while doses of 9.5 and 2.4 millirems are delivered to bone and total body respectively.

Table 9 is a breakdown of dose by radionuclides to various organs. Radon-220 and daughters are the primary contributors to all organs. Second in importance is $^{228}$Ra, delivering 36% of the dose to total body and thyroid. The contribution of various exposure pathways to the dose commitment to total body, bone, and lungs is shown in Table 9. Ingestion is the primary mode of exposure for total body (47%) and bone (61%), while inhalation contributes 99% of the dose to lungs.

Additional AIRDOS-II computer runs were made to determine the increase in dose commitment if the site boundary were located 0.8 km from the point of release of radionuclides, instead of 1.6 km as assumed above. It was determined that the maximum individual dose would increase by a factor of approximately

**Table 8. Maximum individual 50-year dose commitment to total body and various organs from radioactivity released to the atmosphere during one year of facility operation**

| Meteorology | Dose commitment (millirems) | | | | | | |
|---|---|---|---|---|---|---|---|
| | Total body | GI tract | Bone | Thyroid | Lungs | Kidneys | Liver |
| **Lemhi Pass site** | | | | | | | |
| Butte | 2.4 | 4.1 | 9.5 | 2.4 | 35.3 | 4.3 | 2.9 |
| Mullan Pass | 2.4 | 3.7 | 9.4 | 2.4 | 32.0 | 3.9 | 2.7 |
| **Wet Mountains site** | | | | | | | |
| Pueblo | 3.7 | 3.8 | 13.1 | 3.7 | 33.4 | 4.2 | 3.3 |
| Alamosa | 3.2 | 3.3 | 11.2 | 3.2 | 28.7 | 3.7 | 2.8 |

**Table 9. Radionuclide contributors to the dose commitment to various organs for maximally exposed individual**

| Radionuclide | Contribution to dose commitment (%) | | | | | | |
|---|---|---|---|---|---|---|---|
| | Total body | GI tract | Bone | Thyroid | Lungs | Kidneys | Liver |
| **Lehmi Pass site (Butte meteorology)** | | | | | | | |
| $^{232}$Th | 3 | <1 | 10 | 4 | <1 | 1 | 13 |
| $^{228}$Ra | 36 | 2 | 23 | 36 | <1 | 4 | 6 |
| $^{228}$Ac | <1 | <1 | <1 | <1 | <1 | <1 | <1 |
| $^{228}$Th | 1 | <1 | 2 | 2 | <1 | <1 | 3 |
| $^{224}$Ra | <1 | <1 | <1 | <1 | <1 | <1 | <1 |
| $^{220}$Rn + D[a] | 59 | 98 | 65 | 58 | 98 | 95 | 78 |
| **Wet Mountains site (Pueblo meteorology)** | | | | | | | |
| $^{232}$Th | 4 | <1 | 12 | 4 | <1 | 2 | 18 |
| $^{228}$Ra | 59 | 4 | 42 | 59 | <1 | 9 | 13 |
| $^{228}$Ac | <1 | <1 | <1 | <1 | <1 | <1 | <1 |
| $^{228}$Th | 2 | 1 | 3 | 2 | 2 | 1 | 5 |
| $^{224}$Ra | <1 | <1 | <1 | <1 | <1 | <1 | <1 |
| $^{220}$Rn + D[a] | 35 | 94 | 43 | 35 | 96 | 87 | 63 |

[a]Contribution of $^{220}$Rn and daughters of $^{220}$Rn.

2 3 when the distance to the maximally exposed individual is reduced by one-half Individual dose commitments calculated in this report could be reduced as necessary by imposing more stringent controls on the handling of ore and tailings and/or by more effective control of airborne particulates at the mill site

Maximum individual doses from thorium mining and milling appear to be similar in magnitude to those reported in assessments on uranium mining and milling operations [16] However, it is emphasized that many of the assumptions made in this study are based on extrapolations from current practices in the uranium mining and milling industry, and this conclusion is, therefore, subject to uncertainty Consequently, as the thorium production industry expands to accommodate the demand for thorium as a fertile material in nonproliferative fuel cycles, assessments such as this one should be extended to include on-site measurements of source terms and meteorological and environmental parameters

### 4 3.2 Population doses

The population residing within 80 km of the Lemhi Pass site is small (~15,000 people) Consequently, population dose commitments in man-rems for the hypothetical thorium mine and mill (Table 10) are relatively low The critical organs are bone, lungs, and kidneys, with the highest dose, 0 7 man-rem, received by the lungs The most important radionuclides (Table 11) include $^{220}$Rn and its daughters, which deliver 64 to 98% of the dose to these organs Radium-228 is the second most important contributor, with the remainder of the dose due to $^{232}$Th and $^{228}$Th The dose to total body is divided between inhalation (39%), surface exposure (36%), and ingestion (25%) Dose to bone is primarily by inhalation (53%), while surface exposure and ingestion contribute 18 and 27% respectively Exposure to lungs is almost entirely from inhalation of particulate matter (98%)

In addition to the small number of individuals living near the site, one factor leading to relatively low population exposures from thorium mining and milling is the rapid decay of $^{220}$Rn (55.6 sec) compared with that of $^{222}$Rn (3 8 days) This property implies that the mean distance of $^{220}$Rn transport from the site will be relatively short before decay to particulate daughters occurs Since the probability of deposition through washout, rainout, or gravitational settling is greater for particulates than gases, daughters of $^{220}$Rn are less likely than those of $^{222}$Rn to reach populated areas

**Table 10. Population dose commitment to total body and various organs from radioactivity released to the atmosphere during one year of facility operation**

| Meteorology | Dose commitment (man-rems) | | | | | | |
|---|---|---|---|---|---|---|---|
| | Total body | GI tract | Bone | Thyroid | Lungs | Kidneys | Liver |
| **Lemhi Pass site[a]** | | | | | | | |
| Butte | 0 05 | 0 03 | 0 1 | 0 05 | 0 7 | 0 07 | 0 06 |
| Mullan Pass | 0 05 | 0 03 | 0 1 | 0 05 | 0 8 | 0 08 | 0 06 |
| **Wet Mountains site[b]** | | | | | | | |
| Pueblo | 0 3 | 0 1 | 0 7 | 0 2 | 2 8 | 0 3 | 0 3 |
| Alamosa | 0 3 | 0 2 | 0 9 | 0 3 | 3 7 | 0 4 | 0 3 |

[a]Total population within 80 km = 15,000
[b]Total population within 80 km = 260,000

