MASTER

CONF-771008--2

RADIOLOGICAL ENVIRONMENTAL ASSESSMENT OF THE RECYCLE OF LMFBR ADVANCED FUELS*

V. J. Tennery, E. S. Bomar, J. E. Till, and L. E. Morse

Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

and

M. Pobereskin and W. J. Madia

Battelle Columbus Laboratories
Columbus, Ohio 43201

1. ABSTRACT

The environmental impact resulting from the release of radioactive material during reprocessing and refabrication of spent LMFBR advanced fuels was compared with that from similar treatment of reference oxide fuel. Candidate advanced fuels include carbide [(U,Pu)C] in addition to nitride [(U,Pu)N] with selected concentrations of ^{15}N. Several techniques for preparing enriched ^{15}N were reviewed and estimates were made of the cost of preparing nitrogen enriched to >99% by each method. Core neutronics, fuel management, and designs appropriate for each fuel were used with the ORIGEN code to calculate the compositions of spent core and blanket fuel. The mass of fuel recycled annually was that providing 50 GW(e)-years of energy at the burnup attained by each fuel. Confinement factors for each isotope were identified for reprocessing and refabrication operations and were used to calculate source terms describing isotopic release rates. These source terms were used in the AIRDOS-II code to estimate the 50-year dose to the maximally exposed individual and to both the local and world populations. Total body dose commitments to the maximally exposed individual for oxide and carbide fuels are about 2.8 millirem, while nitride fuel would result in a range of 59 to 3.4 millirem as the ^{14}N content in fresh fuel is varied from 99.64% to zero.

2. INTRODUCTION

Small quantities of radioactive material in the form of gases, gas-borne particles, and liquids are released to the environment during the chemical and mechanical treatments required to recover and refabricate the residual fissile material in spent nuclear fuel elements. In order to predict the environmental impact of these releases, their magnitude must be calculated and a model selected to describe their interaction with the biosphere.

Flow sheets were prepared to identify the various steps in the reprocessing and refabrication of reference oxide and advanced fuels. Mass flow and fractional release values were then used to calculate "source terms" which describe the rate of release of various isotopes to the environment.

The projected environmental impact resulting from recycle of spent LMFBR reference-oxide fuel was described in WASH-1535;[1] both normal operating conditions and accidents were considered. The work reported here compares the environmental impact resulting from recycle of the reference oxide fuel with that for advanced carbide or nitride fuels under normal conditions but the effect of accidents is not included.

The mass of oxide, carbide, or nitride fuel recycled annually was taken as that necessary to generate 50 GW(e)-years of energy at the burnup attained for the respective fuels and a thermal-to-electrical conversion efficiency of 41%.

3. FUEL CYCLE CONSIDERATIONS

The radioisotopes which are present in the spent fuel at the time of reprocessing depend on the starting fuel composition, the characteristics of the neutron flux in the reactor, and fuel management during and after irradiation.

3.1 Determination of Fuel Compositions and Masses

Core loading, fuel management, and equilibrium composition information for 5000 MW(t) LMFBRs fueled with oxide, carbide, and nitride was provided by members of the Applied Physics Division of Argonne National Laboratory (ANL).[2] Using this information, the fissile and fertile isotopes inventory at discharge and after one year of storage was calculated for each

fuel type by using the ORIGEN[3] code. Concurrently, an extensive inventory of fission products was calculated for spent fuel which had been stored for one year prior to reprocessing. Finally, the isotopic change in the composition of the separated plutonium, which was stored for an additional year prior to refabrication, was also calculated.

ORIGEN is an isotopic generation and depletion code which provides an inventory of several hundred fission and activation products, including the actinides through ^{245}Cm. Masses of fissile and fertile isotopes at discharge for each metric ton of uranium plus plutonium charged as carbide fuel is shown in Fig. 1. Similar mass flow data was calculated for oxide and nitride fuels.[4]
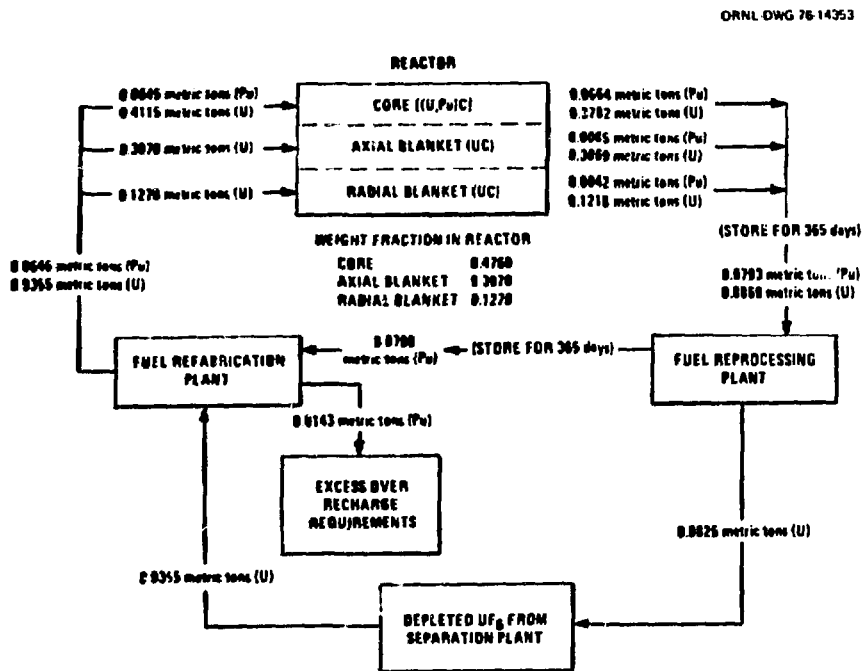

ORNL DWG 76-14353

Fig. 1. Flow of UC and PuC through an LMFBR reactor
and fuel cycle
(1 metric ton U + Pu)

The ^{238}Pu in the fuel charges does not have a significant effect on the core neutronics and, therefore, was not included in ANL's calculations. Plutonium-238 makes an important

contribution to the environmental effect of any plutonium released during recycle, therefore, the isotopic compositions provided by ANL were modified by a 1% addition of ^{238}Pu.[5] The isotopic composition of the original charges and those calculated by ORIGEN after a single additional recycle are given in Table I.

