# Carbon-14 Production in Nuclear Reactors

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## ABSTRACT

Quantities of $^{14}C$ that may be formed in the fuel and core structural materials of light-water-cooled reactors (LWRs), in high-temperature gas-cooled reactors (HTGRs), and in liquid-metal-cooled fast breeder reactors (LMFBRs) have been calculated by use of the ORIGEN code.[1] Information supplied by five LWR-fuel manufacturers pertaining to nitride nitrogen and gaseous nitrogen in their fuels and fuel-rod void spaces was used in these calculations. Average nitride nitrogen values range from 3 to 50 ppm (by weight) in LWR fuels, whereas gaseous nitrogen in one case is equivalent to an additional 10 to 16 ppm. Nitride nitrogen concentrations in fast-flux test facility (FFTF) fuels are 10 to 20 ppm. The principal reactions that produce $^{14}C$ involve $^{14}N$, $^{17}O$, and (in the HTGR) $^{13}C$. Reference reactor burnups are 27,500 MWd per metric ton of uranium (MTU) for boiling water reactors (BWRs), 33,000 MWd for pressurized water reactors (PWRs), about 95,000 MWd per metric ton of heavy metal (MTHM) for HTGRs, and 24,800 MWd/MTHM for an LMFBR with nuclear parameters that pertain to the Clinch River Breeder Reactor. Nitride nitrogen, at a median concentration of 25 ppm, contributes 14, 15, and 6 Ci of $^{14}C/GW(e)$-yr to BWR, PWR, and LMFBR fuels, respectively. The contribution of $^{17}O$ in BWR and PWR fuels is 3.3 and 3.5 Ci of $^{14}C/GW(e)$-yr, respectively, but it is less than 0.2 Ci/GW(e)-yr, in blended LMFBR fuel. In the HTGR fuel particles ($UC_2$ or $ThO_2$), 10 Ci of $^{14}C/GW(e)$-yr will be formed from 25 ppm of nitrogen, whereas $^{17}O$ in the $ThO_2$ will contribute an additional 2 Ci/GW(e)-yr. All $^{14}C$ contained in the fuels may be released in a gas mixture ($CO_2$, CO, $CH_4$, etc.) during fuel dissolution at the fuel reprocessing plants. However, some small fraction may remain in aqueous raffinates and will not be released until these are converted to solids. The gases would be released from the plant unless special equipment is installed to retain the $^{14}C$-bearing gases.

Cladding metals and other core hardware will contain significant quantities of $^{14}C$. Very little of this will be released from BWR, PWR, and LMFBR hardware at fuel reprocessing plants; instead, the contained $^{14}C$, 30 to 60 Ci/GW(e)-yr for LWRs and about 13 Ci/GW(e)-yr for a CRBR, will remain within the metal, which will be retained on site or in a Federal repository. The only core structural material of HTGRs will be graphite, which will contain 37 to 190 Ci of $^{14}C/GW(e)$-yr, exclusive of that in the fuel particles, if the graphite (fuel block and reflector block) initially contains 0 to 30 ppm of nitrogen. All of this is available for release at a fuel reprocessing plant if the graphite is burned to release the fuel particles for further processing. Special equipment could be installed to retain the $^{14}C$-bearing gases.

## 1.0 INTRODUCTION

The radioactive nuclide $^{14}C$ is, and will be, formed in all nuclear reactors due to absorption of neutrons by carbon, nitrogen, or oxygen. These may be present as components of the fuel, moderator, or structural hardware, or they may be present as impurities. Most of the $^{14}C$ formed in the fuels or in the graphite of HTGRs will be converted to a gaseous form at the fuel reprocessing plant, primarily as carbon dioxide; this will be released to the environment unless special equipment is installed to collect it and convert it to a solid for essentially permanent storage. If the $^{14}C$ is released as carbon dioxide or in any other chemical form, it will enter the biosphere, be inhaled or ingested as food by nearly all living organisms including man, and will thus contribute to the radiation burden of these organisms. Carbon-14 is formed naturally by reaction of neutrons of cosmic ray origin in the upper atmosphere with nitrogen and, to a lesser extent, with oxygen and carbon. Large amounts of $^{14}C$ have also been formed in the atmosphere as a result of nuclear weapons explosions.

For the last two decades, the quantities of $^{14}C$ in the environment, and the mechanisms of transfer of this nuclide between the atmosphere, land biota, and the shallow and deep seas have been the subject of many research studies.[2-9] These studies have shown that most of the $^{14}C$ is actually contained in the deep oceans, at depths greater than 100 m. The nuclear weapons tests increased the total $^{14}C$ inventory of the earth by only a few percent,[5] but the atmospheric content was approximately doubled. Since atmospheric weapons tests are no longer being conducted, the atmospheric concentration of $^{14}C$ is now decreasing as it enters the oceans as $CO_2$ and is approaching the pretest value.

Some estimates of the amounts of $^{14}C$ released from or formed in LWRs,[10-15] HTGR,[13,14] and LMFBR[15] have been made previously on the basis of calculations or measurements. The purpose of this report is to present detailed estimates of the production of $^{14}C$ with emphasis on those pathways that are likely to lead to the release of this nuclide, either at the reactor site or at the fuel reprocessing plant.