Table 11. Radionuclide contributions to the population dose commitment to various organs

| Radionuclide | Contribution to the dose commitment (%) | | | | | |
|---|---|---|---|---|---|---|
| | Total body | GI tract | Bone | Thyroid | Lungs | Kidneys |
| **Lemhi Pass site (Butte meteorology)** | | | | | | |
| $^{232}$Th | 3 | <1 | 12 | 3 | <1 | 1 |
| $^{228}$Ra | 28 | 7 | 22 | 28 | <1 | 4 |
| $^{228}$Ac | <1 | <1 | <1 | <1 | <1 | <1 |
| $^{228}$Th | 2 | 2 | 2 | 2 | <1 | <1 |
| $^{224}$Ra | <1 | <1 | <1 | <1 | <1 | <1 |
| $^{220}$Rn + D[a] | 67 | 91 | 64 | 67 | 98 | 94 |
| **Wet Mountains site (Alamosa meteorology)** | | | | | | |
| $^{232}$Th | 3 | <1 | 13 | 3 | <1 | 1 |
| $^{228}$Ra | 48 | 16 | 40 | 48 | <1 | 10 |
| $^{228}$Ac | <1 | <1 | <1 | <1 | <1 | <1 |
| $^{228}$Th | 3 | 3 | 3 | 3 | 2 | 1 |
| $^{224}$Ra | <1 | <1 | <1 | <1 | <1 | <1 |
| $^{220}$Rn + D[a] | 46 | 81 | 45 | 46 | 97 | 87 |

[a]Contribution of $^{220}$Rn and daughters of $^{220}$Rn.

## 5. REPROCESSING AND REFABRICATION OF ($^{232}$Th, $^{233}$U) CARBIDE FUELS

### 5.1 Description of Reprocessing and Refabrication Plants

Plants for the reprocessing and refabrication of irradiated nuclear fuels are designed and constructed to minimize exposure of operating personnel to radioactivity and to control the release of radioactivity to the environment. The principal control is provided by a primary cell which for recently irradiated and recycled thorium and thorium-$^{233}$U fuels must have thick concrete walls for protection from gamma radiation. A tightly constructed building serves as the secondary container. Additional control over release of radioactivity is obtained by maintaining pressure differentials between the work cells, building interior, and the building exterior to assure that any atmospheric in-leakage will be from a clean area into the contaminated or potentially contaminated areas. The working volumes of cells and reaction vessels located in the cells are purged or vented to remove gaseous reaction products. These gaseous streams are treated before venting to the atmosphere to remove acids, various radioactive species, and entrained particles.

The operations used in aqueous reprocessing of spent thorium-uranium carbide fuels and the gaseous radioactive isotopes which will be present in the off-gas system are shown in Fig. 7. The methods of treating the off-gas to control the release of these isotopes is shown in Fig. 8. The principal steps in reprocessing are a mechanical shearing operation to expose the fuel for chemical treatment, an oxidation step to convert the carbide fuel to oxide, nitric acid dissolution, solvent extraction to separate thorium and uranium from fission and decay products, and a conversion step to yield thorium oxide and uranium oxide products. Off-gas treatment is designed to separate radioactive nuclides from the nonradioactive gases in the effluent and to convert some radioactive gases to a condensed form, such as liquid or solid. Tritium and $^{14}$C may be converted to tritiated water and $CO_2$; the latter is then reacted further to form a solid carbonate. Krypton-85 remains a gas and is compressed for storage in metallic cylinders. The high-level liquid effluent from reprocessing is concentrated by evaporation. The reader is referred to earlier publications for additional information on the choice of flowsheets for reprocessing and effluent treatment.[2, 20, 21] The

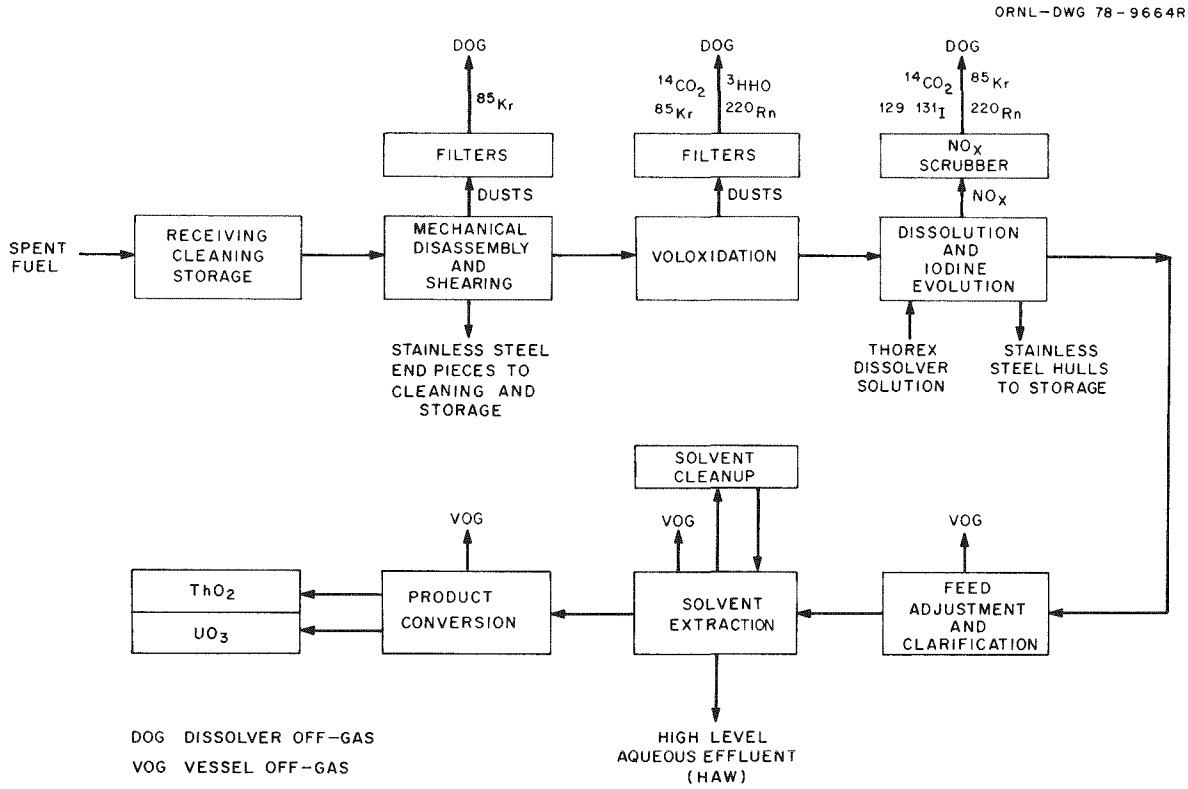


**Fig. 7. Conceptual chemical flow diagram for reprocessing spent thorium-uranium carbide fuels.**

ultimate disposal of these concentrated wastes is the subject of an ongoing DOE development program and is not considered here