Table I. Isotopic content of plutonium in
fuel charge and in blended spent fuel

Radio-nuclide	Oxide (wt % of Pu)		Carbide (wt % of Pu)		Nitride (wt % of Pu)	
	Original charge[a]	Recycle charge[b]	Original charge[a]	Recycle charge[b]	Original charge[a]	Recycle charge[b]
Pu-238	0.99	0.65	0.99	0.56	0.99	0.60
Pu-239	71.26	73.05	75.46	77.99	74.44	76.65
Pu-240	23.80	23.02	21.19	19.44	22.00	20.53
Pu-241	2.87	2.15	1.84	1.48	1.97	1.63
Pu-242	1.12	1.18	0.52	0.53	0.57	0.61

[a]Equilibrium isotopic composition (ref. 3) adjusted for addition of 1 wt % of ^{238}Pu.

[b]Isotopic composition of Pu in spent fuel as calculated by ORIGEN code following a single additional cycle through the LMFBR.

The total mass of fuel of each type which must be recycled following generation of 50 GW(e)-years of energy depends on the burnup levels reached by the fuels. ANL's calculations included burnup levels at discharge which, incidentally, were limits resulting from fuel pin swelling which reduced coolant flow. The total mass of heavy metal in the three fuel types required to supply 50 GW(e)-years of energy is given in Table II.

Table II. Mass of heavy metal required to generate
50 GW(e)-years of energy from oxide, carbide,
and nitride fuels[a]

Fuel	Core		Axial blanket		Radial blanket		Burnup for reactor[b] (weighted average)[c]	Blended fuel (U + Pu) required (metric tons)
	Burnup[b]	Reactor charge (wt fraction)	Burnup[b]	Reactor charge (wt fraction)	Burnup[b]	Reactor charge (wt fraction)		
Oxide	52,854	0.4353	1860	0.3369	2325	0.2278	24,164	1840
Carbide	60,984	0.4760	3774	0.3970	7032	0.1270	31,420	1415
Nitride	58,056	0.4763	3420	0.3967	5988	0.1270	29,769	1495

[a]Assuming thermal-to-electric conversion efficiency of 41%.

[b]Thermal megawatt-days per metric ton of uranium plus plutonium.

[c]Burnup average for blended fuel: core plus axial and radial blankets.

The effect of ^{15}N enrichment on ^{14}C generation was calculated using the mass and burnup determined for nitride fuel containing nitrogen of natural isotopic composition. Therefore, changes in reactor neutronics resulting from ^{15}N are not taken into account.

3.2 Reprocessing of Spent LMFBR Fuels

Three assumptions are made concerning reprocessing of the reference oxide and advanced fuels: (1) the feed is a blend of spent core and axial and radial blanket fuel, (2) only 10% of the ^3H formed in the fuel elements during irradiation remains in the fuel at the time of reprocessing (ref. 1, p. 4.4-43 and -45), and (3) the confinement factors (CF) for the various radioactive species in the spent fuel are the same as used in WASH-1535 (ref. 1, Sect. 4.4). Confinement factor is defined as the ratio of units processed/units released. A flow sheet was selected[4] which is applicable to all three fuel types and is made up of three major subdivisions: (1) head-end treatment to prepare a feed for the extraction steps, (2) solvent extraction to separate plutonium and uranium from fission products, and (3) final treatment of the uranium and plutonium. Two types of wastes are produced during reprocessing; solid wastes and process off-gas which contains radioactive gases, semivolatile materials, and some particulates. Only the gaseous effluent is considered in this assessment. Any liquid residuals from reprocessing are evaporated to dryness preparatory to disposal as solid waste.

External sodium is removed by reacting with water or steam, extraneous hardware is mechanically removed from the fuel assemblies and the fueled sections are sheared to expose the fuel and internal sodium, if present. Fractional quantities of ^3H, volatile ^{14}C compounds, I_2, the noble fission gases, and some particulate matter are released during shearing and are routed to the process off-gas.

The exposed fuel is oxidized at 450–550°C by a controlled treatment called voloxidation. Direct dissolution of carbide leads to formation of aqueous-soluble carbon compounds which interfere with the subsequent solvent extraction step. While direct dissolution of oxide and perhaps nitride fuels[6] is practical a preoxidation treatment may still be used as an aid in controlling ^3H and ^{14}C emissions by forming ^3H_2O and $^{14}CO_2$ which can be removed along with fission noble gases, $^{129-131}I_2$, and semivolatile oxides of ruthenium by treatment of the process

off-gas. The sequence of head-end operations described above are believed to be capable of releasing >90% of these radioactive species from the fuel.[7] The aqueous feed for solvent extraction is prepared by dissolving the oxidized fuels in nitric acid. Radioactivity released from the dissolver to the process off-gas is treated by a confinement system as described in WASH-1535.[1] Tritiated water is removed by adsorption on molecular sieve.[7] The Iodox[7] system is used to remove various iodine compounds from the off-gas and noble fission gases are removed by absorption in fluorocarbon[7] solvents. Concurrently ^{14}C as $^{14}CO_2$ is absorbed in the fluorocarbon and recovered for storage by fractionation and conversion to solid $Ca^{14}CO_3$ by reaction with an aqueous slurry of $Ca(OH)_2$. Some economic advantage may accrue to nitride fuels over carbide fuels due to the smaller amount of CO_2 containing ^{14}C which must be recovered and stored. Recovery of ^{14}C as $^{14}CO_2$ is discussed in more detail elsewhere.[8]

Semivolatile ruthenium species are removed from the process off-gas by scrubbing with caustic while particulate matter is removed with appropriate deentrainment devices in concert with high-efficiency particulate air (HEPA) filters and deep-bed sand filters.

