

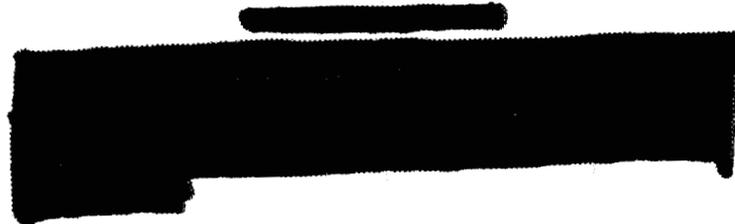


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# Potential Generation and Radiological Impacts of Gaseous <sup>14</sup>C Released During Reprocessing of Advanced LMFBR Fuels

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ENVIRONMENTAL SCIENCES DIVISION  
METALS AND CERAMICS DIVISION

POTENTIAL GENERATION AND RADIOLOGICAL IMPACTS OF GASEOUS  $^{14}\text{C}$   
RELEASED DURING REPROCESSING OF ADVANCED LMFBR FUELS

V. J. Tennery, E. S. Bomar, W. D. Bond, S. V. Kaye,  
L. E. Morse and J. E. Till

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ABSTRACT

Use of advanced carbide or nitride fuels in place of the reference oxide fuel in LMFBRs may be required in the future to optimize the utilization of this reactor system. Differences in the environmental impact associated with such fuel substitutions are of critical importance in determining the real future viability of various advanced fuels. Calculations indicate that the concentration of <sup>14</sup>C can be significantly different in the various fuels following their residence in the reactor. The major source of <sup>14</sup>C generation in mixed oxide, carbide, and nitride fuels is <sup>14</sup>N. Most of the <sup>14</sup>C present in the spent fuels is converted to <sup>14</sup>CO<sub>2</sub> during fuel reprocessing, and <sup>14</sup>C released to the environment will be in this form. Best estimates indicate a release to the environment of no more than 1% of the <sup>14</sup>C contained in the spent fuels.

Radiation dose commitments due to <sup>14</sup>CO<sub>2</sub> release from advanced-fuel reprocessing plants were calculated and compared with that previously determined for the reference oxide on the basis of 50 GW(e)-years of energy generation. The primary source of exposure to the maximally exposed individual is ingestion of food, which contributes greater than 99% of the 50-year dose commitment. Dose commitments to the total body range between  $1.5 \times 10^{-5}$  mrem from nitride fuel containing no <sup>14</sup>N to a maximum of 50 mrem from nitride fuel synthesized with natural nitrogen. This calculated dose may be compared with the natural background exposure from <sup>14</sup>C, which was estimated by the United Nations to be 1.3 mrem to an average individual in the general population.

Population doses within an 80-km (50-mile) radius of the reprocessing plant were calculated to be 13 man-rems for carbide fuel containing 1000 ppm <sup>14</sup>N and 760 man-rems for nitride fuels synthesized from natural nitrogen. The dose commitment to the same population from natural <sup>14</sup>C is estimated to be 1300 man-rems. Ultimate assessment of the significance of the dose estimated from <sup>14</sup>C cannot be determined until doses resulting from all other radionuclides released from the reprocessing plant have been calculated.

The radiological impact of <sup>14</sup>C can be reduced by ensuring low <sup>14</sup>N concentrations in fabricated fuels and/or significant improvements in the confinement factor of 100 applicable to current fuel reprocessing plant technology.

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## INTRODUCTION

Potential environmental, economic, and sociological effects of LMFBRs containing the reference oxide fuel were described and evaluated in U.S. Government documents.<sup>1,2</sup> These studies were done in compliance with the requirements of the National Environmental Policy Act of 1969. Generation of  $^{14}\text{C}$  in the reference oxide fuel is discussed in ref. 2. The work reported here was done as part of an ongoing program supported by the Reactor Development and Demonstration Division of ERDA to determine the environmental consequences of substituting carbide or nitride fuel for the reference oxide fuel in LMFBRs. The amount of  $^{14}\text{C}$  present in spent fuel is of particular interest in the fuel reprocessing step of the LMFBR fuel recycle since this is the point in the fuel cycle where  $^{14}\text{C}$  is most likely to be released to the environment.

The  $^{14}\text{C}$  content of spent fuel depends on the degree of burnup at discharge from the reactor. A limiting value of burnup of a fuel depends on the properties of the fuel and the fuel management procedures followed; therefore, we have selected the amount of a given fuel necessary to generate 50 GW-years of electric energy as a basic unit of comparison rather than use a unit of mass. A thermal-to-electric conversion efficiency of 41% is assumed for this study.

Carbon-14 is a beta emitter (0.156 MeV max) with a radioactive half-life of  $5745 \pm 50$  years. It is found in naturally occurring carbon at a concentration of about one part in  $10^{12}$  and is produced by interaction of cosmic-ray neutrons with atmospheric nitrogen.<sup>3</sup> The concentration of  $^{14}\text{C}$  in deeply buried reservoirs of carbon, such as coal and oil, is much lower. Because of this, the ratio of  $^{14}\text{C}/^{12}\text{C}$  in the atmosphere decreased (2 to 3%) during the industrial revolution as a result of the generation of large quantities of  $\text{CO}_2$  during the combustion of fossil fuels. During the 1960s the  $^{14}\text{C}/^{12}\text{C}$  ratio was increased by 10% as a result of nuclear testing in the atmosphere.<sup>3</sup>

1. THE PRODUCTION OF  $^{14}\text{C}$  IN LMFBR FUELS

## 1.1 INTRODUCTION

Experimentally measured levels of  $^{14}\text{C}$  production in LMFBR fuels are not available, so this information was calculated with the ORIGEN<sup>4</sup> computer code. ORIGEN is an isotope generation and depletion code and is capable of providing an inventory of several hundred fission products and transmutation products, including the actinides through  $^{245}\text{Cm}$ . ORIGEN requires input data on spectrum-averaged cross sections for the nuclear reactions of interest, the initial composition of the core loading, and the core management schedule.

## 1.2 OXIDE FUEL

Various neutron-induced reactions resulting in the production of  $^{14}\text{C}$  in reference-oxide fuel and stainless-steel cladding were reviewed<sup>2</sup>

in ERDA-1535. The LMFBR model assumed for the reference oxide review was the Atomics International Follow-On Design.<sup>5</sup> The energy-dependent cross sections for several reactions that yield  $^{14}\text{C}$  were combined into single effective cross sections. These cross sections were incorporated into the ORIGEN code. The code was then used, along with some hand calculations, to calculate the  $^{14}\text{C}$  yield. The  $^{14}\text{C}$  produced in the  $(\text{U,Pu})\text{O}_2$  fuel of the reference LMFBR<sup>2</sup> is given in Table 1. The assumed carbon and nitrogen levels in the fuel were 20 ppm. The contribution of the  $^{13}\text{C}(n,\gamma)^{14}\text{C}$  reaction was inconsequential, whereas the  $^{14}\text{N}(n,p)^{14}\text{C}$  reaction produces most of the  $^{14}\text{C}$  activity in reference oxide fuel; therefore, the amount of  $^{14}\text{C}$  produced is primarily a function of the amount of  $^{14}\text{N}$  present.