The synthesis and refabrication of thorium-uranium carbide fuel pellets requires a series of mechanical steps and high-temperature treatments As shown in Fig 9, thorium oxide or a mixture of thorium oxide plus uranium oxide is mixed with carbon then heated at a high temperature to remove oxygen and form carbide phases (carbothermic reduction) The resulting carbides are ground to fine powders, pressed, and sintered to the required density. Since the carbide product will oxidize readily, especially in the fine powder state, a protective atmosphere of chemically inert gas, such as argon, must be provided An inert-gas purge is also used to remove the gaseous products of the reduction process Radioactivity is introduced into the off-gas in the form of small entrained particles and radon gas Entrainment of particles results from handling fuel powders at several steps in the preparation of the fuel In addition, the radioisotopes of lead ($^{212}Pb$), bismuth ($^{212}Bi$), and polonium ($^{212}Po$ and $^{216}Po$), formed from decay of thorium ($^{228}Th$), are expected to be released from the fuel during heating at the high temperatures required for carbide synthesis and sintering of pellets It is assumed that no contaminated process-liquid waste is released directly to the environment, but some liquid, potentially contaminated at very low levels, may be released as a result of collection from laboratory sinks, mopping of floors, laundry water, and showers The radioactive content cannot exceed and must not routinely equal the limits set by Title 10, Code of Federal Regulations, Part 20 (10CFR Part 20) (Table II, Column 2, Appendix B) Experience reported by the Nuclear Regulatory Commission[22] in fabricating $(U,Pu)O_2$ fuel suggests that an annual average concentration equal to 7% of the 10CFR 20 limits may be expected in potentially contaminated liquid from a refabrication plant

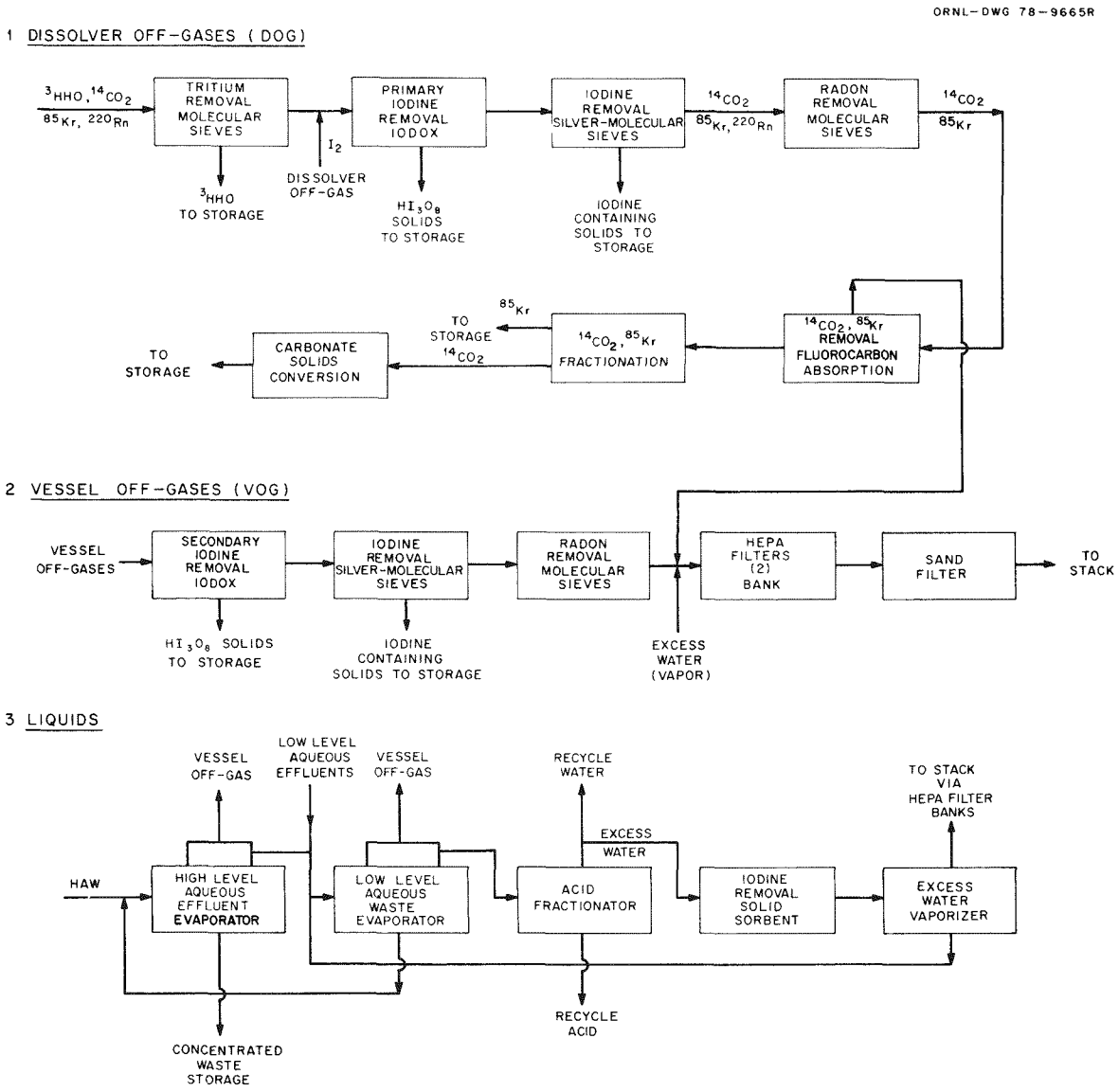


**Fig. 8. Radioactive effluents control system.**

## 5.2 Source Terms from Reprocessing and Refabrication

### 5.2.1 Reprocessing

The composition of the (Th,U)C reactor fuel at discharge from the reactor and after being held in storage for one year was calculated by the ORIGEN computer code, appropriate confinement factors for reprocessing[4] were applied, and the annual release calculated for a reprocessing plant in which the fuel required to generate 50 GW(e)-years of energy was treated. These results are given in Table 12, along with the source terms, for comparison of radioactivity released during reprocessing of (Th,U)C fuel with that from (U,Pu)C fuel calculated in an earlier study.[23]