The radioactivity released during reprocessing of several types of fuels was calculated including a reference oxide fuel, $(U,Pu)O_2$, containing 20 ppm of natural N, two carbide fuels, $(U,Pu)C$, containing 0 and 150 ppm of natural N, and three nitride fuels, $(U,Pu)N$, containing N (1) of natural isotopic composition, (2) enriched to 99 at. % ^{15}N, and (3) enriched to 100 at. % ^{15}N. The radionuclide ^{14}C was included with the volatile species in accordance with ERDA-1535[9] and the radionuclides ^{242}Pu, ^{243}Am, and ^{243}Cm were included with the particulate species because their radiological inhalation hazards are of the same order of magnitude as the actinides listed in WASH-1535. All other isotopes constituting less than 0.02% of the total actinide hazard were not included, in agreement with the WASH-1535[1] cut-off value. Since confinement of ^3H was not provided for in WASH-1535[1] a CF of 1 was assigned in this study; however, a CF of 100 for ^3H is indicated[10,11] if voloxidation and molecular sieve adsorption is used. Curies of activity released during reprocessing are listed in Table III.

Table III. Calculated gas-borne effluents from plant reprocessing one-year-decayed spent fuel

Nuclide	Radioactivity released (µCi) (Blended core plus blankets)			Confinement factor[e]
	Oxide, $(U,Pu)O_2$ [a,b]	Carbide, $(U,Pu)C$ [a,c]	Nitride, $(U,Pu)N$ [a,d]	
		Volatiles		
H-3	9.043 E 10[f]	9.220 E 10	9.172 E 10	1 E 0
Kr-85	7.370 E 10	7.507 E 10	7.469 E 10	1 E 2
I-129	5.797 E 3	5.779 E 3	5.754 E 3	1 E 4
I-131	4.644 E 3	4.669 E 3	4.653 E 3	1 E 4
C-14[g]	2.408 E 6	1.044 E 4	8.267 E 9	1 E 2
	(20 ppm N)	(0 ppm N)	(nat. N)	
		2.019 E 7	1.700 E 8	1 E 2
		(150 ppm N)	(99% N-15)	
			8.782 E 7	1 E 2
			(100% N-15)	
		Semivolatiles		
Ru-103	6.644 E 3	6.721 E 3	6.684 E 3	1 E 9
Ru-106	7.717 E 5	7.705 E 5	7.666 E 5	1 E 9
		Uranium		
U-234	2.446 E 1	1.666 E 1	1.848 E 1	5 E 8
U-235	1.170 E 2	8.055 E 3	8.738 E 3	5 E 8
U-236	2.081 E 2	2.103 E 2	1.979 E 2	5 E 8
U-238	1.099 E 0	8.359 E 1	8.858 E 1	5 E 8
		Plutonium		
Pu-237	3.182 E 1	2.685 E 1	3.151 E 1	2 E 9
Pu-238	7.898 E 3	5.355 E 3	5.921 E 3	2 E 9
Pu-239	3.176 E 3	2.685 E 3	2.723 E 3	2 E 9
Pu-240	3.595 E 3	2.408 E 3	2.622 E 3	2 E 9
Pu-241	1.622 E 5	8.647 E 4	9.884 E 4	2 E 9
Pu-242	3.091 E 0	1.164 E 0	1.383 E 0	2 E 9
		Particulates		
Sr-89	1.740 E 3	1.786 E 3	1.782 E 3	5 E 9
Sr-90	1.082 E 4	1.111 E 4	1.106 E 4	5 E 9
Y-90	1.083 E 4	1.111 E 4	1.106 E 4	5 E 9
Y-91	4.259 E 3	4.355 E 3	4.345 E 3	5 E 9
Zr-95	1.204 E 4	1.218 E 4	1.214 E 4	5 E 9
Nb-95	2.545 E 4	2.756 E 4	2.566 E 4	5 E 9
Ag-110	1.857 E 3	2.214 E 3	2.104 E 3	5 E 9
Sb-125	3.369 E 3	3.332 E 3	3.314 E 3	5 E 9
Te-127m	7.597 E 2	7.500 E 2	7.452 E 2	5 E 9
Cs-134	4.303 E 3	5.358 E 3	5.066 E 3	5 E 9
Cs-137	2.811 E 4	2.842 E 4	2.825 E 4	5 E 9
Ce-144	1.453 E 5	1.476 E 5	1.471 E 5	5 E 9
Pm-147	6.617 E 4	6.664 E 4	6.654 E 4	5 E 9
Sm-151	1.054 E 3	1.055 E 3	1.053 E 3	5 E 9
Eu-154	3.865 E 2	4.550 E 2	4.378 E 2	5 E 9
Eu-155	3.438 E 3	3.496 E 3	3.463 E 3	5 E 9
Am-241	2.682 E 2	1.379 E 2	1.573 E 2	5 E 9
Am-243	3.465 E 0	1.397 E 0	1.545 E 0	5 E 9
Cm-242	1.277 E 3	6.746 E 2	6.989 E 2	5 E 9
Cm-243	2.431 E 0	1.607 E 0	1.569 E 0	5 E 9
Cm-244	4.539 E 1	2.012 E 1	2.021 E 1	5 E 9

[a] Fabricated with recycle plutonium.