Table 1. Production of  $^{14}\text{C}$  in Reference LMFBR  $(\text{U,Pu})\text{O}_2$  Fuel<sup>a</sup>

Reaction	Reaction Cross Section (mb)	$^{14}\text{C}$ Activity	
		(Ci/metric ton) <sup>b</sup>	(% of Total $^{14}\text{C}$ Activity)
$^{13}\text{C}(n,\gamma)^{14}\text{C}$	0.0005	$9.62 \times 10^{-8}$	$4.6 \times 10^{-5}$
$^{14}\text{N}(n,p)^{14}\text{C}$	12.6	0.202	96
$^{15}\text{N}(n,d)^{14}\text{C}$	1.0	$5.70 \times 10^{-5}$	$2.7 \times 10^{-2}$
$^{16}\text{O}(n,^3\text{He})^{14}\text{C}$	0.00005	$4.53 \times 10^{-3}$	2.5
$^{17}\text{O}(n,\alpha)^{14}\text{C}$	0.12	$4.03 \times 10^{-3}$	1.9
TOTAL		0.211	~100

<sup>a</sup> Assumed impurity content: C = 20 ppm and natural nitrogen (99.64%  $^{14}\text{N}$ ) = 20 ppm.

<sup>b</sup> Metric ton = metric ton of heavy metal content. Heavy metal content of  $\text{U}_{0.8}\text{Pu}_{0.2}\text{O}_2 = 88.16\%$  by weight. Average burnup of fuel (driver plus blankets) was 37,100 MW(t)d/metric ton.

### 1.3 CARBIDE FUEL

Carbon-14 generation in advanced LMFBR carbide fuels was examined in this study using core loading, isotopic compositions, and burnup data taken from a 5000 MW(t) advanced LMFBR design prepared at Argonne National Laboratory.<sup>6</sup> In the absence of specific neutron-spectrum information for the carbide-fueled core the cross-section data used in the reference oxide analysis were also used in this investigation. Although use of cross-section data derived for an oxide core will result in some error, we feel that the error will be small.

Two calculations were made with the ORIGEN computer code to assess the  $^{14}\text{C}$  content of the spent carbide fuel. The first assumed no nitrogen impurity in the (U,Pu)C fuel and the second assumed 25 ppm of natural nitrogen impurity. The result of the latter computation was extrapolated to the equivalent of 1000 ppm of nitrogen, which is presently a proposed limit for this impurity.<sup>7</sup> The results are given in Table 2 in terms of  $^{14}\text{C}$  content per metric ton of heavy metal in carbide fuel.

Table 2. Production of  $^{14}\text{C}$  in Advanced LMFBR (U,Pu)C Fuel<sup>a</sup>

Nitrogen Content (ppm)	Reaction	Reaction Cross Section (mb)	$^{14}\text{C}$ Activity	
			(Ci/metric ton) <sup>b</sup>	(% of Total $^{14}\text{C}$ Activity)
0	$^{13}\text{C}(n,\gamma)^{14}\text{C}$	0.0005	$2.39 \times 10^{-4}$	100
1000	$^{13}\text{C}(n,\gamma)^{14}\text{C}$	0.0005	$2.39 \times 10^{-4}$	$2.6 \times 10^{-3}$
	$^{14}\text{N}(n,p)^{14}\text{C}$	12.6	9.07	~100

<sup>a</sup>Fuel may contain up to 10% by volume of the sesquicarbide phase, nitrogen impurity as indicated.

<sup>b</sup>Metric ton = metric ton of heavy metal content; heavy metal content of  $\text{U}_{0.8}\text{Pu}_{0.2}\text{C} = 95.20\%$  by weight. Average burnup of fuel (driver plus blankets) was 29,600 MWd/metric ton.

#### 1.4 NITRIDE FUEL

A 5000-MW(t) advanced LMFBR design prepared by Argonne National Laboratory was also used in conjunction with ORIGEN calculations to determine the  $^{14}\text{C}$  produced in cores having nitride fuel. The core loading data and burnup were unique to the nitride core, but again the nuclear cross-section information used in the reference oxide LMFBR analysis was employed. The effect on  $^{14}\text{C}$  generation of substituting  $^{15}\text{N}$  for  $^{14}\text{N}$  in the fuel was also calculated. The results for fuel represented by one metric ton of heavy metal containing natural nitrogen or enriched with  $^{15}\text{N}$  are given in Table 3.

#### 1.5 CLADDING AND OTHER METALLIC PARTS OF FUEL ELEMENTS

Calculations similar to those made for the fuel materials were also obtained for the stainless steel associated with one metric ton of the oxide fuel.<sup>2</sup> The results, listed in Table 4, were obtained for typical carbon and nitrogen contents of 500 ppm and 45 ppm, respectively.

Table 3. Production of  $^{14}\text{C}$  in Advanced LMFBR (U,Pu)N Fuel

Enrichment Level of Fuel in $^{15}\text{N}$ (%)	$^{14}\text{N}$ in Fuel (%)	Reaction	Reaction Cross Section (mb)	$^{14}\text{C}$ Activity	
				(Ci/metric ton) <sup>a</sup>	(% of Total $^{14}\text{C}$ Activity)
0.36 <sup>b</sup>	5.53	$^{14}\text{N}(n,p)^{14}\text{C}$	12.6	521	100
90	0.55	$^{14}\text{N}(n,p)^{14}\text{C}$	12.6	52.3	100
99	0.055	$^{14}\text{N}(n,p)^{14}\text{C}$	12.6	5.23	100
100	0	$^{15}\text{N}(n,2n)^{14}\text{N}$	0.0022		0
		$^{14}\text{N}(n,p)^{14}\text{C}$	12.6	$1.11 \times 10^{-4}$	100

<sup>a</sup>Metric ton = metric ton of heavy metal content; heavy metal content of  $\text{U}_{0.8}\text{Pu}_{0.2}\text{N} = 94.45\%$  by weight. Average burnup of fuel (driver plus blankets) was 29,400 MWd/metric ton.

<sup>b</sup> $\text{U}_{0.8}\text{Pu}_{0.2}\text{N}$  contains 5.554% N by weight, of which 99.64% by weight of natural nitrogen is  $^{14}\text{N}$ .

Table 4. Production of  $^{14}\text{C}$  in Stainless Steel Components<sup>a</sup> of Reference Oxide LMFBR Fuel Elements

Reaction	Reaction Cross Section (mb)	$^{14}\text{C}$ Activity	
		(Ci/metric ton) <sup>b</sup>	(% of Total $^{14}\text{C}$ Activity)
$^{13}\text{C}(n,\gamma)^{14}\text{C}$	0.0005	$1.30 \times 10^{-6}$	$3.05 \times 10^{-5}$
$^{14}\text{N}(n,p)^{14}\text{C}$	12.6	0.245	~100
$^{15}\text{N}(n,d)^{14}\text{C}$	1.0	$6.92 \times 10^{-5}$	$2.94 \times 10^{-2}$
TOTAL		0.246	~100

<sup>a</sup>Natural contents assumed to be 500 ppm C and 45 ppm N.

<sup>b</sup>Amount of  $^{14}\text{C}$  activity generated in stainless steel associated with one metric ton of fuel; metric ton = metric ton of heavy metal content. Heavy metal content of  $\text{U}_{0.8}\text{Pu}_{0.2}\text{O}_2 = 88.16\%$  by weight; ratio of stainless steel/heavy metal in fuel = 0.54. Average burnup of fuel (driver plus blankets) was 37,100 MWd/metric ton.

Comparison of  $^{14}\text{C}$  values from Tables 1 and 4 shows that the amount of  $^{14}\text{C}$  generated in the stainless steel components is comparable with that produced in the reference oxide fuel. Mixed carbide fuel containing no  $^{14}\text{N}$  will produce about three orders of magnitude less activity from  $^{14}\text{C}$  than would be produced in the associated cladding and duct components. A relatively small  $^{14}\text{N}$  level in the fuel will, however, result in  $^{14}\text{C}$  activity comparable with that from the metal components of the core structure.

## 2. REPROCESSING OF ADVANCED LMFBR FUELS

### 2.1 INTRODUCTION

The distribution of  $^{14}\text{C}$  in gaseous, liquid, and solid process streams as a result of the use of oxidation in the head-end treatment and the aqueous chemical reprocessing of spent advanced LMFBR fuels is considered. However, a source term for assessment of the environmental impact of gaseous effluent alone is presented. Disposition of liquid and solid process waste streams are not considered in the report.