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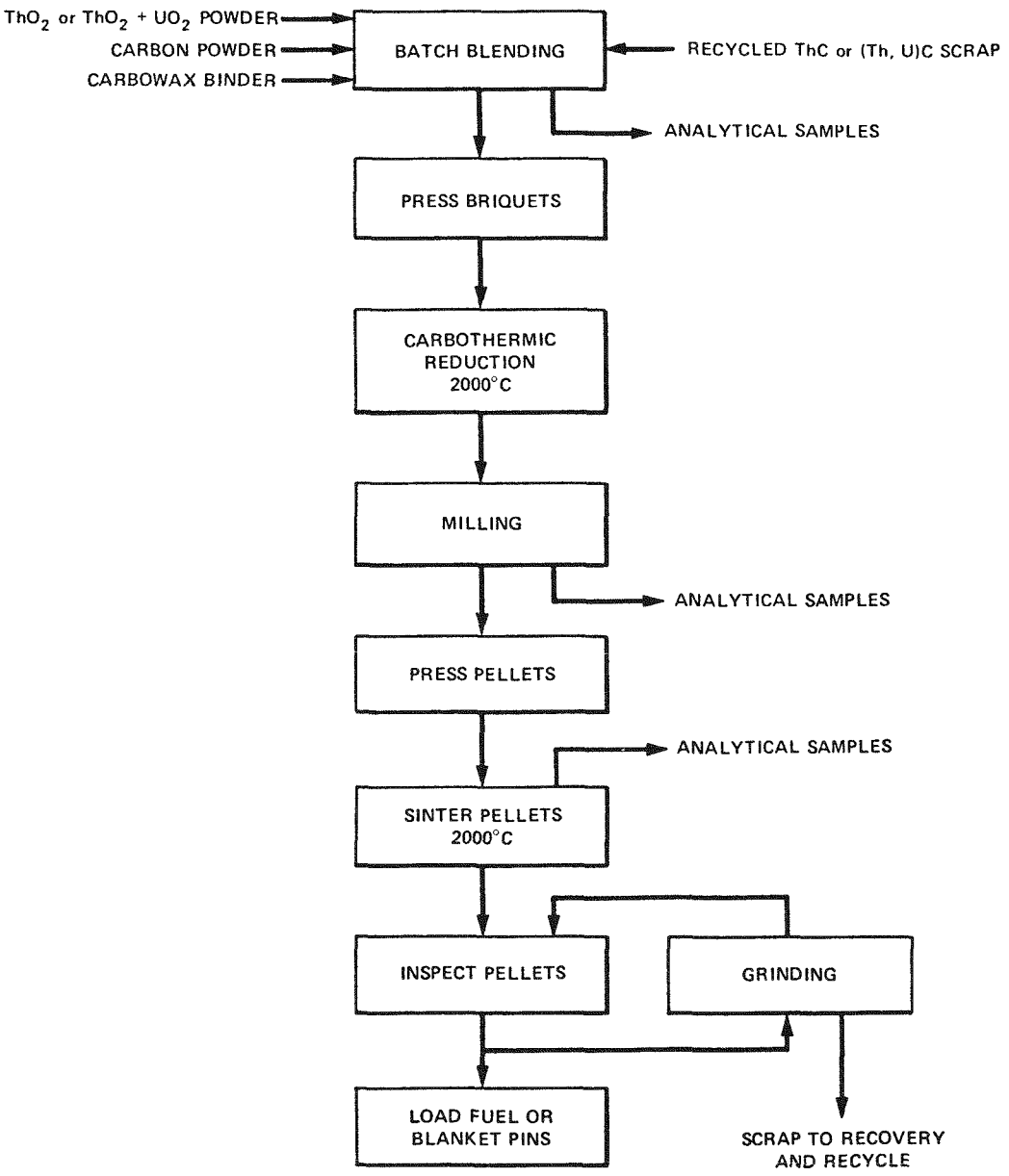


Fig. 9. Flowchart for synthesis of $ThC_{0.95}$ and $(Th,U)C_{0.95}$ and refabrication of pellets.

### 5.2.2 Refabrication

Confinement factors for gaseous and particulate radioactivity entrained in the process and cell off-gas systems during refabrication of (Th,U)C fuel and the resulting amount of airborne activity released from the refabrication plant are given in Table 13, along with releases for (U,Pu)C fuel.[23] The radioactivity which may be contained in potentially contaminated liquid released from the refabrication plant is presented in

Table 12. Comparison of calculated gas-borne effluents from a chemical plant reprocessing one-year-decayed (Th,U)C and (U,Pu)C FBR fuels equivalent to 50 GW(e)-years of energy generated

| Nuclide | Radioactivity releases (Bq/year)[a] | |
|---|---|---|
| | (Th,U)C | (U,Pu)C |
| **Volatiles** | | |
| $^{3}H$ | 3.1E15[b] | 3.4E15 |
| $^{14}C$ | 1.4E12 | 7.5E11 |
| $^{85}Kr$ | 1.2E16 | 2.8E15 |
| $^{129}I$ | 3.0E8 | 2.1E8 |
| $^{131}I$ | 2.4E2 | 1.7E2 |
| $^{220}Rn$ | 9.7E13 | *c* |
| **Semivolatiles** | | |
| $^{103}Ru$ | *c* | 2.5E8 |
| $^{106}Ru$ | 2.9E9 | 2.9E10 |
| **Particulates** | | |
| $^{54}Mn$ | 1.0E4 | *c* |
| $^{55}Fe$ | 2.2E7 | *c* |
| $^{58}Co$ | 1.1E6 | *c* |
| $^{89}Sr$ | 2.9E8 | 6.6E7 |
| $^{90}Sr$ | 1.4E9 | 4.1E8 |
| $^{90}Y$ | 1.4E9 | 4.1E8 |
| $^{91}Y$ | 5.3E8 | 1.6E8 |
| $^{95}Zr$ | 8.0E8 | 4.5E8 |
| $^{95}Nb$ | 1.7E9 | 9.5E8 |
| $^{99}Tc$ | 1.4E5 | *c* |
| $^{110}Ag$ | *c* | 8.2E7 |
| $^{125}Sb$ | 1.8E8 | 1.2E8 |
| $^{125}Te$ | 4.5E7 | *c* |
| $^{127}Te$ | 6.7E7 | 2.8E7 |
| $^{134}Cs$ | 1.7E8 | 2.0E8 |
| $^{137}Cs$ | 1.3E9 | 1.1E9 |
| $^{144}Ce$ | 8.6E9 | 5.5E9 |
| $^{147}Pm$ | 2.6E9 | 2.5E9 |
| $^{151}Sm$ | *c* | 3.9E7 |
| $^{154}Eu$ | *c* | 1.7E7 |
| $^{155}Eu$ | *c* | 1.3E8 |
| $^{212}Pb$ | 1.9E7 | *c* |
| $^{212}Bi$ | 1.9E7 | *c* |
| $^{224}Ra$ | 1.9E7 | *c* |
| $^{228}Ra$ | 3.6E2 | *c* |
| $^{227}Ac$ | 8.4E3 | *c* |
| $^{231}Pa$ | 1.4E6 | *c* |
| $^{241}Am$ | *c* | 5.1E6 |
| $^{243}Am$ | *c* | 5.2E4 |
| $^{242}Cm$ | *c* | 2.5E7 |
| $^{243}Cm$ | *c* | 5.9E4 |
| $^{244}Cm$ | *c* | 7.4E5 |
| **Thorium** | | |
| $^{228}Th$ | 1.9E8 | *c* |
| $^{229}Th$ | 1.9E4 | *c* |
| $^{230}Th$ | 4.9E3 | *c* |
| $^{232}Th$ | 1.5E4 | *c* |