[b] Burnup, 24,164 MW(t)-days per metric ton of uranium plus plutonium (averaged burnup for core plus blankets); reprocessing rate, 1840 metric tons of uranium plus plutonium reprocessed per year for 50 GW(e)-years of energy generated.

[c] Burnup, 31,420 MW(t)-days per metric ton of uranium plus plutonium (averaged burnup for core plus blankets); reprocessing rate, 1415 metric tons of uranium plus plutonium reprocessed per year for 50 GW(e)-years of energy generated.

[d] Burnup, 29,769 MW(t)-days per metric ton of uranium plus plutonium (averaged burnup for core plus blankets); reprocessing rate, 1495 metric tons of uranium plus plutonium reprocessed per year for 50 GW(e)-years of energy generated.

[e] Ratio of curies processed to curies released.

[f] To be read as 9.043×10^{10}.

[g] Nitrogen content of fuel as indicated.

3.3 Refabrication of LMFBR Fuels

As shown in Table II, the fuel refabrication plant must fabricate 1840 metric tons of oxide fuel to provide for an annual production of 50 GW(e)-years of energy making the plant similar in size to that described in WASH-1535. The refabrication plant contains two principal sections, one for blanket pellets and a second section in which the plutonium-bearing mixed fuel is fabricated. The fractional mass flows of 1 metric ton of uranium plus plutonium through these two sections for oxide, carbide, and nitride fuels is shown elsewhere.[4]

Mechanical mixing of UO_2 and PuO_2 was selected for this study because it has been used in the preparation of FTR fuels and can be used to prepare carbide or nitride fuels. Carbide is produced by adding carbon to the oxide mixture and carrying out a carbothermic reduction under vacuum or flowing inert gas. Nitride fuel is formed if reduction is done in a flowing nitrogen atmosphere. Carbon is removed from the nitride fuel by heating in contact with a hydrogen containing gas mixture. Refer to ref. 4 for a flow sheet showing the preparation of the three fuel types.

Purge air, or inert gas in the case of the advanced fuels, from glove boxes or cells used for fuel preparation passes through three high-efficiency particulate (HEPA) filters before being exhausted from the building. The combined transmission factor for three HEPA filters in series is 1.25×10^{-10} (ref. 1, p. 4.3-15). It is assumed that 0.1% of the fuel being refabricated becomes suspended in the glove box atmosphere and is carried to the first HEPA filter. This provides an overall CF of 8×10^{12} (ref. 1, p. 4.3-33) for the mixed fuel section. A CF of 10^{12} was used in calculating release of plutonium-bearing particulates. The two additional steps for carbide or nitride preparation increase the amount of fuel suspended and would, therefore, decrease the confinement factor for the fabrication plant but this is offset by the smaller mass of advanced fuel refabricated, a reduced flow of inert gas through the glove boxes, or complete elimination of glove box effluent if the inert-gas purge is recirculated. Venting of the purge to the atmosphere is assumed here because it has the greater environmental effect. Gas-borne particulates released during fuel refabrication equal the quotient of the mass of fuel processed and the plant CF. The source term describing the type and amount of radionuclides released with gas-borne particulate

matter from the mixed-fuel section of the refabrication plant was calculated.[4] Potentially contaminated liquid will amount to about 1000 gal/day (ref. 1, Fig. 4.3-6) and will contain an annual average concentration equal to 7% of that permitted by federal regulations (10 CFR 20, Table II, column 2, Appendix B) as was assumed in NUREG-0002[12] and was selected here to calculate the environmental release of liquid from the mixed-fuel section of the refabrication plant.[4]

Operation of the uranium portion of the refabrication plant is essentially like that described in the LMFBR Program environmental statement (ref. 1, p. 4.3-13). Uranium recovered from reprocessed fuel is sent to storage for future use or is greatly diluted with fresh depleted uranium, therefore the isotopic composition of the uranium is essentially unchanged.

The uranium feed is received as UF_6 and is converted to UO_2 by the ammonium diuranate (ADU) process (ref. 1, Fig. 4.3-8). Blanket pellets of UO_2 are prepared by conventional methods of pressing and sintering. Uranium carbide and UN are prepared by a carbothermic process.

The requirements for containment of uranium are much less demanding than for plutonium. A CF of 2.6×10^5, used in an earlier environmental assessment (ref. 1, p. 4.3-42), is based on commercial experience. The quantity of gas-borne release was obtained[4] by prorating the mass flow of uranium in three fuels examined here to the 720 metric ton/year required in the earlier study (ref. 1, Table 4.3-6).

Similarly, the radioactivity contained in liquid-effluent from the blanket section of the refabrication plant was obtained by prorating the release reported in WASH-1535 (ref. 1, Table 4.3-7) to comparative mass flows in the two plants and the attendant proportional change in volume of liquid effluent.[4]

3.4 Transportation

The differences in the environmental effects of transporting spent advanced fuels compared to the reference oxide fuel are minimal. The small differences in heat generation rates and neutron fluxes from the three fuel types are readily compensated for by adjusting the number of assemblies transported at one time to meet the limits set by the Department of Transportation for temperature and radiation dose rate at the surface of the carrier.

The reader is referred to an earlier study[4] for a general analysis of the transportation of fuels and waste materials as required by recycle of spent LMFBR fuels.