## 2.0 MECHANISMS OF CARBON-14 FORMATION IN NUCLEAR REACTORS

Carbon-14 is formed from five reactions of neutrons with isotopes of elements that are normal or impurity components of fuel, structural materials, and the cooling water of LWRs. The neutron-induced reactions are as follows:

(1)  $^{13}C(n,\gamma)^{14}C$;

(2)  $^{14}N(n,p)^{14}C$;

(3)  $^{15}N(n,d)^{14}C$;

(4)  $^{16}O(n,^3He)^{14}C$;

(5)  $^{17}O(n,\alpha)$ $^{14}C$.

In these reactions, standard notation has been used in which n refers to a neutron, p to a proton, d to a deuteron ($^2$H), and $\gamma$ to a gamma ray. Reactions 4 and 5 will occur in any reactor containing heavy-metal oxide fuels and/or water as the coolant. Reaction 1 will be important only in the HTGRs, while reactions 2 and 3 will occur in all reactors containing nitrogen as an impurity in the fuel, coolant, or structural materials.

To facilitate calculations, the energy-dependent cross sections of nuclear reactions are typically collapsed into a single, effective cross section that applies to the neutron spectrum of the reactor in question. Such collapsed values are known with fairly good accuracies for reactions 1, 2, and 5 for the thermal-neutron spectra of LWRs and HTGRs. Values listed in Table 1 for the BWR, PWR, and HTGR are taken from the ORIGEN library[1] and its update[16] according to the latest version of the "Barn Book."[17] Because reactions 3 and 4 are highly endothermic, their cross sections are assumed to be 0.0 in thermal reactors, as shown in Table 1. Unfortunately, some of these cross sections for the LMFBR are very uncertain. The following discussion concerning cross sections of reactions 1-5, as they apply to the Clinch River Breeder Reactor (CRBR), has been provided by A. G. Croff.[18]

### Reaction 1 $^{13}$C(n,$\gamma$)$^{14}$C

The cross section for this reaction is not well known for nonthermal neutron energies. The assumed values were taken from ref. 19, in which the $^{13}$C(n,$\gamma$) cross section was calculated on the bases of a few experimental data and nuclear systematics. The cross section obtained when the data are collapsed to a single value using the CRBR neutron spectrum is 0.5 $\mu$b (1 $\mu$b = $10^{-6}$ barns). The fact that the thermal $^{13}$C(n,$\gamma$) cross section is only about 1 mb (Table 1) coupled with the fact that cross sections in the nonthermal energy regions are considerably smaller than thermal cross sections tends to confirm that the 0.5 $\mu$b value is realistic.

### Reaction 2 $^{14}$N(n,p)$^{14}$C

Of the five $^{14}$C-producing reactions listed, this is the only one for which the experimental data may be considered adequate. Energy dependent cross-section data for the $^{14}$N(n,p)$^{14}$C reaction are available from the ENDF/B[20] compilation. Collapsing these data with the CRBR spectrum gives a cross section of 12.6 mb, with an estimated error of ±30%.

### Reaction 3 $^{15}$N(n,d)$^{14}$C

The only cross-section data available for this reaction are some sketchy information on the angular distribution of the deuterons when the neutrons have energies of 14 to 15 MeV. This information, coupled with the fact that the reaction is endothermic (Q = -7.99 MeV), would probably lead to a value of the reaction rate in the 0.01 to 0.1 mb range. However, for calculational purposes, a value of 1.0 mb was used.

### Reaction 4 $^{16}$O(n,$^3$He)$^{14}$C

Of the five reactions considered, the data for this reaction are by far the least well-known. It is highly endothermic (Q = -14.6 MeV), indicating that greater neutron energies are required for the

Table 1. Cross sections for formation and yields of $^{14}$C in BWR, PWR, HTGR, and LMFBR[a]

| Reaction No. | Reaction | Cross section for formation of $^{14}$C in | | | | $^{14}$C formation (curies per gram of parent element) | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | BWR | PWR | HTGR | LMFBR | BWR | PWR | HTGR | LMFBR |
| 1 | $^{13}$C(n,γ)$^{14}$C | 1.00 mb | 1.00 mb | 0.419 mb | 0.5 ub | 1.51E-7 | 1.61E-7 | 3.38E-7 (3.69E+0)[b] | 4.81E-9 |
| 2 | $^{14}$N(n,p)$^{14}$C | 1.48 b | 1.48 b | 1.02 | 12.6 mb | 1.71E-2 | 1.83E-2 | 3.84E-2 | 9.66E-3 |
| 3 | $^{15}$N(n,d)$^{14}$C | 0 | 0 | 0 | 1.0 mb | 0 | 0 | 0 | 2.85E-6 |
| 4 | $^{16}$O(n,$^{3}$He)$^{14}$C | 0 | 0 | 0 | 0.05 μb | 0 | 0 | 0 | 3.82E-8 (4.53E-3)[c] |
| 5 | $^{17}$O(n,$^{4}$He)$^{14}$C | 0.183 b | 0.183 b | 0.110 b | 0.12 mb | 7.31E-7 (1.01E-1)[c] | 7.75E-7 (0.87E-2)[c] | 1.79E-6 (2.25E-1)[d] | 3.40E-8 (4.03E-3)[c] |

[a] All of the values in this table were obtained by collapsing available neutron cross-section data to a single value, using neutron spectra of the individual reactors, as discussed by Bell.[1] These values are not equal to 2200-m/sec cross sections, such as 0.9 mb, 1.81 b, and 0.235 b for reactions 1, 2, and 5, respectively.