Conceptual flowsheets for reprocessing carbide and nitride fuels and for treating the off-gas streams are shown in Figs. 1 and 2, respectively. The darkened areas in the flowsheets depict the principal differences between the reprocessing of oxide fuels and advanced fuels. These flowsheets are based on the limited amount of small-scale hot-cell work and cold laboratory work found in the literature.

The principal basis for the flowsheet in regard to  $^{14}\text{C}$  removal is an oxidation step at 450 to 600°C using oxygen. In this oxidation step, the internal sodium is deactivated and the nitride or carbide is burned to the metal oxide (Fig. 1). The  $\text{CO}_2$  resulting from the oxidation is then separated from the other radioactive gases and is converted to solid  $\text{CaCO}_3$  for storage. In contrast, current flowsheets for oxide fuel reprocessing use a dilute nitric acid wash to deactivate the internal sodium, and then the washed fuel is dissolved in a more concentrated solution of nitric acid.<sup>1</sup>

### 2.2 SIGNIFICANT DIFFERENCES BETWEEN REPROCESSING OXIDE FUEL AND ADVANCED FUEL

The principal difference in the reprocessing of the advanced LMFBR fuels compared with the oxide fuels is that the oxide fuels (after removal of internal sodium) may be dissolved directly in nitric acid to prepare satisfactory solvent extraction feed solutions. Aqueous dissolution of carbide fuels results in the formation of soluble organic compounds, which, despite additional treatment of the solution, may interfere with both the plutonium recovery and process operability in solvent extraction.<sup>8</sup> Further, the presence of these organic compounds would increase the retention of  $^{14}\text{C}$  and contamination of the main process streams by  $^{14}\text{C}$ . No data could be found in the literature to demonstrate that satisfactory solvent extraction feed could be prepared by treating the soluble organic compounds with chemical reagents, such as permanganate, chromate, peroxide, or ozone.<sup>9,10</sup>

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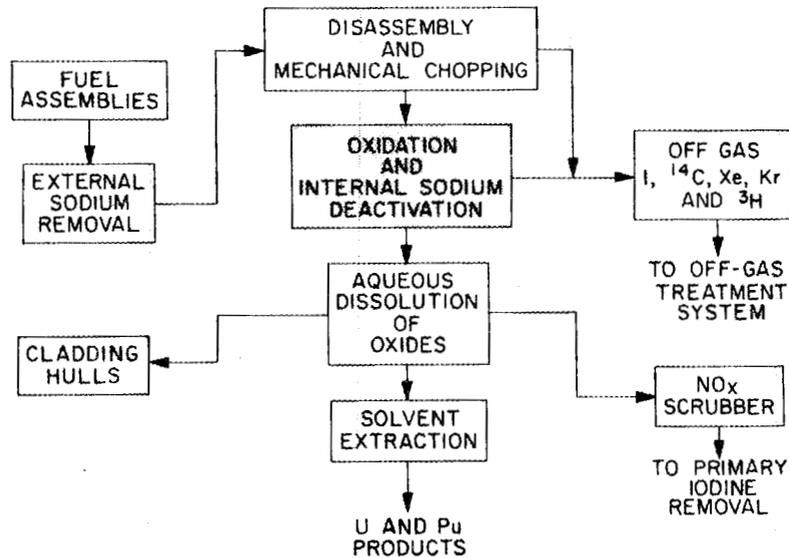


Fig. 1. Conceptual Flow Diagram for the Aqueous Chemical Processing of Advanced Nuclear Fuels.

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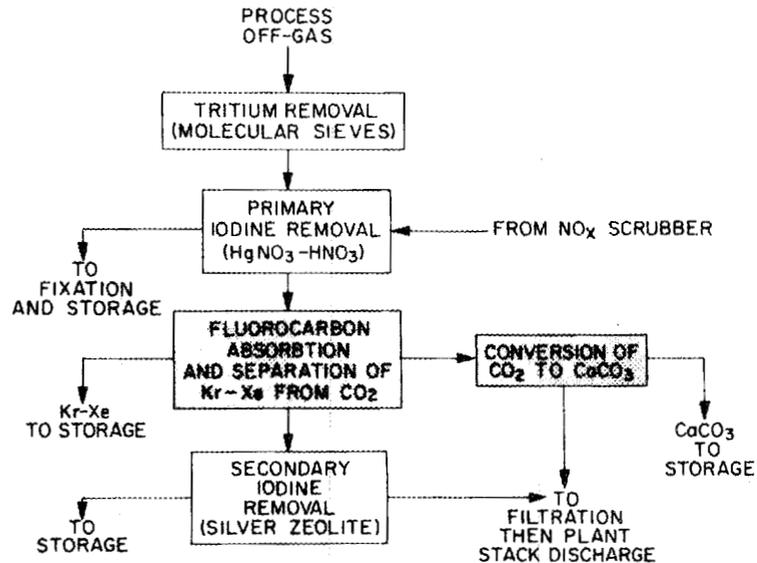


Fig. 2. Conceptual Flow Diagram for Treating Process Off-Gas Stremas from the Aqueous Chemical Processing of Advanced Nuclear Fuels.

If carbonitride fuels were considered for use, direct dissolution of carbonitride fuels is likely to encounter the same problems in the removal of soluble organic compounds.<sup>11,12</sup> Additional development work that clearly demonstrates that soluble organic compounds can be destroyed or removed to make a satisfactory solvent extraction feed is required before direct dissolution can be considered a viable method for use in the reprocessing of spent carbide or carbonitride fuels.

Direct dissolution of spent nitride fuels in nitric acid is feasible for producing satisfactory solvent extraction feed solutions.<sup>13,14</sup> However, if the nitride fuel is prepared with nitrogen enriched in <sup>15</sup>N, recovery of this valuable material would be hindered by isotopic exchange reactions. If natural nitrogen is used, the contamination of the main process streams is increased. Therefore, while the head-end oxidation of spent nitride fuels appears to be optional, it appears to be necessary in the case of carbide or carbonitride fuels.

Oxidation of advanced fuels to oxides before dissolution is a significant departure from the more conventional method of direct dissolution that is employed for oxide fuels. Nevertheless, it is not significantly different from advanced concepts for reprocessing oxide fuels (see Sect. 2.4). The theoretical feasibility of fuel oxidation has been established in small-scale tests in the laboratory and in hot cells. However, practical methods for carrying out the oxidation and dissolution in remote operations are yet to be established. Although not conclusively demonstrated, corrosion considerations probably preclude carrying out the dissolution in the same vessel that is used for the fuel oxidation. Problems related to transfer of the oxide and cladding to a subsequent dissolution vessel would require evaluation on both the laboratory and engineering scale. However, the oxidation step does have the potential advantage that more than 99% of the carbon would be removed in a single step and the distribution of <sup>14</sup>C throughout other process streams would be significantly reduced.

### 2.3 ISOLATION OF <sup>14</sup>C DURING ADVANCED-FUEL REPROCESSING AND ITS RELEASE FROM A FUEL REPROCESSING FACILITY

The movement of <sup>14</sup>C between the gaseous, liquid, and solid process streams within the fuel reprocessing plant is estimated in this section, and these estimates are used to obtain a <sup>14</sup>C source term for the gaseous waste stream leaving the plant. For our purposes here, waste streams are designated as those leaving the boundary of the reprocessing plant, while various flows within the plant boundary are defined as process flow streams. The disposition of solid wastes containing <sup>14</sup>C from this plant is not considered in this report. The fixation of <sup>14</sup>C in a solid waste form suitable for long-term isolation is presently the object of experimental work at ORNL. As this technology is developed and demonstrated, a realistic assessment of the <sup>14</sup>C release associated with the isolated solid waste from the plant will be possible. For the purposes of this report, releases of <sup>14</sup>C resulting from this waste stream are assumed to be negligible.