Table 12 (continued)

| Nuclide | Radioactivity releases (Bq/year)[a] | |
|---|---|---|
| | (Th U)C | (U,Pu)C |
| | **Uranium** | |
| $^{232}U$ | 3 1E8 | *c* |
| $^{233}U$ | 8 2E7 | *c* |
| $^{234}U$ | 1 4E7 | 6 2E3 |
| $^{235}U$ | *c* | 3 0E2 |
| $^{236}U$ | *c* | 7 8E2 |
| $^{238}U$ | *c* | 3 1E4 |
| | **Neptunium** | |
| $^{235}Np$ | 2 4E2 | *c* |
| $^{237}Np$ | 5 0E3 | *c* |
| | **Plutonium** | |
| $^{237}Pu$ | 1 7E1 | 1 1E4 |
| $^{238}Pu$ | 1 7E6 | 2 0E8 |
| $^{239}Pu$ | 8 0E1 | 9 9E7 |
| $^{240}Pu$ | 6 4 E0 | 8 9E7 |
| $^{241}Pu$ | *c* | 3 3E9 |
| $^{242}Pu$ | *c* | 4 3E4 |

[a] 1 Bq/year = 27 pCi/year

[b] 3 1E15 = $3.1 \times 10^{15}$

[c] Nuclide present in concentrations that contribute less than 0 02% of the potential inhalation hazard of the spent fuel

Table 14 In comparing the potential release of isotopes of uranium and thorium contained in the liquid effluent from the refabrication plant, the reader should be aware that feed for preparation of the thorium based fuel is received as oxide, however, the uranium-based fuel is prepared from $UF_6$, therefore the fabrication process contains a step to convert $UF_6$ to $UO_2$ by the ammonium diuranate process with an attendant release of 0 004 $m^3$/sec (86,500 gpd) of water potentially contaminated at a very low level [23] This liquid effluent is assumed to be released to a river having a minimum flow rate of 17 $m^3$/sec (600 cfs) Also, a smaller confinement factor is provided for the fabrication of blanket fuel containing depleted uranium than for recycled thorium because of the lower radiotoxicity of the former

### 5 3 Radiological Impact

The meteorological summaries input to the AIRDOS-II code were the same as those used in the environmental statement for the Liquid-Metal Fast Breeder Reactor (LMFBR) Program [24] Weather data were obtained from 18 stations in the continental United States The data were averaged to obtain joint wind speed and stability categories for the 16 principal compass directions, bounded by radial distances ranging from 1 0 to 80 km Population distribution was also taken from the LMFBR environmental statement

Table 13. Radionuclides released as gas-borne effluents from refabricating (Th,U)C and (U,Pu)C FBR fuels used to generate 50 GW(e)-years of energy[a]

| Radionuclide | Refabrication plant confinement factor (CF) | Radioactivity released (Bq/year)[b] | |
|---|---|---|---|
| | | (Th,U)C | (U,Pu)C |
| $^{212}$Pb | 1E11[c,d] | 1.1E6 | |
| $^{212}$Bi | 1E11 | 1.1E6 | |
| $^{212}$Po | 1E11 | 7.3E5 | |
| $^{216}$Po | 1E11 | 1.1E6 | |
| $^{220}$Rn | 2E3[e] | 1.1E14 | |
| $^{224}$Ra | 1E12 | 1.1E5 | |
| $^{228}$Ra | 1E12 | 7.1E-1 | |
| $^{225}$Ac | 1E12 | 9.5E0 | |
| $^{228}$Th | 1E12 | 1.1E5 | 2.7E-1 |
| $^{229}$Th | 1E12 | 9.5E0 | |
| $^{230}$Th | 1E12 | 2.4E0 | |
| $^{231}$Th | 2.6E5[f] | | 4.8E5 |
| $^{232}$Th | 1E12 | 7.3E0 | |
| $^{234}$Th | 2.6E5[f] | | 3.5E7 |
| $^{232}$U | 1E12 | 1.5E5 | |
| $^{233}$U | 1E12 | 4.1E4 | |
| $^{234}$U | 1E12, 2.6E5 | 7.3E3 | 3.8E6 |
| $^{235}$U | 2.6E5[f] | | 4.8E5 |
| $^{236}$U | 2.6E5[f] | | 6.0E5 |
| $^{238}$U | 2.6E5[f] | | 3.5E7 |
| $^{236}$Pu | 1E12 | | 1.4E2 |
| $^{238}$Pu | 1E12 | | 3.2E5 |
| $^{239}$Pu | 1E12 | | 1.6E5 |
| $^{240}$Pu | 1E12 | | 1.4E5 |
| $^{241}$Pu | 1E12 | | 5.1E6 |
| $^{242}$Pu | 1E12 | | 6.0E1 |

[a]Plant capacity: 6.7 Mg/day of (Th + U) and 4.7 Mg/day of (Pu + U). Plant operation: 300 days per year. Effluent volume: 95 $m^3$/sec ($2 \times 10^5$ scfm) from 18-m-high (60-ft) rooftop stack [as assumed in U.S. AEC, *Environmental Statement, Liquid-Metal Fast Breeder Reactor Program,* WASH-1535, vols. I–VII (December 1974)].

[b]1 Bq/year = 27 pCi/year.

[c]Lower plant confinement factor results from volatility of Pb, Bi, and Po during heat treatments.

[d]1E11 = $1 \times 10^{11}$.

[e]Confinement factor assumed for approximately 10-min holdup of Rn in molecular-sieve bed.

[f]Lower confinement factor applied for depleted uranium than for recycled thorium feed used in fabrication of blanket fuel because of the lower radiotoxicity of the former.