[b] Based on 10.93 MT of carbon/MTHM where HM = thorium plus uranium.

[c] Based on 8383 g-at. of oxygen/MTHM where HM = uranium or uranium plus plutonium, present as $UO_2$ and $PuO_2$.

[d] Based on 0.9094 MT of thorium/MTHM with thorium present as $ThO_2$ and uranium as UC.

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reaction to proceed. Information supplied by the Physics Division of Lawrence Livermore Laboratory indicates that the cross section at 15 MeV should be less than 1 mb, and at 20 MeV it should be less than 10 mb. By combining these "guesstimates" with the CRBR spectrum and a theoretical expression for the availability of high-energy fission neutrons, the reaction cross section is estimated to be about 0.05 $\mu$b. The lack of information on both the high-energy cross sections and the high-energy neutron spectrum makes this value very uncertain.

*Reaction 5 $^{17}O(n,\alpha)^{14}C$*

As with reaction 1, the cross-section data for this reaction are not well known. The data, which again are based on only a few experiments and nuclear systematics, were taken from ref. 19. The cross section, which is calculated and based on the CRBR spectrum, is 0.12 mb.

The assumed LMFBR fuel model was the Atomics International Follow-On Design. Initial concentrations of the isotopes of importance in this case (in g-atoms/MTHM) are:

| | |
|---|---|
| $^{12}C$ | 33.33 |
| $^{13}C$ | 0.374 |
| $^{14}N$ | 1.42 |
| $^{15}N$ | 0.00528 |
| $^{16}O$ | 8383. |
| $^{17}O$ | 3.27 |
| $^{18}O$ | 17.2 |

The ORIGEN code[1] is not capable of explicitly accounting for (n,d) or (n,$^3$He) reactions. This difficulty may be circumvented by combining reaction 4 with reaction 5 and reaction 3 with reaction 2, since the naturally occurring isotopes are present in a fixed ratio for each element. Alternatively, since the depletion of the carbon, nitrogen, and oxygen is relatively small (<2%), the calculation is easily performed by hand.

# 3.0 CARBON-14 FORMATION IN LIGHT-WATER REACTORS

Carbon-14 is formed in the fuel ($UO_2$), in core structural materials, and in the cooling water of LWRs.

## 3.1 Formation in the Fuel

Carbon-14 will be formed primarily by two reactions in the fuel: $^{17}O(n,\alpha)^{14}C$ and $^{14}N(n,p)^{14}C$. The quantity of $^{14}C$ formed from the first of these reactions can be calculated accurately on the basis of the stoichiometry of $UO_2$ (134.5 kg O/MTU) and an abundance of 0.039 at. % $^{17}O$ in normal oxygen, which corresponds with 55.6 g of $^{17}O$/MTU or 3.27 g-atoms of $^{17}O$/MTU. As listed in Table 2, burnup of BWR and PWR fuels to 27,500 and 33,000 MW(t)d/MTU, respectively, leads to the formation of 0.098 and 0.104 Ci of $^{14}C$/MTU, which corresponds with 3.3 and 3.5 Ci/GW(e)-yr, respectively.

Table 2. Production of $^{14}C$ in core hardware and fuel at light-water reactors (BWR and PWR)

| Material | Quantity in core (kg/MTU) | Quantity of element in core (g/MTU) | | | $^{14}C$ existing 160 days after discharge of fuel (Ci/MTU) | | | Total $^{14}C$ production | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | Carbon | Nitrogen | Oxygen | From carbon | From nitrogen | From oxygen | Calculated Ci/MTU | Ci/GW(e)-yr[a] | Observed Ci/GW(e)-yr |
| **Boiling-Water Reactor[b]** | | | | | | | | | | |
| Zircaloy-2 (Grade RA-1) | 316 | ≤85.3 | ≤25.3 | | 1.29E-5 | 4.33E-1 | | 0.433 | 14.5 | |
| 304 stainless steel | 50 | ≤40.0 | 50-80 | | 0.60E-5 | (0.86-1.37)E+0 | | 0.86-1.37 | 28.7-45.9 | |
| Inconel-X | 3.4 | ≤3.4 | | | 0.05E-5 | | | 0.000 | 0.0 | |
| Uranium dioxide | 1135 | | Low  10 | 134,500 | | 1.71E-1 | 9.83E-2 | 0.269 | 9.0 | |
| | | | Med  25 | | | 4.28E-1 | | 0.526 | 17.6 | |
| | | | High  75 | | | 1.28E+0 | | 1.38 | 46.3 | |
| Water | 216 | | | 192,000 | | | 1.40E-1 | 0.140 | 4.7 | 8[c] |
| Totals, Low | | | | | | | | 1.70 | 57 | |
| Med | | | | | | | | 2.21 | 74 | |
| High | | | | | | | | 3.32 | 111 | |
| **Pressurized-Water Reactor[d]** | | | | | | | | | | |
| Zircaloy-4 (Grade RA-2) | 235 | ≤63.5 | ≤18.8 | | 1.02E-5 | 2.74E-1 | | 0.274 | 9.5 | |
| 302 stainless steel | 4.2 | ≤3.4 | 4.2-6.7 | | 0.05E-5 | (0.61-0.98)E-1 | | 0.061-0.098 | 2.1-3.4 | |
| 304 stainless steel | 37.1 | ≤29.7 | 37.1-59.4 | | 0.48E-5 | (5.42-8.67)E-1 | | 0.542-0.867 | 18.8-30.0 | |
| Inconel 718 | 12.8 | ≤1.3 | | | 0.02E-5 | | | 0.000 | 0.0 | |
| Microbraze 50 | 2.6 | 0.3 | 0.2 | 1.1 | 0.00E-5 | 3.66E-3 | 0.85E-6 | 0.004 | 0.12 | |
| Uranium dioxide | 1135 | | Low  10 | 134,500 | | 1.83E-1 | 1.04E-1 | 0.287 | 9.6 | |
| | | | Med  25 | | | 4.57E-1 | | 0.561 | 18.8 | |
| | | | High  75 | | | 1.37E+0 | | 1.48 | 49.5 | |
| Water | 216 | | | 192,000 | | | 1.49E-1 | 0.149 | 5.0 | 6 |
| Totals, Low | | | | | | | | 1.32 | 44 | |
| Med | | | | | | | | 1.77 | 59 | |
| High | | | | | | | | 2.87 | 96 | |