## 2.4 REMOVAL OF $^{14}\text{C}$ IN HEAD-END REPROCESSING

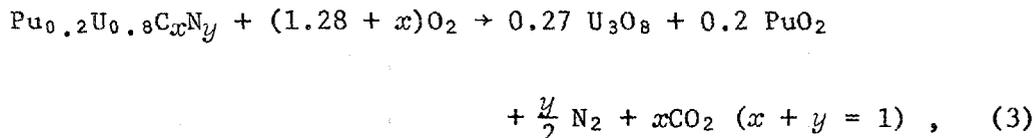
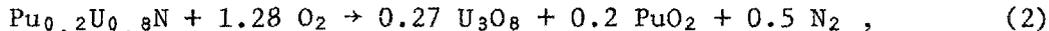
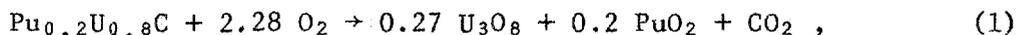
Current concepts of the head-end processing of spent LMFBR fuels consist of the following sequence of operations:

1. dismemberment of fuel assemblies by shearing after external sodium removal;
2. oxidation by heating sheared fuel pieces in oxygen to volatilize tritium, the fission noble gases, and  $^{14}\text{C}$  as  $^{14}\text{CO}_2$ , and to deactivate the internal sodium;
3. dissolution of the sheared and oxidized fuel in  $\text{HNO}_3$ .

Application of this sequence in advanced fuels reprocessing indicates that  $^{14}\text{C}$  may be volatilized as  $^{14}\text{CO}_2$  during the second and third operations but not during the first step because both fuel and cladding retain the carbon during mechanical segmenting.<sup>15,16</sup> The balance of this section evaluates the disposition of  $^{14}\text{C}$  in the oxidation and dissolution steps.

### 2.4.1 Oxidation and Volatilization

Carbon-14 contained in the spent advanced nuclear fuel is converted to  $^{14}\text{CO}_2$  and volatilized from the sheared fuel pieces in the course of the following idealized oxidation reactions at elevated temperatures:



where, more approximately, in Eq. (2) the mole ratio of  $\text{C}/(\text{N} + \text{C}) \approx 0.004$  because of  $^{14}\text{C}$  generation in mononitride fuel at a burnup of 29,400 MWd/metric ton.

The small amount of information on the oxidation of irradiated advanced fuels is available from laboratory-scale experiments with both irradiated uranium-plutonium monocarbide and uranium monocarbide. Oxidation of a mixed 20% PuC-80% UC sample (30,000 MWd/metric ton U + Pu burnup) at 450°C in flowing oxygen resulted<sup>17</sup> in the quantitative conversion of the carbon in the carbide to  $\text{CO}_2$ . Oxidation-reduction ( $\text{UC}-\text{U}_3\text{O}_8-\text{UO}_2$ ) cycles on two samples of irradiated UC (~12,500 and ~6,500 MWd/metric ton burnups) indicated 100% conversion of UC to  $\text{UO}_2$ . The oxidations were accomplished<sup>18</sup> in flowing air by heating to 600°C. Further information on the oxidation behavior of carbide fuels was obtained from laboratory studies on the oxidation of nonirradiated UC or PuC. Careful studies showed that a small amount of carbon was retained by the oxidation product of UC when oxidized at 600°C in flowing oxygen,<sup>19</sup> possibly

trapped or sorbed as carbon dioxide.<sup>20</sup> The ready oxidation of PuC in air at 300°C and in oxygen at 400°C has been reported.<sup>21</sup> The oxidation behavior of unirradiated nitride fuels was derived from laboratory-scale studies on the mononitrides of uranium and plutonium. The oxidation of UN in dry oxygen begins at about 250°C and at about 600°C conversion to U<sub>3</sub>O<sub>8</sub> is complete.<sup>22</sup> The oxidation of PuN to PuO<sub>2</sub> in dry oxygen at 260°C is slower than that of UN but is catalyzed by traces of water vapor.<sup>23</sup> Retention of nitrogen in both oxidation products was noted, but there is disagreement as to the final temperature required for its release. Information about the retention of carbon was not obtained from these experiments since these materials were carbon-free. Laboratory experiments on the oxidation of nonirradiated uranium carbonitrides (UC<sub>0.97</sub>N<sub>0.03</sub>, UC<sub>0.95</sub>N<sub>0.05</sub>, UC<sub>0.90</sub>N<sub>0.10</sub>, and UC<sub>0.80</sub>N<sub>0.20</sub>) by air provide information relative to the oxidation of carbonitride fuels.<sup>24</sup> The behavior of UC<sub>1-x</sub>N<sub>x</sub> was similar to that of UC and UN. The oxidation started at 260 to 310°C, and conversion to U<sub>3</sub>O<sub>8</sub> was complete above 500°C. Retention of carbon by the UC<sub>1-x</sub>N<sub>x</sub> oxidation products was not mentioned; however, some retention of both carbon and nitrogen may be anticipated from the results with UC and UN. The extent of volatilization of <sup>14</sup>C from the stainless steel cladding during oxidation is not known.

The radioactive constituents of the oxidation off-gases resulting from this process step will be HTO, <sup>14</sup>CO<sub>2</sub>, particulates, and the fission noble gases (Kr and Xe). Other gases will be either CO<sub>2</sub> (carbide fuels), N<sub>2</sub> (nitride fuels), or a mixture of both (carbonitride fuels) and excess O<sub>2</sub>. Also, solids will retain some of the gaseous oxidation products. The off-gases after cleanup to remove HTO and particulates are routed to the process off-gas system.

#### 2.4.2 Aqueous Dissolution of Oxidized Fuel

The solids from the oxidation process, stainless steel cladding and the heavy metal and fission product oxides, are transferred to the dissolver. Under the prevailing oxidizing conditions the acid (HNO<sub>3</sub>) will dissolve the oxides and about 1% of the stainless steel cladding,<sup>2</sup> liberating the <sup>14</sup>C trapped in the oxide solids and in the dissolved cladding as <sup>14</sup>CO<sub>2</sub>. Cladding dissolution will contribute about 2 Ci <sup>14</sup>C per year to the total <sup>14</sup>C contained in the process stream within the reprocessing plant. The radioactive constituents in the dissolver off-gases may be <sup>14</sup>CO<sub>2</sub>, I<sub>2</sub>, Kr, Xe, and particulates; the other gases will include NO<sub>x</sub>, O<sub>2</sub>, and, depending on the fuel composition, either CO<sub>2</sub>, N<sub>2</sub>, or a mixture of both. The dissolver off-gas, after treatment to remove some of the NO<sub>x</sub> and I<sub>2</sub>, is routed into the process off-gas system. The balance of the <sup>14</sup>C in the stainless steel cladding, approximately 99%,<sup>2</sup> not released in dissolution will remain trapped in the metal and is managed as a separate solid waste. An additional quantity of CO<sub>2</sub> will be soluble in the dissolver solution; however, it is difficult to estimate this concentration because of the unknown effects of the I<sub>2</sub> removal step on the concentration of dissolved CO<sub>2</sub> and the lack of CO<sub>2</sub> solubility data for dissolver solution conditions.

It is reasonable to assume that solubility of CO<sub>2</sub> will not be greater than that in water at the same temperature because of the high electrolyte concentrations in the dissolver solutions (see Sect. 2.6). The solubility of CO<sub>2</sub> in the dissolver solution provides a potential path for the introduction of <sup>14</sup>C into the solvent extraction system.