4. RADIOLOGICAL IMPACT OF REPROCESSING AND REFABRICATION

4.1 Methodology for the Radiological Assessment

The radiological impact of the fuel reprocessing plant was assessed by calculating radiation doses to individuals and populations living in the vicinity of the nuclear facility. Radiological impact was estimated as the 50-year dose commitment to individuals or populations in units of millirems or man-rems per year of facility operation. In this work, the terms dose and dose commitments are used interchangeably, and each implies a 50-year commitment. Doses to different organs may vary considerably for internal exposure from ingested or inhaled materials because some radionuclides concentrate in certain organs of the body. For this reason, estimates of radiation dose to the total body and major organs are considered for all pathways of internal exposure, based on parameters applicable to an average adult.

The AIRDOS-II computer code[13] was used to estimate 50-year dose commitments from airborne effluents to the maximally exposed individual and to the population in the vicinity of the nuclear facility handling reference oxide or advanced LMFBR fuel. The AIRDOS-II code includes the dose commitment from pathways of inhalation, immersion in air, exposure to ground surfaces, ingestion of food produced in the area, and swimming in water contaminated with radionuclides. Doses were estimated for the total body, GI tract, bone, thyroid, lungs, muscle, kidney, liver, spleen, testes, and ovaries. Meteorological data and population distribution were taken from WASH-1535.[1] The global impact of releases of ^{14}C from a fuel reprocessing plant was estimated using a multicompartment diffusion model developed by Killough.[13,14]

4.2 Maximum Individual Dose Commitments

Essentially no differences were found to exist in the radiological impact when advanced fuels are substituted for oxide fuel at an LMFBR fuel refabrication facility. Therefore, we will focus on the results of the reprocessing step, since this is where an impact difference exists for various fuels.

Since no liquid effluents are to be released from the fuel
reprocessing facility being analyzed, the only anticipated
exposure to man results from radioactivity released to the
atmosphere. The maximum dose to an individual living near a
reprocessing plant handling LMFBR fuels would occur at the plant
boundary, which for this study was assumed to be 1000 m from the
stack. Dose commitments for the maximally exposed individual
to the total body, GI tract, and bone for each type of fuel were
calculated and are listed in Table IV.

Table IV. Summary of 50-year dose
commitments to individuals living
at the boundary of a reprocessing
plant handling LMFBR fuels

Type of Fuel	50-year dose commitment[a] (millirems)		
	Total body	GI tract	Bone
Oxide (20 ppm N)	2.8	14	8.0
Carbide (0 ppm N)	2.8	14	6.7
Carbide (150 ppm N)	2.9	14	6.9
Nitride (natural N)	59	59	104
Nitride (99% N-15)	3.9	14	8.9
Nitride (100% N-15)	3.4	14	7.9

[a] For one year of facility operation.

Except for the case where nitrogen of natural isotopic
content is employed in nitride fuel, the dose commitments to all
organs do not vary significantly, and the organ receiving the
highest dose is the GI tract. With nitride fuel fabricated
using natural nitrogen, the bone receives the highest dose, 104
millirems, followed by the total body and the GI tract, each
with 59 millirems. Dose commitments to other organs from that
nitride fuel are also significantly greater than those for oxide
or carbide, or for nitride fuel enriched with 99 or 100% ^{15}N.
Doses listed in Table IV represent maximum values and would
decrease rapidly as the individual's distance from the stack is
increased.

Tritium is the primary contributor to the dose for oxide fuel, carbide fuel, and nitride fuel that is 99 and 100% enriched with ^{15}N and ^{14}C is the second most important contributor. However, for nitride fuel containing natural nitrogen, ^{14}C is responsible for 95% of the dose.

Ingestion is the dominant exposure pathway, and inhalation is second in importance for each type of fuel. As the quantity of ^{14}C released to the environment increases, the contribution to the dose from the ingestion pathway increases. In the case of nitride fuel made with natural nitrogen, ingestion is responsible for more than 98% of the dose.

Figure 2 illustrates the maximum individual dose commitment

Fig. 2. Effect of
fuel on the maximum indiv
total body from all radion
from a reprocessing plant handling (U,Pu,
fuel. Confinement factor for ^{14}C is assumed to
be 100.

and the contribution of ^{14}C to total body near a reprocessing plant as a function of the ^{15}N enrichment in nitride fuel. The dose commitment to total body includes contributions from all radionuclides released from the facility. As the ^{15}N enrichment is increased, the dose commitment decreases and is reduced by approximately 50% when the ^{15}N enrichment is 50% ^{15}N. However, at 50% ^{15}N enrichment, ^{14}C is still responsible for 91% of the dose to the total body. The reduction in dose with increasing ^{15}N enrichment is approximately linear, because the primary mode of production of ^{14}C is by the ^{14}N(n,p)^{14}C reaction and because ^{14}C is the primary contributor to the dose commitment, even up to ^{15}N enrichment values as high as 95%.

The effect of increasing the confinement factor for ^{14}C on the maximum dose commitment to the total body from all radionuclides is illustrated in Fig. 3. In this figure, only the ^{14}C confinement factor is varied, while the confinement factors for all other radionuclides remain constant at the values listed in Table III. The dose commitment from nitride fuel containing natural nitrogen would be significantly greater than the dose commitment from other fuels under consideration in this study, even when the ^{14}C confinement factor is increased to a value of 1000. A reprocessing plant similar to that discussed in this study handling nitride fuel enriched with 99 and 100% ^{15}N or carbide fuel with 150 ppm N would probably require some confinement of ^{14}C to maintain exposures at an acceptable level.