[a] Based on 33.5 MTU/GW(e)-yr.

[b] ORIGEN calculations assume 18.823 MW(t)/MTU, 4 years in reactor, to 27,500 MWd/MTU; 2.6 wt % $^{235}U$. Quantities of metal in core from ref. 21.

[c] The measured value[12] at the Nine Mile Point reactor [625 MW(e)] was 8 Ci/yr; see text for comments on power density and steam/liquid water volume.

[d] ORIGEN calculations assume 30.0 MW(t)/MTU, 3 years in reactor, to 33,000 MWd/MTU; 3.3 wt % $^{235}U$. Quantities of metal in core from ref. 22.

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There is considerable variation in production of $^{14}C$ from the $^{14}N(n,p)$ reaction because of variations in the nitrogen content of LWR fuels. Crow[23] presented the following brief summary of a survey of five fuel fabrication plants:

| | |
|---|---|
| Maximum nitrogen allowed by specification, ppm | 75-100 |
| Maximum nitrogen reported, ppm | 100 |
| Minimum nitrogen reported, ppm | 1 |
| Average nitrogen in reactor fuel, ppm | 25 ±5 |

He has indicated that the 25 ±5 ppm average is not a true arithmetic average but a consensus derived from discussions with representatives of fuel manufacturers.

Table 3 contains the results of a much more extensive survey of the nitrogen content of fuels made at these same five plants. The current average nitrogen content varies from 3 to 50 ppm and the standard deviation of each average is in the range of 40 to 70% of the average. The data shown in Table 3 suggest that the median value of fuel from all plants is about 25 ppm.

The differences in the nitride-nitrogen concentrations in LWR fuels from the five manufacturers listed in Table 3 are due to many variables. Some of these have been described qualitatively and are discussed by Pechin et al.[24] without reference to reaction times, temperatures, and concentrations. Uranium hexafluoride from gaseous diffusion plants, enriched to 2 to 4 wt % in $^{235}U$, is the starting material in the manufacture of LWR fuels. Four of the manufacturers use the ammonium diuranate (ADU) process, and one uses the direct (dry) conversion (DC) process. Powdered $UO_2$ is obtained from both processes, cracked $NH_3$ being the preferred source of hydrogen reductant. Pellets are obtained by pressing the powder into pellet form and sintering these in hydrogen, as in the uranium-valence reduction step. Pellet pressing is performed as a dry operation (except for a little lubricant). Sintering is performed at temperatures ranging from $\leq 1600°C$ to $\geq 1750°C$. After cooling, the pellets are loaded into Zircaloy fuel tubes (closed at one end), usually without any additional treatment. Before the fuel tube is welded closed in a helium atmosphere at all plants, air is removed in a vacuum degassing step at four plants, but is left in place at one of the plants. During the degassing operation, pellets in the fuel rods are unheated in some plants and heated in others. All vaccum degassing operations are followed by filling the fuel rod with high-purity helium and closing the second end by welding in a helium atmosphere. Helium is added under pressure to fuel tubes at the plant at which the the vacuum degassing step is not employed. The gaseous nitrogen from 18 to 30 cc of air in a single fuel tube containing about 1.75 kg of $UO_2$ corresponds to an additional 10 to 16 ppm of $N_2$ that is not included in Table 3.

Because of the wide range of nitrogen concentrations, three values of $^{14}C$ production from the $^{14}N(n,p)$ reaction are listed in Table 2. These correspond to 10, 25, and 75 ppm of nitrogen. At these three levels, $^{14}C$ production for the listed burnup conditions are 0.171, 0.428, and 1.28 Ci/MTU, respectively, which corresponds to 5.7, 14.3, and 42.9 Ci/GW(e)-yr for the BWR. Similar values for the PWR are 0.183, 0.457, and 1.37 Ci/MTU, respectively, and 6.1, 15.3, and 45.9 Ci/GW(e)-yr.

It may be noted that the same quantity of $^{14}C$ will be produced from $^{17}O(n,\alpha)$ and $^{14}N(n,p)$ reactions when the nitrogen content of the fuel is about 5.7 ppm for both PWRs and BWRs.