### 5.3.1 Maximum individual doses

The maximum individual doses were calculated assuming a plant boundary located 1 km from the reprocessing and refabrication plants. Dose commitments from the reprocessing plant and a listing of the principal contributors are given in Tables 15 and 16. The thyroid receives the highest dose (6.8 millirems); however, allowable exposure for this organ is generally recognized to be higher than those for the total body and other organs.[25] Tritium contributes more than 60% of the dose to total body, lungs, and kidneys. Cesium-137 and $^{14}$C are also significant contributors; $^{232}$U makes an appreciable contribution to the total body and all organs.

**Table 14. Radionuclides in potentially contaminated liquid effluent from refabricating (Th,U)C and (U,Pu)C FBR fuels used to generate 50 GW(e)-years of energy[a]**

| Radionuclide | Radioactivity concentration guide, water[c] (Bq/ml) | Potential activity released (Bq/year)[b] | |
|---|---|---|---|
| | | (Th,U)C[d] | (U,Pu)C[d] |
| $^{208}$Tl | 1 9E2[e] | 9 4E3 | |
| $^{212}$Pb | 7 4E-1 | 2 6F4 | |
| $^{212}$Bi | 1 5E1 | 2 6E4 | |
| $^{212}$Po | 3 7E4 | 1 7E4 | |
| $^{216}$Po | 3 7E4 | 2 6E4 | |
| $^{220}$Rn | 3 7E4 | 2 6E4 | |
| $^{224}$Ra | 7 4L 2 | 2 6E4 | |
| $^{225}$Ra | 1 9E-2 | 3 0E0 | |
| $^{228}$Ra | 1 1E-3 | 1 6E-1 | |
| $^{225}$Ac | 1 9F-1 | 3 0E0 | |
| $^{228}$Th | 2 6F-1 | 2 6E4 | |
| $^{229}$Th | 1 5L-2 | 3 0E0 | |
| $^{230}$Th | 7 4E-2 | 5 7E-1 | |
| $^{231}$Th | 7 5E0 | 9 6E-2 | 1 2E8[f] |
| $^{232}$Th | 7 4E-2 | 1 7E0 | |
| $^{234}$Th | 7 4E-1 | | 9 3E9[f] |
| $^{232}$U | 1 1E0 | 3 5E4 | |
| $^{233}$U | 1 1E0 | 9 3E3 | |
| $^{234}$U | 1 1E0 | 1 6E3 | 1 0E9[f] |
| $^{235}$U | 1 1E0 | 9 6E-2 | 1 2E8[f] |
| $^{236}$U | 1 1E0 | 7 2E-1 | 1 6E8[f] |
| $^{238}$U | 1 5E0 | | 9 3E9[f] |
| $^{236}$Pu | 1 1E0 | | 2 6E3 |
| $^{238}$Pu | 1 9E-1 | | 6 3E6 |
| $^{239}$Pu | 1 9E-1 | | 3 1E6 |
| $^{240}$Pu | 1 9E-1 | | 2 8E6 |
| $^{241}$Pu | 7 4E0 | | 1 0E8 |
| $^{242}$Pu | 1 9E-1 | | 1 2E3 |

[a] Plant capacity 6 7 Mg/day of (Th + U) and 4 7 Mg/day of (Pu + U) Plant operation 300 days per year Effluent volume 0 012 $m^3$/sec (2 75 × $10^5$ g/d) total after in-plant dilution

[b] 1 Bq/year = 27 pCi/year

[c] Radioactivity concentration guide in unrestricted areas, Title 10, Code of Federal Regulations, Part 20, revised January 1, 1976

[d] (U,Pu)C in 3 8 $m^3$ (1000 gal) and (Th,U)C in 7 6 $m^3$ (2000 gal) volume based on scaling of the assumptions in U S Atomic Energy Commission, *Environmental Statement, Liquid Metal Fast Breeder Reactor Program,* WASH-1535, vols I–VII (December 1974) Results for (U,Pu)C fuel reported in V J Tennery et al , *Environmental Assessment of LMFBR Advanced Fuels A Radiological Analysis of Fuel Reprocessing, Refabrication, and Transportation,* ORNL-5230 (November 1976) Total radioactivity in liquid assumed equal to 7% of 10 CFR Part 20 limit Seven percent of CFR limit based on industrial small plant experience reported in Nuclear Regulatory Commission, *Final Generic Environmental Statement on Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors,* NUREG-0002, Chap IV, Sect D, p IV D-29 (August 1976)

[e] 1 9E2 = 1 9 × $10^2$

[f] Contained in 0 004 $m^3$/sec (86,500 g/d) of aqueous effluent from ammonium diuranate conversion process (based on USAEC and V J Tennery references in footnote *d*)

**Table 15. Maximum individual dose commitment (millirems) to total body and various organs due to radionuclide releases to the atmosphere during one year of reprocessing plant operation**

| Total body | GI tract | Bone | Thyroid | Lungs | Kidneys |
|---|---|---|---|---|---|
| 3.1 | 4.6 | 4.1 | 6.8 | 3.3 | 2.9 |

**Table 16. Significant contributors to maximum individual dose commitment due to radionuclide releases to the atmosphere during reprocessing plant operation**

| Radionuclide | Contribution (%) | | | | | |
|---|---|---|---|---|---|---|
| | Total body | GI tract | Bone | Thyroid | Lungs | Kidneys |
| $^{3}H$ | 64 | 43 | 49 | 29 | 61 | 69 |
| $^{14}C$ | 8 | 4 | 11 | 2 | 3 | 5 |
| $^{85}Kr$ | 1 | <1 | 1 | <1 | 2 | 1 |
| $^{90}Sr$ | 1 | 1 | 9 | <1 | <1 | <1 |
| $^{106}Ru$ | 1 | 32 | 1 | 1 | 1 | 2 |
| $^{129}I$ | <1 | <1 | <1 | 57 | <1 | <1 |
| $^{137}Cs$ | 13 | 6 | 12 | 6 | 10 | 13 |
| $^{144}Ce$ | <1 | 6 | <1 | <1 | 1 | <1 |
| $^{220}Rn$ + daughters | <1 | <1 | 1 | <1 | 5 | 1 |
| $^{228}Th$ | 1 | <1 | 3 | <1 | 3 | <1 |
| $^{232}U$ | 9 | 5 | 12 | 4 | 12 | 7 |
| $^{233}U$ | <1 | <1 | <1 | <1 | 1 | <1 |