4.3 Local and World Population Dose Commitments

Doses were also calculated for the population living within a 50-mile radius of the fuel reprocessing plant and, in general, follow a pattern similar to the maximum individual dose commitment. There is little difference between the dose commitments calculated for oxide, carbide, and nitride fuel that is enriched to 99 and 100% with ^{15}N. The GI tract receives the highest organ doses in the population for both the oxide and carbide fuels and for nitride fuel with 99 and 100% ^{15}N (approximately 125 man-rems). The highest population dose resulting from reprocessing of nitride fuel synthesized with natural nitrogen is 1240 man-rems to the bone (for 1 million persons).

An analysis of world population doses from ^{14}C released to the atmosphere during reprocessing of advanced fuels has also been completed.[15] This analysis revealed that world population doses from ^{14}C released to the atmosphere at a reprocessing plant
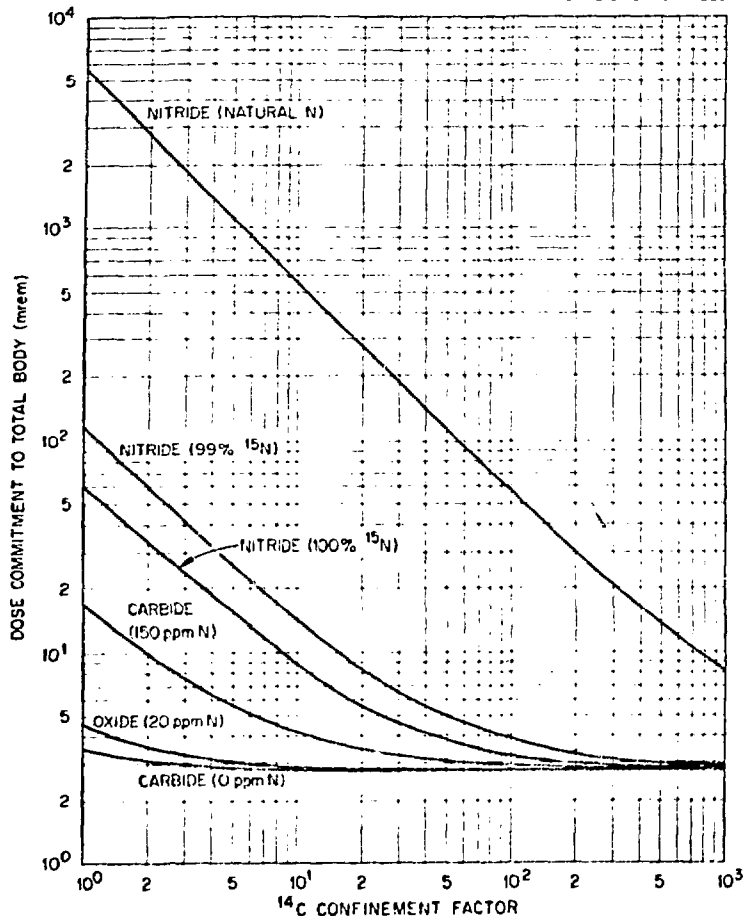
ORNL-DWG 76-15229R

Fig. 3. Maximum individual dose commit-
ment to total body from all radionuclides
annually released from a reprocessing plant
handling LMFBR fuels as a function of the ^{14}C
confinement factor.

handling nitride fuel could be a significant factor in
determining whether this fuel is acceptable from an environ-
mental standpoint. It appears that significant improvements
would have to be made on ^{14}C effluent treatment systems
before nitride fuel fabricated with natural nitrogen could
be used in LMFBRs. Optimization of ^{15}N enrichment in the fuel
and confinement of ^{14}C at the reprocessing plant must be
obtained in terms of economical and technological feasibility
if mixed nitride fuel is to be used on a large scale.

5. TECHNOLOGY AND ECONOMICS OF ^{15}N ENRICHMENT

Material presented in the preceding sections shows the importance of ^{14}C generation and release on the environmental impact of oxide and carbide fuels and especially nitride fuels. As a means of reducing ^{14}C releases an order of magnitude increase in the CF for ^{14}C is presently attainable; however, a further reduction in ^{14}C release may be desirable for nitride fuel. This can be accomplished by substituting ^{15}N for ^{14}N in the fuel with the concurrent benefit of improving the breeding gain for a nitride core.[16] Estimates of the amount of ^{15}N required, methods of enrichment, and estimates of cost are discussed in the following and were reviewed in more detail in an earlier report.[17]

5.1 Break-Even Enrichment Cost and Nitrogen-15 Requirements for Advanced LMFBR Application

Chang and Barthold[16] have estimated the level at which the cost of ^{15}N enrichment would be offset by the savings resulting due to reduced parasitic neutron absorption and the attendant increased breeding ratio. An interpolation of their data to 0.37-in. (0.0094-m) fuel pin diameter gives a break-even value of about $960/kg of ^{15}N for a reactor in which the core and axial blanket contain N enriched to >99% [15]N but the radial blanket contains N of natural isotopic content. Neither the cost decrease due to reduced ^{14}C production nor the increase resulting from separate recycling of enriched and nonenriched fuel are taken into account.