The chemical form of $^{14}C$ in the fuel is not known. When formed from any of the five nuclear reactions presented in Sect. 2, this nuclide might become bound to uranium as carbide, remain as impurity atoms, or be converted to carbon monoxide or carbon dioxide. A nitrogen impurity of 75 ppm corresponds to 1.28 Ci of $^{14}C/MTU$ in the case of the reference BWR and to 1.37 Ci of $^{14}C/MTU$ in the case of the reference PWR (Table 2). These maximum expected activities

Table 3. Nitrogen content of $UO_2$ fuels for LWRs and of FFTF fuels[a]

| | Current production of LWR fuels ($UO_2$) | | | | | FFTF fuels[b] [(U,Pu)$O_2$] | | | |
| | Company | | | | | Company A fuel | | Company B fuel | |
| | | | | | | Analyzed by | | Analyzed by | |
| | 1 | 2 | 3 | 4 | 5 | Company A | HEDL | Company B | HEDL |
|---|---|---|---|---|---|---|---|---|---|
| No. of measurements | 358 | 408 | 38 | 206 | 70 | 80 | 10 | 80 | 10 |
| **Percent of measurements with nitrogen, ppm** | | | | | | | | | |
| <10 | 100 | 75 | 42 | 14 | 10 | 68 | 100 | 78 | 90 |
| 10 – 20 | | 12 | 53 | 39 | 1 | 4 | | 17 | |
| 20 – 35 | | 9 | | 36 | 16 | 12 | | 5 | 10 |
| >35 | | 4 | 5 | | | | | | |
| 35 – 50 | | | | 10 | 27 | 2 | | | |
| >50 | | | | 1 | 46 | 14 | | | |
| Mass-weighted av nitrogen, ppm | 2.8 | 13.3 | 13.7 | 21.6 | 47.8 | <21.6[c] | <10[c] | <11.1[c] | <9.2[c] |
| Std deviation, ppm[d] | 1.4 | 8.3 | 9.8 | 11.1 | 21.2 | N.A | N.A | N.A | N.A. |

[a] Primarily nitride nitrogen.

[b] From ref. 52.

[c] Numerical values are based on using the many values <10 ppm as 10.0 ppm.

[d] It is emphasized that the distribution of nitrogen analyses is not normal. N.A. (not available) is used because a meaningful standard deviation cannot be calculated.

correspond to a ratio of about 1 $^{14}$C atom/200,000 uranium atoms. Ferris and Bradley[25] studied the reactions of uranium carbides with nitric acid and found that 50 to 80% of the carbide carbon was converted to carbon dioxide; the remaining carbide carbon was converted to nitric acid-soluble chemicals such as oxalic acid, mellitic acid, and other species, probably aromatics highly substituted with -COOH and -OH groups. Formation of such compounds can be reconciled with the existence of the polymeric -C-C- bonds of uranium carbides. However, at a ratio of 1 $^{14}$C atom/200,000 uranium atoms, or even at a ratio 1 C atom/500 uranium atoms, which would correspond to an impurity of 100 ppm of carbon in the $UO_2$, there will be a very low concentration of -C-C- bonds in the $UO_2$ fuels. This suggests that a larger quantity of any carbide carbon, including that formed from nuclear reactions, will be converted to $CO_2$ in dissolving operations at the fuel reprocessing plant than the 50 to 80% reported by Ferris and Bradley[25] for pure uranium carbides. An experimental program to measure $^{14}$C liberated during fuel dissolution is now in progress.[26]

## 3.2 Formation in Core Hardware

Core structural materials include stainless steel support hardware, Zircaloy cladding, and nickel alloys used as springs and fuel tube separators. According to specifications,[27-31] the primary source of $^{14}$C in these materials is the nitrogen that is present in quantities listed in Table 4. The quantities of each of the types of metal (i.e., stainless steel, Zircaloy, Inconel-X) are somewhat dependent on the reactor type (BWR[32-34] or PWR[34-37]) and on the year and size of the design within a reactor type. For example, Fuller et al.[32] have presented data on the fifth and sixth generation BWRs (BWR/5 and BWR/6) from which the weight ratios are calculated to be 247 and 265 kg of Zircaloy-2/MTU, respectively. Other estimates of quantities of structural hardware have been given by Griggs[38] and by Levitz et al.[39] However, the quantities of these metals, the contained nitrogen, and the $^{14}$C produced (as listed in Table 2) are based on information pertaining to present reactor designs provided by Marlowe[21] and Kilp.[22] Carbon-14 values are based on calculations with the ORIGEN code[1] for a BWR operated to a burnup of 27,500 MW(t)d/MTU in 4 yr and a PWR to a burnup of 33,000 MW(t)d/MTU in 3 yr. The revised light-element library[16] was used in these calculations. Most of the $^{14}$C formed in these structural components will be retained within the metal when the latter is encapsulated for long-term disposal, although a very small fraction in the Zircaloy might be dissolved in fuel leaching solutions at the fuel reprocessing plant. Experiments have never been performed to evaluate this possibility.