Maximum individual doses due to refabrication, and the major contributors to dose, are given in Tables 17 and 18. Doses are generally reduced with respect to those for reprocessing, with the exception of lung doses. Virtually all of the dose resulting from refabrication of fuel derives from $^{220}Rn$ and its daughters; $^{232}U$ as such contributes about 1% of the total. Radon-220 daughters, primarily $^{212}Pb$, contribute essentially all of the dose to GI tract, bone, lungs, and kidneys and 99% of the dose to total body and thyroid. Holdup of $^{220}Rn$ for additional time prior to release from the refabrication plant, followed by HEPA

**Table 17. Maximum individual dose commitment (man-rems) to total body and various organs due to radionuclide releases to the atmosphere during one year of refabrication plant operation**

| Total body | GI tract | Bone | Thyroid | Lungs | Kidneys |
|---|---|---|---|---|---|
| 0.15 | 0.41 | 0.63 | 0.15 | 3.8 | 0.42 |

**Table 18. Significant contributors to maximum individual dose commitment due to radionuclide releases to the atmosphere during refabrication plant operation**

| Radionuclide | Contribution (%) | | | | | |
|---|---|---|---|---|---|---|
| | Total body | GI tract | Bone | Thyroid | Lungs | Kidneys |
| $^{220}Rn$ + daughters | 99 | ~100 | ~100 | 99 | ~100 | ~100 |
| $^{232}U$ | 1 | <1 | <1 | 1 | <1 | <1 |

filtration of daughter $^{212}Pb$ particulates, and/or an increase in the effective stack height for the refabrication facility, would greatly decrease the dose to the lung for this plant Either procedure is technologically feasible if later found to be cost effective

The annual total-body, bone, and kidney doses to individuals via aquatic pathways are summarized in Table 19 Dose commitments are dominated by $^{228}Th$, which contributes 80% of the dose to total body and about 90% of the dose to bone and kidney Liquid releases containing the radioactivity levels assumed in this work will cause very low total doses to individuals residing near a fuel refabrication facility

### 5.3 2 Population doses

Population exposures are calculated assuming approximately 1 million people live within an 80-km radius from the reprocessing and refabrication plants Thyroid receives the largest dose due to operation of the reprocessing plant (see Table 20) Relative doses to the total body, GI tract, bone, lungs, and kidneys parallel those for the maximally exposed individual Significant radionuclide contributors to the population dose are listed in Table 21 These results indicate that tritium is the most important contributor to all organs Ruthenium-106 and $^{129}I$ are seen to act as primary contributors to GI tract and thyroid respectively, while causing little dose to other organs Table 22 presents dose commitments to the population within 80 km of the refabrication plant and indicates that lungs receive the highest dose The major contributors are $^{220}Rn$ and its daughters (see Table 18) These results are similar to those observed for the maximally exposed individual located adjacent to the refabrication facility boundary

**Table 19. Summary of dose commitments to individuals due to one year of release of liquid effluents from a (Th,U)C fuels refabrication plant**

| Radionuclide | Dose commitment (millirems) | | |
|---|---|---|---|
| | Total body | Bone | Kidney |
| $^{212}Pb$ | 6 1E-7 | 7.6E-6 | 1 8E-5 |
| $^{212}Bi$ | 7 3E-9 | 1 1E-8 | 5 1E-7 |
| $^{224}Ra$ | 4 1E-5 | 3 2E-4 | 4 1E-5 |
| $^{228}Th$ | 4 8E-4 | 1 4E 2 | 1 3E-3 |
| $^{232}U$ | 7 9E 5 | 1 1E-3 | 1 2E-4 |
| $^{233}U$ | 3 8E-6 | 6 2E-5 | 1 4E-5 |
| $^{234}U$ | 6 5E-7 | 1 0E 5 | 2 5E-6 |
| Total doses | 6 1E-4 | 1 6E 2 | 1 5E-3 |

**Table 20. Population dose commitment (man-rems) to total body and various organs due to radionuclide releases to the atmosphere during one year of reprocessing plant operation**

| Total body | GI tract | Bone | Thyroid | Lungs | Kidneys |
|---|---|---|---|---|---|
| 39 2 | 50 7 | 47 6 | 60 1 | 40 2 | 36 7 |

**Table 21. Significant contributors to population dose commitment due to radionuclide releases to the atmosphere during reprocessing plant operation**

| Radionuclide | Contribution (%) | | | | | |
|---|---|---|---|---|---|---|
| | Total body | GI tract | Bone | Thyroid | Lungs | Kidneys |
| $^{3}H$ | 74 | 57 | 61 | 48 | 72 | 79 |
| $^{14}C$ | 9.3 | 5.8 | 13 | 2.8 | 4 | 5.7 |
| $^{85}Kr$ | 1 | 1 | 1 | 1 | 2 | 1 |
| $^{90}Sr$ | <1 | <1 | 4.8 | <1 | <1 | <1 |
| $^{106}Ru$ | 1 | 23 | 1 | 1 | 1 | 1.2 |
| $^{129}I$ | <1 | <1 | <1 | 39 | <1 | <1 |
| $^{137}Cs$ | 7.3 | 4.0 | 7.4 | 5.1 | 6.5 | 7.2 |
| $^{144}Ce$ | <1 | 4.6 | <1 | <1 | 1 | <1 |
| $^{220}Rn$ + daughters | <1 | <1 | 1 | <1 | 2.7 | <1 |
| $^{228}Th$ | <1 | <1 | 1.9 | <1 | 2.0 | <1 |
| $^{232}U$ | 5.5 | 3.2 | 7.8 | 3.1 | 8.2 | 4.4 |
| $^{233}U$ | <1 | <1 | <1 | <1 | 1 | <1 |

**Table 22. Population dose commitment (man-rems) to total body and various organs due to radionuclide releases to the atmosphere during one year of refabrication plant operation**

| Total body | GI tract | Bone | Thyroid | Lungs | Kidneys |
|---|---|---|---|---|---|
| 0.12 | 0.35 | 0.52 | 0.12 | 2.6 | 0.12 |

### 5.3.3 Radiological importance of $^{232}U$ in recycle of ($^{232}Th$,$^{233}U$)C fuel

The decay chain for $^{232}U$ ($t_{1/2}$ = 72 years) contains no "stopping" nuclide such as is found in the $^{238}Pu$, $^{239}Pu$, $^{240}Pu$, and $^{241}Pu$ chains. This property implies that the effective absorbed energy per disintegration for $^{232}U$ is high when compared with most other radionuclide chains.[26] Estimated levels of the $^{232}U$ content of fuels range to >4000 ppm $^{232}U/U$. The effect of $^{232}U$ level on the dose to the maximally exposed individual was calculated for a reprocessing plant and a refabrication plant, with the results shown in Tables 23 and 24. For the reprocessing plant, variation of $^{232}U$ content from 10 ppm to 5000 ppm raises the total-body dose by only a factor of 1.4, from 2.8 millirems to 4.0 millirems. The highest dose is delivered to bone, 6.0 millirems for 5000 ppm $^{232}U$. Similar results for increasing $^{232}U$ content were observed for population doses.