The ^{15}N required by a nitride-fueled LMFBR has been calculated by Barthold[2] and Caspersson et al.[18] Typical ^{15}N requirements for various arrangements of enriched and nonenriched nitride fuel were calculated and show that depending upon the LMFBR introduction scenario assumed, the annual ^{15}N needs vary between 50 and 300 metric ton per year. These requirements are to be compared with the estimated 1976 U.S. capacity of 8 kg for the LASL nitric oxide distillation facility. Thus ^{15}N enrichment capacity would have to be increased by factors of 12,000 and 25,000 before large scale commercialization of nitride fueled LMFBRs could be practical. The present cost of 99% enriched ^{15}N is about $68,000/kg.[19]

5.2 Nitrogen Enrichment Techniques

There are several methods of obtaining enrichment of ^{15}N on a laboratory scale and most effort has been applied to chemical exchange and distillation processes. In both processes two phases are brought together under conditions which favor the concentration of ^{15}N in one of the phases and the energy consumed in the overall processes is strongly dependent on the refluxing step at the end of the exchange columns. A comprehensive description of the exchange process is presented by Johns and Bigeleisen.[20]

Taylor and Spindel[21,22] examined the systems $NO-HNO_3$ (Nitrox process) and $NH_3 - NH_4^+$. Hayford[23] et al. reported on a feasibility study for production of 50 metric ton/year of ^{15}N based on these processes and estimated costs of $1000 to $2000/kg in 1955 dollars and a plant cost of the order of $150 million. In 1972, Williams and Blumkin[24] designed an enrichment plant based on the Nitrox process with a capacity of 15 kg of ^{15}N per year and a concentration of 99.9%. Under their assumptions they estimated a cost of $1900/kg.

Michaels and Schwind[25] recently designed a plant based on the $NO-HNO_3$ exchange and a capacity of 300 metric ton/year. They proposed a bottom reflux system based on the following half-cell reactions:

$$HNO_3 + 3H^+ + 3e \rightleftarrows NO + 2H_2O \qquad (1)$$

$$H_2O \rightleftarrows \frac{1}{2}O_2 + 2H^+ + 2e \qquad (2)$$
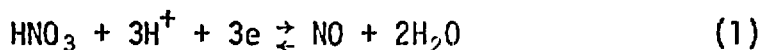
The top reflux system would be a similar cell with the driving potential reversed. A cost analysis for this plant indicated a product cost of $150/kg based upon a reflux energy efficiency of 90%. It was also assumed that the large diameter columns required for this plant would perform similar to that observed to date for 1- to 4-in.-diam columns and that the plant would be sited in conjunction with a large NH_3 complex having a capacity of 1500 metric tons/day and from which HNO_3 would be drawn, ^{15}N removed, and ^{15}N depleted HNO_3 returned, resulting in a negligible feed cost.

A second process for isotopic enrichment is fractional distillation and is based on vapor pressure differences of different isotopic species.[26] It is also characterized by low

power consumption. Michaels and Schwind[25] assumed isotopic redistribution in the design of a ^{15}N fractional distillation enrichment plant of 300 metric ton/year capacity. The resultant ^{15}N cost of \$370/kg is twice that for the $NO-HNO_3$ process.

A third method of ^{15}N enrichment is based on Laser Isotope Separation (LIS) processes. The reader is referred to ref. 17 for a discussion of technical aspects of LIS. The cost/kg of ^{15}N enriched to $\geq 99\%$ in a single step laser stimulation process was estimated as \$300/kg ^{15}N using NO_2 feed. If two stages of laser stimulation are required or the second stage is done using NO fractional distillation, a marked increase in cost to \$1000–8000/kg of ^{15}N results.

6. CONCLUSIONS

The work reported here points to the practicality of using certain fuel and reactor characteristics in conjunction with computer codes and recycle-plant flow sheets to project the environmental impact of the recycle of spent LMFBR fuel. The major environmental impact derives from the fuel reprocessing plant.

The environmental impact resulting from reprocessing carbide fuel or nitride fuel made with nitrogen containing >99 at. % ^{15}N is essentially the same as that due to reprocessing of the reference oxide fuel. The primary source of impact on the environment from these fuels is ^3H with ^{14}C as the second major source. The contribution of ^3H to the dosage of exposed individuals derived here is very conservative since a confinement factor of 1 was assumed, that is, complete release during reprocessing. This was done to permit a direct comparison with the results for ^3H reported in WASH-1535.[1] A large reduction in ^3H release will result from use of voloxidation as a head-end treatment plus drying of the process off-gas with molecular sieves. The greater impact of ^3H occurs before reprocessing since only 10% of the ^3H generated during irradiation is expected to be retained in stainless-steel clad fuel rods.

Carbon-14 becomes the principal source of dosage for nitride fuels made with natural nitrogen or with <60 at. % ^{15}N enrichment. It appears that confinement of ^{14}C will have to be improved before nitride fuels made with natural nitrogen can be used. This can be accomplished by increasing the confinement factor for $^{14}CO_2$ formed during voloxidation, which is a reasonable

expectation. Replacing ^{14}N with ^{15}N reduces the yield of the reaction ^{14}N$(n,p)^{14}$C. The cost of enrichment is at least partially and may be completely offset by an attendant increase in breeding ratio because of the lower neutron-absorption cross section of ^{15}N. Nitrogen can be enriched with ^{15}N as a result of exchange processes which occur between chemical compounds and during fractional distillation of nitric oxide. Performance of exchange processes on the scale required for a large number of nitride fueled LMFBRs remains to be demonstrated, however, since only relatively small quantities of ^{15}N have been prepared to date. Laser isotope separation methods show promise but their evaluation for ^{15}N enrichment is limited at present.

ACKNOWLEDGMENTS

REFERENCES

1. USAEC, *Proposed Final Environmental Statement, Liquid-Metal Fast Breeder Reactor Program*, WASH-1535, Vol. II (December 1974).