## 3.3 Formation in Cooling Water

Oxygen of the cooling water and nitrogen-containing chemicals in this water are sources of $^{14}$C. An accurate calculation of the quantity of $^{14}$C that will be formed would require integrating the flux over the volume of water in and surrounding the core. Data to perform such an integration do not appear to be readily available, but reasonable approximations can be made. Reference 34 gives values for the atomic ratio H/U of 3.74 and 4.23 for BWRs and PWRs, respectively; these correspond to 7860 and 8890 g-atoms of O (as $H_2O$)/MTU. Fuller et al.[32] give values of the water/fuel volume ratio of 2.52 for BWR 5 and 2.50 for BWR/6. A water density of 0.805 g/cm$^3$ and a $UO_2$ density of 10 g/cm$^3$, both at 550°F, indicate a ratio of about 13,000 g-atoms of O/MTU for the BWR cores. Reference 36 gives a hot, first core $H_2O$/$UO_2$ volume ratio (for a PWR) of 2.08,

Table 4. Specifications for carbon and nitrogen in reactor structural and cladding metals

| | Reactor type | Specifications (wt %) | | References for specifications |
| --- | --- | --- | --- | --- |
| | | Carbon | Nitrogen | |
| Stainless steel 304 | BWR | ≤0.08 | 0.10-0.16 | ASME SA213-73[27] and ASME SA-240[28] |
| 304 | PWR | ≤0.08 | 0.10-0.16 | ASME SA213-73[27] and ASME SA-240[28] |
| 316 | LMFBR | 0.040-0.060 | ≤0.010 | RDT M3-28T[29] |
| Zircaloy-2 | BWR | ≤0.027 | ≤0.008 | ASTM B353-71 (ANSI N124-1973)[30] |
| Zircaloy-4 | PWR | ≤0.027 | ≤0.008 | ASTM B353-71 (ANSI N124-1973)[30] |
| Inconel-X | BWR | ≤0.10 | | International Nickel Co.[31] |
| Inconel 718 | PWR | ≤0.10 | | International Nickel Co.[31] |
| Nicrobraze 50 | PWR | 0.01 | 0.0066 | |

which corresponds to about 10,500 g-atoms of O/MTU. For the purpose of this report, it is thus assumed that the rate of reaction $^{17}O(n,\alpha)^{14}C$ is specified by a ratio 12,000 g-atoms of O/MTU and a natural $^{17}O$ abundance of 0.039 at. % in oxygen for both BWRs and PWRs. This corresponds (Table 2) to about 4.7 and 5.0 Ci of $^{14}C/GW(e)$-yr for BWRs and PWRs, respectively, from the $^{17}O(n,\alpha)^{14}C$ reaction; it also corresponds to an initial atomic ratio $H/^{235}U$ of about 220 for BWRs and 175 for PWRs using fuels containing 2.6% and 3.3% $^{235}U$, respectively.

The quantity of $^{14}C$ formed from impurity nitrogen cannot be estimated since there do not appear to be any analyses pertaining to the concentration of this element in reactor cooling water. Although its concentration may be no more than a few parts per million, Cohen[40] mentions a value as high as 50 ppm $NH_3$ in the primary cooling water of PWRs.

Quantities of $^{14}C$ actually released from a BWR and three PWRs, as measured by Kunz and his coworkers,[11,12] are listed in Table 2. From the BWR at Nine Mile Point [625 MW(e)] they observed[12] a release rate of 8 Ci of $^{14}C/yr$. These authors also reported 6 Ci of $^{14}C/GW(e)$-yr on the basis of their analyses of gaseous effluents from the Ginna, Indian Point 1, and Indian Point 2 PWRs. At the PWR stations,[11] over 80% of the $^{14}C$ activity was chemically bound as $CH_4$ and $C_2H_6$; only small quantities were bound as $CO_2$. At the Nine Mile Point BWR station[12] the chemical form of $^{14}C$ was greatly different, with 95% as $CO_2$, 2.5% as CO, and 2.5% as hydrocarbons.

On the bases of the fuel isotopic compositions and burnups shown in the footnotes of Table 2 and for the assumed ratio of 12,000 g-atoms of O/MTU, an impurity of 1 ppm of nitrogen in the cooling water (corresponding to 0.216 g of N/MTU) would lead to the formation of 0.124 and 0.132 Ci of $^{14}C/GW(e)$-yr in BWRs and PWRs, respectively. The difference between a calculated 5 Ci of $^{14}C/GW(e)$-yr from the $^{17}O(n,\alpha)$ reaction and the observed 6 Ci/yr at the PWR stations[11] (Table 2) is probably well within limits of analytical uncertainty. The extrapolation to 16 Ci of $^{14}C/GW(e)$-yr from the measured 8 Ci/yr at the Nine Mile Point BWR is based on maintenance of a constant power density and a constant volume ratio $H_2O/UO_2$. Values of this ratio tabulated for the Nine Mile Point reactor[41] and for newer, larger reactors, such as those at Brown's Ferry,[42] do not differ significantly (2.38 vs 2.43); the average power densities for the two reactors are 41 and 50.732 kW/liter, respectively. When these ratios are combined with data on the average void fractions within a fuel assembly (a measure of steam/liquid water, and having values of 0.3 for the Nine Mile Point core and 0.4 for the Brown's Ferry core), it is apparent that $^{14}C$ formation in a new 1100 MW(e) BWR (such as BWR/5[32]) would be larger than 8 Ci/GW(e)-yr, but significantly less than 16 Ci/GW(e)-yr.

## 4.0 CARBON-14 FORMATION IN HIGH-TEMPERATURE GAS-COOLED REACTORS

The only structural materials in HTGRs in which $^{14}C$ will be formed to any significant extent are the fuel containing and reflector blocks of graphite. There will be some nitrogen and oxygen in the helium coolant.[43] However, the rate of $^{14}C$ formation from coolant impurities will be very small in comparison with similar rates in the fuel blocks; in addition, the helium cleanup system is expected to remove $CO_2$, a probable form of part of the $^{14}C$ in the coolant.