## 2.5 TECHNIQUE FOR <sup>14</sup>C CONTAINMENT DURING FUEL REPROCESSING

The process vessel off-gas flow is estimated to be about 0.47 m<sup>3</sup>/sec (1000 cfm) for a chemical plant processing oxide fuel at the rate of 5 metric tons/day of equivalent heavy metal content.<sup>25</sup> The additional gas generated by reprocessing the advanced nuclear fuels through an oxidation procedure is calculated to be approximately 5.7 liters/sec (12 cfm) CO<sub>2</sub> for carbides, approximately 2.8 liters/sec (6 cfm) N<sub>2</sub> for nitrides, and an intermediate value for CO<sub>2</sub> + N<sub>2</sub> for carbonitrides. Therefore, the maximum contribution from these sources would be less than 1.5% of the total process off-gas volume. The process off-gas would be decontaminated by selective absorption in a fluorocarbon solvent before venting to the atmosphere. The retained gases are fractionated, and the radioactive fractions are placed in permanent storage.

### 2.5.1 Fluorocarbon Absorption Process

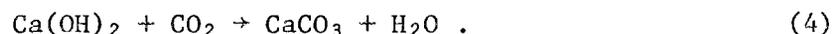
Selective absorption in fluorocarbon solvents is a processing technique for the decontamination of gaseous effluent streams from nuclear facilities. It utilizes the differences in gas-liquid solubilities that exist between the solvent and the various gas constituents to effect their separation.<sup>26</sup> Pilot-plant-scale development and demonstration of this process have been in progress since 1968. Tests have shown that a fluorocarbon process designed to remove 99.9% of the off-gas krypton can also be relied upon to remove and concentrate at least that much <sup>14</sup>C present as CO<sub>2</sub>. Although the potential capability of the basic process needs to be better defined, removal of 99% of the CO<sub>2</sub> appears to be readily obtainable. The absorption process product stream contains CO<sub>2</sub>, N<sub>2</sub>O, the noble gases (Kr and Xe), and some O<sub>2</sub>. The less soluble decontaminated gases are vented to the atmosphere after filtration to remove any entrained particles. Further testing of this process is now proceeding in a pilot facility of one-fifth to one-sixth the scale of a unit required for a 5-metric ton/day fuel processing plant.

### 2.5.2 Separation of <sup>14</sup>CO<sub>2</sub>-Kr

The primary purpose of this separation is to alleviate some of the problems associated with the permanent storage of <sup>85</sup>Kr and <sup>14</sup>C. Separations based on cold trapping and use of molecular sieves are being developed to fractionate the absorption-process product stream.<sup>26</sup>

### 2.5.3 Fixation of $^{14}\text{C}$ as $\text{CaCO}_3$

The  $^{14}\text{C}$  is prepared for final storage by converting  $\text{CO}_2$  into a solid compound. The compound selected is  $\text{CaCO}_3$ , which is precipitated by the reaction of  $\text{CO}_2$  with an aqueous  $\text{Ca}(\text{OH})_2$  slurry:<sup>27</sup>



In principle this should be a highly efficient method to fix the  $^{14}\text{C}$  since the calculated pressure of  $\text{CO}_2(\text{g})$  in equilibrium with a solution saturated with both  $\text{CaCO}_3$  and  $\text{Ca}(\text{OH})_2$  is about  $10^{-9}$  Pa ( $10^{-14}$  atm) and the calculated carbon concentration in liquid phase due to soluble  $\text{CaCO}_3$  is about  $3 \times 10^{-6}$  g/liter.<sup>28</sup> The precipitate is filtered off and prepared for final disposal, and the filtrate is recycled. The off-gas also may be recycled if necessary. Engineering development for the fixation of  $^{14}\text{C}$  by this process is already under way in the HTGR program.

### 2.6 ESTIMATION OF THE $^{14}\text{C}$ CONTENT IN LIQUID PROCESS STREAMS INTERNAL TO THE REPROCESSING PLANT

The principal source of  $^{14}\text{C}$  in liquid streams is assumed to be the dissolved  $\text{CO}_2$  in the fuel solution leaving the fuel dissolver. The  $^{14}\text{C}$  concentration in the aqueous process stream may be estimated if it is assumed that all the  $^{14}\text{C}$  entering the solvent extraction system as a result of the solubility of  $\text{CO}_2$  in the fuel dissolver solution is retained in the aqueous stream. On the premise that the  $\text{CO}_2$  solubility will not be greater than that in water at  $100^\circ\text{C}$  ( $\approx 1.3 \times 10^{-5}$  M), and that the volume of concentrated aqueous waste placed in temporary storage is 5000 liters/day,<sup>25</sup> an upper limit for the  $^{14}\text{C}$  content was calculated. On an annual basis, that is processing spent fuel used to generate 50 GW-year of electrical energy, the stored wastes would contain 43 mCi  $^{14}\text{C}$  for a carbide fuel with a 1000 ppm  $^{14}\text{N}$  impurity and 1091 Ci  $^{14}\text{C}$  for a nitride fuel prepared with natural nitrogen. The difference in contents results from the dilution of  $^{14}\text{C}$  by natural carbon in the carbide fuel (see Sect. 1.0). The need for an experimental program to determine the actual  $^{14}\text{C}$  contents in these wastes and methods for their reduction to acceptable levels, where necessary, is indicated.

Sufficient data are not presently available to define the quantity and chemical form of  $^{14}\text{C}$  trapped (or sorbed) by the oxide as a result of the oxidation of the spent fuel. Residual  $^{14}\text{C}$  in the oxide will be the determining factor for the  $^{14}\text{C}$  content of the process streams from subsequent aqueous reprocessing. Although it may be theoretically possible to convert the residual carbon to gaseous species during and after aqueous dissolution of the oxide and to collect the gases by absorption processes, quantitative data for such processes have not been obtained. The limitations of practical processes will preclude complete separation of  $^{14}\text{C}$  from the liquid process streams. Therefore, experimental work is required to define the  $^{14}\text{C}$  decontamination factors that can be realistically achieved for liquid process streams.

2.7 ESTIMATE OF  $^{14}\text{C}$  SOURCE TERM FOR GASEOUS EFFLUENT

Estimates of the  $^{14}\text{C}$  content of the gaseous effluent from an LMFBR fuel processing plant are based on the levels of production reported in Sect. 1 of this report and the assumption that 99% (confinement factor = 100) of the  $\text{CO}_2$  generated during reprocessing is removed from the off-gas stream by the fluorocarbon selective absorption process previously discussed. A comparison of the  $^{14}\text{C}$  produced in the oxide fuel with that in the carbide and nitride fuels on the basis of a metric ton of heavy metal processed does not take into account the difference in burnup of the fuels. Production of  $^{14}\text{C}$  was therefore based on processing an amount of fuel equivalent to 50 GW-year of electricity and assuming a thermal-to-electric conversion efficiency of 41% for the power plant. This is equivalent to the energy generated in 1500 metric tons of heavy metal at an average burnup of 29,500 MWd. The resulting source terms are given in Table 5.

Table 5. Carbon-14 Content of Gaseous Effluent from LMFBR Fuel Processing Plant<sup>a</sup>

Fuel	Concentration of $^{14}\text{N}$ in Fuel (%)	Radioactivity Released <sup>b</sup> [Ci/50 GW(e)-year]
Reference oxide <sup>c</sup>	0.002	2.5
Carbide <sup>d</sup>	0	$3.58 \times 10^{-3}$
	0.1	136
Nitride <sup>e</sup>	99.64 (natural N)	$7.82 \times 10^3$
	10	785
	1	78.5
	0	$1.68 \times 10^{-3}$

<sup>a</sup>A confinement factor of 100 assumed for  $^{14}\text{C}$ . Confinement Factor = Ci Processed Through Plant/Ci Released.

<sup>b</sup>Release based on reprocessing an amount of fuel equivalent to 50 GW-year of electricity, assuming a thermal-to-electric conversion efficiency of 41%.