2. W. P. Barthold, Applied Physics Division, Argonne National Laboratory, Argonne, IL, private communication.

3. M. J. Bell, "Calculated Radiation Properties of Spent Plutonium Fuels," *Nucl. Technol.* 18, 5–14 (April 1973).

4. V. J. Tennery, L. E. Morse, E. S. Bomar, R. D. Seagren, W. D. Bond, L. B. Shappert, G. S. Hill, and J. E. Till, *Environmental Assessment of LMFBR Advanced Fuels: A Radiological Analysis of Fuel Reprocessing Refabrication, and Transportation*, ORNL-5230 (November 1976).

5. R. A. Doncals, Project Manager, Physics Evaluation and Applications, Westinghouse Advanced Reactor Division, private communication.

6. M. J. Maurice, J. Fischer, and G. N. Kramer, "Recovery of Plutonium from Non-Irradiated Refractory Plutonium and Uranium-Plutonium Oxides, Nitrides, and Carbides," *J. Applied Chemistry* 19, 15–17 (Jan. 1969).

7. O. O. Yarbro, W. S. Groenier, and M. J. Stevenson, "Airborne Effluent Control for LMFBR Fuel Reprocessing Plants," *Proc. Topical Meeting on Controlling Airborne Effluents from Fuel Cycle Plants*, Amer. Nucl. Soc. and Amer. Institute of Chemical Engineers, Sun Valley, Idaho, Aug. 5–6, 1976, pp. 15-1 to 15-12.

8. V. J. Tennery, E. S. Bomar, W. D. Bond, S. V. Kaye, L. E. Morse, and J. E. Till, *Potential Generation and Radiological Impacts of Gaseous ^{14}C Release During Reprocessing of Advanced LMFBR Fuels*, ORNL/TM-5538 (June 1976).

9. ERDA, "Radiological Assessment of Carbon-14 Releases from the LMFBR Fuel Cycle and Radioactive Waste Management," Sect. III D, *Final Environmental Statement, Liquid Metal Fast Breeder Reactor Program*, ERDA-1535, Vol. I (December 1975).

10. O. O. Yarbro, F. E. Harrington, and D. S. Joy, *Effluent Control in Fuel Reprocessing Plants*, ORNL/TM-3899 (March 1974).

11. B. C. Finney, R. E. Blanco, R. C. Dahlman, F. G. Kitts, and J. P. Witherspoon, *Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle for Use in Establishing "as Low as Practicable Guides" — Nuclear Fuel Reprocessing*, ORNL/TM-4901 (May 1975).

12. NRC, *Final Generic Environmental Statement on Use of Recycle Plutonium in Mixed Oxide Fuel in Light-Water Cooled Reactors*, NUREG-0002, Chapter IV, Section D, p. IV D-29.

13. R. E. Moore, *AIRDOS: A Computer Code for Estimating Population and Individual Doses Resulting from Atmospheric Releases of Radionuclides from Nuclear Facilities*, ORNL/TM-4687 (January 1975).

14. G. G. Killough, *A Diffusion-Type Model of the Global Carbon Cycle for Estimation of Dose to the World Population from Release of Carbon-14 to the Atmosphere*, ORNL-5269 (in press).

15. J. E. Till, E. S. Bomar, L. F. Morse, and V. J. Tennery, "A Radiological Assessment of Reprocessing Advanced LMFBR Fuels, *Nucl. Technol.* (in press).

16. Y. I. Chang and W. P. Barthold, "Impact of Nitrogen Enrichment on Breeding Performance of LMFBR Nitride Fuel," *Trans. Am. Nucl. Soc.* <u>21</u>, 407-08 (1975).

17. V. J. Tennery, *Nitrogen-15 Enrichment and Its Potential Use in Advanced LMFBR Mixed-Nitride Fuels*, ORNL/TM-5621 (November 1976).

18. S. A. Caspersson, R. C. Noyes, and J. A. Vitti, "Economic and Nuclear Performance Characteristics of 500-MW(e) Oxide, Carbide, and Nitride LMFBRs," pp. 125-34 in Advanced Reactors: Physics, Design, and Economics, *Proceedings of the International Conference Held at Atlanta, Georgia, Sept. 8-11, 1974*, ed. J. M. Kallfelz and R. A. Karam, Pergamon Press, New York, 1975.

19. V. L. Avona, Mound Laboratory, Miamisburg, Ohio, personal communication.

20. T. F. Johns and J. Bigeleisen, p. 488 in *Separation of Isotopes*, ed. H. L. London, George Newnes, London, 1961.

21. T. I. Taylor and W. Spindel, "Preparation of Highly Enriched Nitrogen-15 by Chemical Exchange of NO with HNO_3," Chap. 10 in *Proceedings of the International Symposium on Isotope Separation, Amsterdam, 1957*, ed. J. Kistemaker et al., North Holland, Amsterdam, 1958.

22. W. Spindel and T. I. Taylor, "Separation of Nitrogen Isotopes by Chemical Exchange Between NO and HNO_3," *J. Chem. Phys.* 23, 981 (1955).

23. D. A. Hayford, W. N. Johnson, S. A. Levin, J. Shorter, and E. Von Halle, *Feasibility of Large-Scale Nitrogen-15 Production for Nuclear Reactors*, K-1232, Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tenn. (August 1955).

24. K. A. Williams and S. Blumkin, Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tenn., personal communication on 1972 plant analysis.

25. E. Michaels and R. Schwind, Mound Laboratory, Miamisburg, Ohio, personal communication, May 1976, Mound Technical Report (in press).

26. H. London, "Isotope Separation by Fractional Distillation," Chap. 25 in *Proceedings of the International Symposium on Isotope Separation, Amsterdam, 1957*, ed. J. Kistemaker et al., North Holland, Amsterdam, 1958.