### 4.1 Formation in the Fuel

The compositions of fertile and fissile fuel for HTGRs have not been positively established since commercial reactors are not yet being made. However, it is highly probable[44] that the initial and

makeup (the IM stream) fuel will be in the form of about 93 wt % of $^{235}$U as $UC_2$, that $^{233}$U bred from the fertile thorium will be recycled as $UC_2$ (the 23R stream), and that uranium recovered from the IM stream after reprocessing, if it is recycled as the 25R stream, will also be in the form of $UC_2$. Similarly, the fertile thorium is expected to be in the form of $ThO_2$. Uranium in the IM stream will have a chemical history different than that of uranium in the 23R and 25R streams. In particular, uranium for the IM stream will be received at a fresh-fuel fabrication plant[45] as $UF_6$, which will be hydrolyzed with steam to $UO_2F_2$; this, in turn, will be reduced at about $650^0$C with $H_2$ ( from cracked ammonia) to $UO_2$. Subsequently, the $UO_2$ will be mixed with carbon flour, ethyl cellulose and methylene chloride. It will then be dried, ground, separated into appropriate sizes, and heated in a vacuum to cause the formation of $UC_2$. Finally, it will be cooled in an inert atmosphere, which may either be nitrogen or argon. In these successive processes, the uranium-bearing material never exists as a nitrogen-containing compound, although it is exposed to $N_2$ from cracked ammonia at a high temperature and may be exposed to nitrogen after formation of $UC_2$.

On the other hand,[14] recycle uranium, both 23R and 25R streams, will pass through the uranyl nitrate [$UO_2(NO_3)_2$] state in a fuel reprocessing plant. These materials will be denitrated and converted to $UO_2$ before subsequent carbonizing steps that are similar to those described for the IM material. The significance of the differences in histories is that recycle uranium may contain more nitrogen (from undecomposed nitrate) than does the initial or makeup 93% $^{235}$U.

There are limited data concerning the quantities of nitrogen in potential HTGR fuel since this fuel is not made on a routine basis. It is therefore assumed that all forms of $UC_2$ and $ThO_2$ contain the same quantity of nitrogen (i.e., 25 ppm) used in this report as an industry concensus for LWR fuels. On this basis, about 0.96 Ci of $^{14}$C/MTHM, or about 9.7 Ci/GW(e)-yr will be formed from the $^{14}$N(n,p) reaction.

Carbon-14 will also be formed to the extent of 0.225 Ci/MTHM, or 2.3 Ci/GW(e)-yr, from the reaction $^{17}$O(n,$\alpha$)$^{14}$C of oxygen present as $ThO_2$ (Table 5).

## 4.2 Formation in Graphite Blocks

Independently of the $^{14}$N(n,p)$^{14}$C reaction, significant quantities of $^{14}$C will be formed in graphite of fuel and reflector blocks due to the reaction $^{13}$C(n,$\gamma$)$^{14}$C. Based on a lifetime average ratio of 10.93 MTC in fuel blocks/MTHM, about 3.7 Ci of $^{14}$C/MTHM, or 37 Ci/GW(e)-yr, will be formed from this (n,$\gamma$) reaction (Table 5). Additional $^{14}$C will be formed in reflector blocks, which are present to the extent of 16.2% of fuel blocks on a lifetime average basis. The neutron flux in reflector blocks will be about 70 to 80% of the core-average flux, although the $^{14}$C production listed in Table 5 is based on a flux in these reflector blocks equal to the core average. The total $^{14}$C formed from the $^{13}$C(n,$\gamma$) reaction in fuel blocks and reflector blocks is less than 4.3 Ci/MTHM, or less than 43 Ci/GW(e)-yr.

The amount of nitrogen present in fuel-block or reflector-block graphite is uncertain. Four samples of graphite were irradiated in the Oak Ridge Research Reactor (ORR) and were subsequently analyzed for $^{14}$C.[46] The quantity of this nuclide in excess of that calculated to be formed from the $^{13}$C(n,$\gamma$)$^{14}$C reaction was ascribed to the reaction $^{14}$N(n,p)$^{14}$C. On the basis of this assumption, the equivalent nitrogen impurity was calculated to be 3.2 to 8.4 ppm on a graphite-weight basis. The only other estimate of nitrogen content in an in-use graphite is 26 ppm,[14] and is used here as the basis for the value of 30 ppm of nitrogen in fuel blocks and reflector blocks listed in Table 5. Carbon-14 formed in graphite containing 30 ppm of nitrogen corresponds to 12.6 Ci/MTHM or 127 Ci/GW(e)-yr.

Table 5. Production of $^{14}$C in graphite and fuel of High-Temperature Gas-Cooled Reactors

| Material | Impurity content | | Material in core (MT/MTHM) | Quantity of element in core (g/MTHM) | | | $^{14}$C existing 160 days after discharge of fuel (Ci/MTHM) | | | Total $^{14}$C | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | Nitrogen (ppm) | Oxygen (wt %) | | Carbon | Nitrogen | Oxygen | From carbon | From nitrogen | From oxygen | Ci/MTHM | Ci/GW(e)-yr[a] |
| Graphite in fuel blocks | 30[b] | | 10.93[c] | 1.093E+7 | 3.28E+2 | | 3.69 | 12.58 | | 16.27 | 164 |
| Graphite in reflector blocks | 30[b] | | 1.77[c] | 1.77E+6 | 3.54E+1 | | <0.60[d] | <2.04 | | <2.63 | <26.6 |
| IM uranium (UC$_2$) | 25[e] | | 0.04548[f] | | 2.50E+1 | | | 0.959 | | 0.044 | 0.44 |
| Recycle uranium (UC$_2$) | 25[e] | | 0.04512[f] | | 2.50E+1 | | | 0.959 | | 0.043 | 0.44 |
| Thorium dioxide | 25[e] | 12.12 | 0.90941[f] | | 2.50E+1 | 1.25E+5 | | 0.959 | 0.225 | 1.08 | 10.9 |
| Total | | | | | | | | | | <19.9[g] | 202[g] |

[a]Based on 10.11 MTHM/GW(e)-yr (equivalent to 38.5% efficiency in converting heat to electricity).