<sup>c</sup>Blend of  $\text{UO}_2$  and  $(\text{U,Pu})\text{O}_2$  fuel (driver plus blankets). Average burnup: 37,100 MWd/metric ton of heavy metal.

<sup>d</sup>Blend of UC and  $(\text{U,Pu})\text{C}$  fuel pellets (driver plus blankets) irradiated in a reactor presumed to have same neutronics as the GE Follow-On Reactor. Average burnup; 29,610 MWd/metric ton of heavy metal.

<sup>e</sup>Blend of UN and  $(\text{U,Pu})\text{N}$  fuel pellets (driver plus blankets) irradiated in a reactor presumed to have the same neutronics as the GE Follow-On Reactor. Average burnup; 29,406 MWd/metric ton of heavy metal.  $(\text{U,Pu})\text{N}$  contains 5.554% N by weight.

### 3. RADIOLOGICAL IMPACT OF $^{14}\text{C}$

#### 3.1 INTRODUCTION

Carbon is a major constituent of all living cells, and because of this, radiocarbon is taken up readily by biochemical processes regulating carbon metabolism. The uptake of  $^{14}\text{C}$  by living organisms depends upon the amount of carbon present, since cells do not distinguish radioactive from stable atoms of carbon. For the purposes of this report, releases of  $^{14}\text{C}$  from the model fuel reprocessing facility are assumed to be gaseous  $^{14}\text{CO}_2$ . By assuming that all of the  $^{14}\text{C}$  released from the reprocessing plant will be in the form of  $^{14}\text{CO}_2$ , we are choosing the chemical form that will result in maximum  $^{14}\text{C}$  uptake by biota.

The released  $^{14}\text{CO}_2$  will become quickly diluted by the stable  $\text{CO}_2$  in the atmosphere. The ratio of radioactive to stable atoms of carbon is an important relationship for the environmental assessment of releases. It is usually referred to as the specific activity and is written:

$$\text{SA} = \frac{\mu\text{Ci } ^{14}\text{C}}{\text{g } ^{14}\text{C} + \text{g } ^{12}\text{C}} \approx \frac{\mu\text{Ci } ^{14}\text{C}}{\text{g } ^{12}\text{C}}, \quad (5)$$

where

SA = specific activity,

$\mu\text{Ci}$  = microcuries of radioactivity, and

g = grams of carbon.

The specific activity concept is used in this study to assess the consequences of  $^{14}\text{C}$  releases to the environment.

The initial behavior of this  $^{14}\text{CO}_2$  in the local atmospheric environment will depend on several factors, including the height of the off-gas stack, the quantity of heat released with the  $^{14}\text{CO}_2$ , and the local meteorological conditions. These parameters are usually specified in atmospheric diffusion models of the type used to calculate the concentration and/or specific activity of  $^{14}\text{C}$  at short distances from a tall stack.

Although crops may be immersed in  $^{14}\text{CO}_2$  continuously, they take up  $^{14}\text{C}$  only when photosynthesis occurs. That is,  $^{14}\text{C}$  will be incorporated in plant tissues only during daylight hours of the growing season. Plants take up  $^{14}\text{C}$  at the leaf surface as  $^{14}\text{CO}_2$ , and  $\text{CO}_2$  gas passes through the stomata to be used as building blocks in the manufacture of chlorophyll and other molecules for growth and maintenance. Thus, most of the carbon atoms entering the leaves initially are translocated to other parts of the plant.

### 3.2 DOSIMETRY MODEL FOR $^{14}\text{C}$

#### 3.2.1 Individual Dose Commitment

##### 3.2.1.1 Ingestion Model

Ingestion is the dominant mode of exposure for  $^{14}\text{C}$  released to the environment, principally because almost 99% of reference man's carbon intake is via ingestion.<sup>29</sup> This quantity is listed in ICRP Publication 23 as approximately 300 g/day, and essentially all this intake is assimilated.<sup>30</sup> The usual assumption in modeling food chain transport of  $^{14}\text{C}$  is that the SA (specific activity) of food is equal to the SA in man. It follows that cattle, poultry, and other livestock feeding upon vegetation contaminated with  $^{14}\text{C}$  will likewise attain a steady-state SA at the same level, as will man as the final consumer.

The annual dose rate to a reference organ of an individual from uptake of  $^{14}\text{C}$  by ingestion is given by Killough et al. as<sup>31</sup>

$$\dot{D}_w = Q(\chi/Q)F_w(\text{DRF}), \quad (6)$$

where

$\dot{D}_w$  = annual dose rate (mrem/year),

$Q$  = rate of release of radioisotope from source (Ci/sec),

$\chi/Q$  = estimated dilution factor (sec/m<sup>3</sup>) for the receptor point of interest,

$F_w$  = fractional contribution of ingestion to the total uptake of carbon by the reference organ

$$= \frac{f_w \times (300 \text{ g C/day})}{f_w \times (300 \text{ g C/day}) + f_a \times (23 \text{ m}^3 \text{ air/day}) \times (0.16 \text{ g C/m}^3 \text{ air})},$$

$f_w$  = fraction of ingested carbon taken up by reference organ  
 $\cong 1$  for total body,

$f_a$  = fraction of inhaled carbon taken up by reference organ  
 $\cong 0.01$  for total body, and

(DRF) = dose rate factor for  $^{14}\text{C}$  in reference organ.

##### 3.2.1.2 Inhalation Model

Inhalation is a minor mode of exposure because reference man's daily intake is computed as

$$23 \text{ m}^3 \frac{\text{air}}{\text{day}} \times \frac{0.16 \text{ g C}}{\text{m}^3 \text{ air}} \cong 3.7 \frac{\text{g C}}{\text{day}}. \quad (7)$$

The total body uptake via inhalation is actually very small because only 1% is retained by the body; the balance is exhaled before uptake by tissues occurs.<sup>29</sup>

The model for computing the annual dose rate due to inhalation is identical to Eq. (6) except  $F_{\alpha}$  is used in place of  $F_w$ . This new quantity,  $F_{\alpha}$ , is defined as the fractional contribution of inhalation to the total uptake of carbon by the reference organ such that  $F_{\alpha} + F_w = 1$ .

If inhalation takes place at a location different from food production, then the appropriate  $\chi/Q$  must be substituted in Eq. (6) to reflect a different concentration of  $^{14}\text{C}$  breathed by man. If the concentration of  $^{14}\text{C}$  in air that man breathes is significantly greater (>100×) than the concentration of  $^{14}\text{C}$  in air where his food is produced, then the inhalation pathway may be the most important mode of exposure. This would be the only situation where the dose from inhalation could provide a significant part of the total dose.

### 3.2.1.3 Immersion Model

If a person is exposed by inhalation, then he will also be exposed at the same time by immersion in the plume. The dose received will depend upon the types of radiation and their energies. In the case of  $^{14}\text{C}$  the parameters are such that the dose from immersion can be neglected relative to the doses from ingestion and even inhalation. The EXREM III computer code was used to calculate a skin dose rate factor for immersion in an infinite cloud.<sup>32</sup>

### 3.2.2 Population Dose Commitment

Carbon-14 released to the atmosphere would mix rapidly with the  $1.8 \times 10^{18}$  g of carbon estimated to be present in the atmosphere, land biosphere, and the mixed layers of the ocean.<sup>31</sup> Mixing with the estimated  $3.0 \times 10^{19}$  g of carbon in the deep ocean occurs with a mean life of 59 years.<sup>33</sup> Because of this mixing throughout the atmosphere, biosphere, and ocean waters, and because  $^{14}\text{C}$  has a long radioactive half-life, the radiological impact to world populations from  $^{14}\text{C}$  will be present for many years after it is released to the environment.