An increase of $^{232}U$ content to 5000 ppm had an appreciable effect on the relative dose to the individual exposed to a fuel refabrication facility. However, the resulting absolute dose was still moderate, with values of 0.59 millirem to the total body and 15.1 millirems to the lungs. The population dose also increased, but even at 5000 ppm, $^{232}U$ content does not appear to create an unacceptable insult.

### 5.3.4 Radiological importance of tritium in reprocessing of ($^{232}Th$,$^{233}U$)C fuel

Tritium can be a major contributor to the dose to individuals and populations in the vicinity of a nuclear fuel reprocessing plant. Its potential contribution to exposure may be even greater than currently

**Table 23. Effects of variation in $^{232}U$ content on maximum individual dose commitment to total body, bone, and lungs due to radionuclide releases to the atmosphere during one year of reprocessing facility operation**

| $^{232}U$ content (ppm) | Dose commitment (millirems) | | |
|---|---|---|---|
| | Total body | Bone | Lungs |
| 10 | 2.8 | 3.5 | 2.6 |
| 100 | 2.8 | 3.5 | 2.7 |
| 500 | 2.9 | 3.7 | 2.9 |
| 1260 | 3.1 | 4.1 | 3.3 |
| 2500 | 3.4 | 4.7 | 3.9 |
| 5000 | 4.0 | 6.0 | 5.3 |

**Table 24. Effects of variation in $^{232}U$ content on maximum individual dose commitment to total body, bone, and lungs due to radionuclide releases to the atmosphere during one year of refabrication facility operation**

| $^{232}U$ content (ppm) | Dose commitment (millirems) | | |
|---|---|---|---|
| | Total body | Bone | Lungs |
| 10 | <0.01 | <0.01 | 0.03 |
| 100 | 0.01 | 0.05 | 0.30 |
| 500 | 0.06 | 0.25 | 1.5 |
| 1260 | 0.15 | 0.63 | 3.8 |
| 2500 | 0.29 | 1.3 | 7.5 |
| 5000 | 0.59 | 2.5 | 15.1 |

assumed, since recent data[27, 28] predict the yield of $^{3}H$ from fast fission in various nuclear fuels to be larger than previously calculated. The effects on individual and population doses of increasing the confinement factor from a value of 1 (used in this work) to 100 are shown in Table 25. The contribution of $^{3}H$ decreases to a few percent when a confinement factor of 100 is imposed. This does not imply that increased confinement of $^{3}H$ is needed. Investigators should be aware, however, of the importance of $^{3}H$ in the source term of a reprocessing facility.

**Table 25. Effects of variation in $^{3}H$ confinement factor on total-body dose commitment due to reprocessing operations**

| $^{3}H$ confinement factor | Maximum individual | | Population | |
|---|---|---|---|---|
| | Total body dose (millirems) | Contribution (%) | Total body dose (man-rems) | Contribution (%) |
| 1 | 3.1 | 64 | 39 | 74 |
| 10 | 1.3 | 15 | 13 | 22 |
| 100 | 1.1 | 2 | 11 | 3 |

# 6. CONCLUSIONS

## 6.1 Mining and Milling

This radiological assessment of thorium mining and milling suggests that the resulting doses should be similar to those from uranium operations. An absolute comparison cannot be made, however, due to differences in some assumptions utilized, including the use of site-specific meteorology and populations at thorium resource sites in the western United States. A distinct difference resulting from the short half-life of $^{220}Rn$ ($t_{1/2}$ = 55.6 sec) in the thorium decay chain compared to that for $^{222}Rn$ ($t_{1/2}$ = 3.82 days) in uranium decay was found in analyzing emission levels following mill shutdown; release of gaseous and particulate radioactivity from a thorium tailings impoundment can be virtually eliminated with a shallow covering of earth. Also, the residual radioactivity in the thorium tailings decreases rapidly because of the relatively short half-life ($t_{1/2}$ = 5.75 years) of $^{228}Ra$. It was determined that additional information is needed regarding several characteristics of thorium ores and site meteorology to improve the quality of future environmental assessments of thorium mining and milling.

## 6.2 Reprocessing and Refabrication

Airborne doses to the maximally exposed individual and the population were found to be generally less for operation of a refabrication plant than for a reprocessing facility. Tritium is the principal contributor to reprocessing plant doses to both the maximally exposed individual and the population. Carbon-14, $^{137}Cs$, and $^{232}U$ account for most of the remaining dose. Dose commitments calculated for the refabrication plant are due almost entirely to $^{220}Rn$ and its daughters. Only 1% of the dose is caused directly by $^{232}U$; however, the $^{220}Rn$ and daughter content of (Th,U)C fuel in the refabrication plant is roughly proportional to $^{232}U$ concentration.

Since $^{3}H$ release and the $^{232}U$ content of (Th,U)C fuels are important in determining exposure, the effects on dose of a range of $^{3}H$ confinement factors (CFs) and of $^{232}U$ concentrations were calculated. A tenfold increase in reprocessing plant CF for $^{3}H$ reduces the individual and population doses by about 60%. An additional tenfold increase in CF causes an additional 15% reduction in dose. The strong dependence of the refabrication plant dose on $^{232}U$ content of the fuel was demonstrated by calculating dose to the individual while varying the $^{232}U$ content from 10 ppm to 5000 ppm. A near-linear dependence was obtained.

A comparison of the radiological impact of recycling (Th,U)C and (U,Pu)C showed little difference. However, the larger content of $^{106}Ru$ in (U,Pu)C fuel caused an increased GI tract dose from the reprocessing plant. Uranium-232 and $^{228}Th$, radionuclides specific to the thorium cycle, contribute 10% of the (Th,U)C fuel dose. The dose from $^{14}C$ varied somewhat due to variations in content assumed for nitrogen as an impurity in (Th,U)C and (U,Pu)C fuels. The presence of $^{232}U$ in the (Th,U) fuel (with attendant $^{220}Rn$ and daughters) causes an appreciable increase in the refabrication plant dose over that previously calculated for (U,Pu)-type fuels.

Release from the (Th,U) fuel refabrication plant of liquids contaminated to a level typical of that reported for industrial experience in fabricating (U,Pu) fuels contributes little to individual doses.

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