Following UNSCEAR,<sup>34</sup> the global population dose commitment over all time following release of 1 Ci of  $^{14}\text{C}$  may be estimated from the rate of production of natural  $^{14}\text{C}$ ,  $3 \times 10^4$  Ci/year, and the average dose rate in human tissue from natural  $^{14}\text{C}$ ,  $1.3 \times 10^{-3}$  rem/year.

$$D = \gamma WN/B , \quad (8)$$

where

$D$  = global population dose commitment over all time following release of  $^{14}\text{C}$  (man-rem),

$\gamma$  = average dose rate in human tissue from natural  $^{14}\text{C}$  (rem/year),

$W$  = amount of  $^{14}\text{C}$  released (Ci),

$B$  = rate of production of natural  $^{14}\text{C}$  (Ci/year), and

$N$  = constant world population.

For a constant world population of  $6 \times 10^9$  and a release of 1 Ci this yields with substitution:

$$D = (1.3 \times 10^{-3} \text{ rem/year}) \frac{1 \text{ Ci} (6 \times 10^9)}{(3 \times 10^4 \text{ Ci/year})},$$

$$D = 260 \text{ man-rems}.$$

### 3.3 DOSES FROM $^{14}\text{C}$ IN THE VICINITY OF A REPROCESSING FACILITY HANDLING ADVANCED LMFBR FUELS

In this report radiological impact is estimated as the 50-year dose commitment (mrem). The "dose commitment" is associated with a specified intake of  $^{14}\text{C}$  and is defined as the total dose to a reference organ, resulting from one year of intake, that will accrue during the remaining lifetime of the individual. The exposed individual is assumed to be an adult (20 years of age) at the time of intake who will live to an age of 70 years. Thus, the dose commitment is calculated by integrating the dose rate over a 50-year period, and the result is called the "50-year dose commitment." In the case of  $^{14}\text{C}$ , which has a relatively short biological half-life of 28 days,<sup>29</sup> the dose commitment integrated to 50 years is not significantly different from the annual dose. Therefore, although it is technically correct to estimate the radiation exposure in units of the 50-year dose commitment, the annual exposure to an individual from  $^{14}\text{C}$  in mrem/year would be approximately the same value as the 50-year dose commitment. In this report, dose, dose commitment, and 50-year dose commitment have the same meaning when individual exposures are under discussion.

#### 3.3.1 Maximum Individual Doses

The AIRDOS computer code<sup>35</sup> was used to estimate 50-year dose commitments to the maximally exposed individual in the vicinity of a reprocessing facility handling advanced LMFBR fuels. The reprocessing plant was assumed to have a stack height of 100 m. Meteorology and other stack parameters were assumed to be identical to those described in the LMFBR Proposed Final Environmental Statement.<sup>1</sup> Release rates for  $^{14}\text{C}$  from the stack as  $^{14}\text{CO}_2$  are listed in Table 5 of this report.

The point of maximum exposure occurs at the plant boundary, which is 1000 m from the stack. Table 6 lists doses to various organs of an individual at this location. The dose estimates given are 50-year dose commitments calculated for one year of radionuclide release from the facility. Doses presented are for an adult individual residing constantly

Table 6. Estimated Maximum Individual 50-Year Dose Commitments<sup>a</sup> From <sup>14</sup>C Released From a Reprocessing Facility Handling LMFBR Fuels

Organ	Dose, mrem, for each Type of Fuel, <sup>14</sup> C Content (%), and Release Rate <sup>b</sup> in Ci/Year						
	Oxide	Carbide		Nitride			
	0.002, 2.5	0, $3.58 \times 10^{-3}$	0.1, $1.36 \times 10^2$	Natural, $7.82 \times 10^3$	10, 785	1, 78.5	0, $1.68 \times 10^{-3}$
Total body	$1.6 \times 10^{-2}$	$2.3 \times 10^{-5}$	0.87	50	5.0	0.50	$1.5 \times 10^{-5}$
Skeleton							
Endosteal cells	2.5	3.7	1.4	79	8.0	0.80	1.7
Red marrow	2.7	4.1	1.5	87	8.7	0.87	1.9
Lungs	0.70	1.0	0.38	22	2.2	0.22	0.47
Muscle	1.6	2.3	0.87	50	5.0	0.50	1.1
Adipose Tissue	4.7	6.7	2.5	145	14.5	1.45	3.2
Testes	0.62	0.90	0.34	19	1.9	0.19	0.43

<sup>a</sup>Calculated for an individual residing at the plant boundary, 1000 m from the stack.

<sup>b</sup>Carbon-14 release based on reprocessing fuel required to generate 50 GW-year of electric energy.

at the site boundary and consuming only food and beverages produced at that site. The primary exposure pathway is via ingestion which contributes greater than 99% of the dose.

Estimates of doses to an individual at the same location from radionuclides other than  $^{14}\text{C}$  released by a reprocessing plant handling mixed-oxide fuel were summarized in WASH-1535.<sup>1</sup> The dose estimate for the total body presented there is 2.0 mrem. Assuming the total body dose from radionuclides other than  $^{14}\text{C}$  associated with releases from an advanced fuel reprocessing plant is also 2.0 mrem, we can estimate the relative contribution of  $^{14}\text{C}$  to the total source term. The addition of  $^{14}\text{C}$  to the source term would significantly increase the total body dose from carbide fuel having 1000 ppm  $^{14}\text{N}$  and from nitride fuel having natural nitrogen, 10%  $^{14}\text{N}$  (enriched to 90%  $^{15}\text{N}$ ), or 1%  $^{14}\text{N}$  (enriched to 99%  $^{15}\text{N}$ ).

### 3.3.2 Average Individual and Population Doses

Population doses were calculated with the AIRDOS computer code<sup>35</sup> and assuming a population of one million living within a 50-mile radius of the facility. The meteorology and plant physical characteristics were identical to those reported in WASH-1535.<sup>1</sup>

Table 7 lists population doses in man-rems for each type of LMFBR fuel using total body as a reference organ. Dose estimates are 50-year dose commitments resulting from one year of radionuclide release. Estimates of population dose for other reference tissues would scale to the total body value as the individual dose estimates given for the respective tissues in Table 6. This calculation assumes that individuals living within an 80-km (50-mile) radius of the facility obtain all of their food and beverages from the point where they reside. Greater than 99% of the estimated dose is received via the ingestion pathway. Estimates for the average dose to all individuals living within the 50-mile radius are also listed in Table 7.

The total population [1 million in 80-km (50-mile) radius] dose from all radionuclides other than  $^{14}\text{C}$  released from a reprocessing facility handling mixed-oxide fuel was reported in WASH-1535<sup>1</sup> to be 35 man-rem per year of facility operation. Assuming that the population dose would be similar to this for a reprocessing plant handling carbide or nitride fuel instead of oxide fuel, the addition of  $^{14}\text{C}$  to the source term would significantly increase the population dose for carbide fuel having 1000 ppm  $^{14}\text{N}$  and for nitride fuel having natural nitrogen, 10%  $^{14}\text{N}$  (enriched to 90%  $^{15}\text{N}$ ), or 1%  $^{14}\text{N}$  (enriched to 99%  $^{15}\text{N}$ ).

### 3.3.3 The Effect of $^{14}\text{N}$ Content in Advanced Fuels on the Maximum Individual Dose

As discussed in Sect. 1 of this report, the principal mode of production of  $^{14}\text{C}$  in LMFBR fuels is by the  $^{14}\text{N}(n,p)^{14}\text{C}$  reaction, which has an effective cross section of 12.6 mb. Figure 3 illustrates the maximum individual 50-year dose commitment from  $^{14}\text{C}$  as a function of the  $^{14}\text{N}$

Table 7. Estimated 50-Year Total Body Dose Commitment to the Population and the Average Individual Within an 80-km (50-mile) Radius Due to  $^{14}\text{C}$  Released From a Reprocessing Facility Handling LMFBR Fuels

Type	Fuel $^{14}\text{N}$ Content (%)	Release Rate <sup>a</sup> (Ci/year)	Population Dose <sup>b</sup> (Man-rem)	Average Individual Dose (mrem)
Oxide	0.002	2.5	0.24	$2.4 \times 10^{-4}$
Carbide	0	$3.58 \times 10^{-3}$	$3.4 \times 10^{-4}$	$3.4 \times 10^{-7}$
	0.1	136	13	0.013
Nitride	99.64 (natural)	$7.82 \times 10^3$	760	0.76
	10	785	76	0.076
	1	78.5	7.6	$7.6 \times 10^{-3}$
	0	$1.68 \times 10^{-3}$	$1.6 \times 10^{-4}$	$1.6 \times 10^{-7}$

<sup>a</sup>Carbon-14 release based on reprocessing fuel required to generate 50 GW-year of electric energy.

<sup>b</sup>Population =  $1 \times 10^6$ .

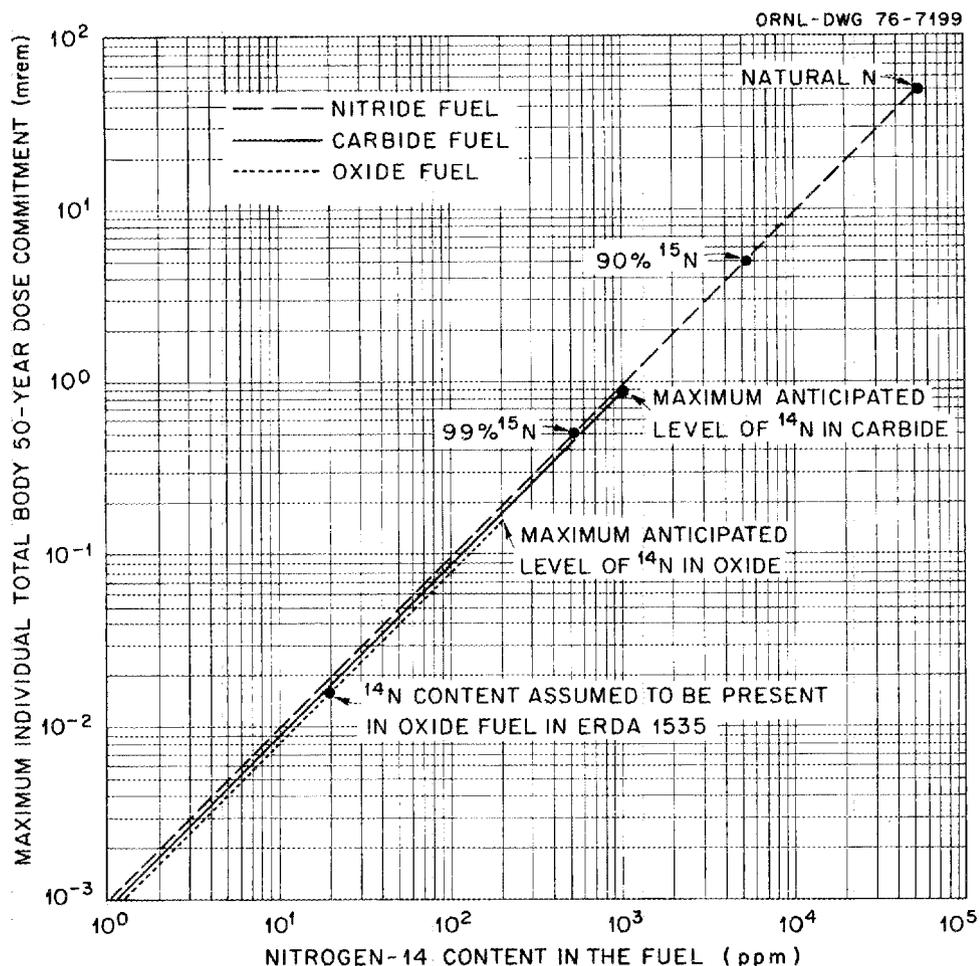


Fig. 3. Maximum Individual 50-Year Total Body Dose Commitment as a Function of  $^{14}\text{N}$  Content in Mixed-Oxide, Carbide, or Nitride LMFBR Fuel:

content in oxide, carbide, and nitride LMFBR fuels. This graph assumes a linear relationship between the  $^{14}\text{C}$  inventory and  $^{14}\text{N}$  content in the fuel. A containment factor for  $^{14}\text{C}$  of 100 in the fuel processing plant was used.

Several conclusions may be drawn from Fig. 3. First, in the range of 1 to 1000 ppm  $^{14}\text{N}$  there is little difference between the maximum individual dose commitment from  $^{14}\text{C}$  for each fuel at a given concentration of  $^{14}\text{N}$ . This result may be somewhat artificial, since the neutronic library used for the ORIGEN calculations was specific to an oxide-fuel LMFBR design. Libraries appropriate for advanced fuels and based on advanced fuel LMFBR designs are currently being developed and when available will be used to more accurately compute the  $^{14}\text{C}$  concentrations in spent advanced fuels. A second conclusion from this figure is that even at the maximum anticipated concentration of  $^{14}\text{N}$  of 200 ppm in oxide and 1000 in carbide fuels, the maximum individual dose commitment is less than

1 mrem. Finally, Fig. 3 illustrates that the dose commitment from nitride fuel may be significantly greater than the dose commitment from either oxide or carbide fuel depending on the fraction of the nitrogen that is  $^{15}\text{N}$ .

### 3.4 ANALYSIS OF WORLD POPULATION DOSES FROM $^{14}\text{C}$ PRODUCED IN ADVANCED LMFBR FUELS

Dose commitments to the world population from  $^{14}\text{C}$  produced by LMFBR fuels were estimated by assuming a value of 260 man-rems/Ci of release to the environment calculated in Sect. 3.2.2. These dose commitments are listed in Table 8 and are based on 50 GW(e)-year of energy for each fuel. Assuming the annual dose from  $^{14}\text{C}$  derived from natural sources is 1.3 mrem and a world population of  $6 \times 10^9$ , the annual dose commitment to the world population from natural  $^{14}\text{C}$  would be  $7.8 \times 10^6$  man-rems. This value cannot be directly compared with  $^{14}\text{C}$  from advanced LMFBR fuels because of the uncertainty in predicting the contribution which the LMFBR will make to energy production in the future. It is possible that if nitride fuel with natural nitrogen is selected, when the energy produced by LMFBRs is significantly greater than 50 GW(e)-year, then the dose commitment to the world population from  $^{14}\text{C}$  produced in LMFBRs could exceed that from  $^{14}\text{C}$  that occurs naturally.

Table 8. Dose Commitment to the World Population From  $^{14}\text{C}$  Produced by LMFBRs

Type of Fuel	Release Rate of $^{14}\text{C}$ per 50 GWe-years of Energy from LMFBRs <sup>a</sup> (Ci/year)	Dose Commitment to the World Population <sup>b</sup> (man-rems)
Oxide	2.5	650
Carbide		
No $^{14}\text{N}$	$3.58 \times 10^{-3}$	0.93
1000 ppm $^{14}\text{N}$	136	$3.5 \times 10^4$
Nitride		
Natural N	$7.82 \times 10^3$	$2.0 \times 10^6$
90% $^{15}\text{N}$	785	$2.0 \times 10^5$
99% $^{15}\text{N}$	78.5	$2.0 \times 10^4$
100% $^{15}\text{N}$	$1.68 \times 10^{-3}$	0.44

<sup>a</sup>Assuming a decontamination factor of 100 at the reprocessing plant.

<sup>b</sup>Assuming 260 man-rems/Ci of  $^{14}\text{C}$  released to the environment.

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