[b]This is an estimate based on the assumption that no great efforts will be made to minimize the nitrogen content.

[c]See ref. 13.

[d]Based on a neutron flux in reflector blocks equal to the core-average flux. However, the flux in the reflector blocks will be about 70 to 80% of the core-average value.

[e]Assumed to be the same as in LWR fuels.

[f]From ref. 13 the following values are obtained: 405.06 kg (93% $^{235}$U) IM material, 294.07 kg 23R material, 107.83 kg 25R material, and 8394.79 kg thorium in the lifetime average annual reload. Values listed are MT thorium or uranium/MTHM.

[g]All of this is potentially available for release at the fuel reprocessing plant except about 0.012 Ci/MTHM (0.12 Ci/GW(e)-yr) in the initially fissile particles of the 25R stream which are designated 25W after discharge.[13]

## 5.0 CARBON-14 FORMATION IN LIQUID-METAL FAST BREEDER REACTORS

The primary structural material of the core of an LMFBR will be 316 or A-286 stainless steel. Carbon-14 will be formed from impurities in this metal as well as in the fuel. Since no LMFBR has yet been built, discussion presented here is based on the proposed reference design[47] of the Clinch River Breeder Reactor (CRBR) and on recent updating of fuel composition.[48] A core element for this reactor is shown in Fig. 1.

### 5.1 Formation in the Fuel

In common with LWR fuels, $^{14}$C will be formed by the $^{17}$O(n,$\alpha$) and $^{14}$N(n,p) reactions in LMFBR fuels; in both types of reactor very small quantities of $^{14}$C will be formed by the $^{13}$C(n,$\gamma$) reaction. Two other reactions produce $^{14}$C in the LMFBR (Sect. 2): $^{15}$N(n,d) and $^{16}$O(n,$^3$He). Croff's[18] estimates of cross sections and formation rates are listed in Table 1. Production of $^{14}$C from reactions involving oxygen are listed in Table 6; these values are based on 8383 g-atoms of O/MTHM (in this case, MTHM is uranium plus plutonium) and 0.039 at. % of $^{17}$O in natural oxygen (corresponding to 3.27 g-atoms of $^{17}$O/MTHM).

The specification limit on the nitride nitrogen impurity in plutonium dioxide[49] and driver fuel[50] for the Fast Flux Test Facility (FFTF) is 200 ppm. Air in fuel rods is evacuated and replaced by high-purity helium[51] before the rods are closed by welding in a helium atmosphere. The maximum fuel-pellet gas content of 0.09 cc (STP) per gram of fuel,[50] exclusive of water, would correspond to 120 g of N/MTU if all the gas were nitrogen. Measured nitride nitrogen concentrations in FFTF fuels have been significantly less than specifications, generally in the 10 to 20 ppm range,[52] as shown in Table 3. Therefore, it is assumed in this report that the concentration of nitrogen in CRBR fuel will be about 25 ppm, with a range of 10 to 75 ppm. These values were used to estimate an average and range (Table 7) of $^{14}$C formation due to neutron absorption by $^{14}$N and $^{15}$N. The average value is 0.166 Ci of $^{14}$C/MTHM, or 6.1 Ci of $^{14}$C/GW(e)-yr; the values range from 0.0665 Ci/MTHM [2.45 Ci/GW(e)-yr] to 0.499 Ci/MTHM [18.4 Ci/GW(e)-yr]. Formation of $^{14}$C from oxygen in the fuel, 0.00364 Ci/MTHM, and from nitrogen would be equal if the nitrogen concentration in the fuel were about 0.55 ppm.

### 5.2 Formation in Core Hardware

As noted above, 316 stainless steel (with specifications listed in ref. 29) or A-218, is essentially the only metal in the CRBR core and may be the only metal in future commercial LMFBRs. Specification RDT M3-28T, Table 4, requires that the oxygen and nitrogen concentrations be lower than corresponding values for 304 stainless steel used in LWRs. In particular, the specification of $\leq$0.010 wt % of nitrogen in 316 stainless steel is more than a factor of 10 below the specification of 0.10 to 0.16 wt % of nitrogen in 304 stainless steel for LWR applications.

Calculated quantities of $^{14}$C to be formed in CRBR cladding are listed in Table 7. These are based on 100 ppm (0.01 wt %) of nitrogen and on the "mass ratios" shown in Table 6. These ratios refer only to cladding plus shroud plus wire between bottom and top fuel elevations. The neutron flux decreases very rapidly with elevation away from fuel levels. For this reason, $^{14}$C formation in regions above the fuel level in the upper axial blanket and below the fuel level in the lower axial blanket is neglected.
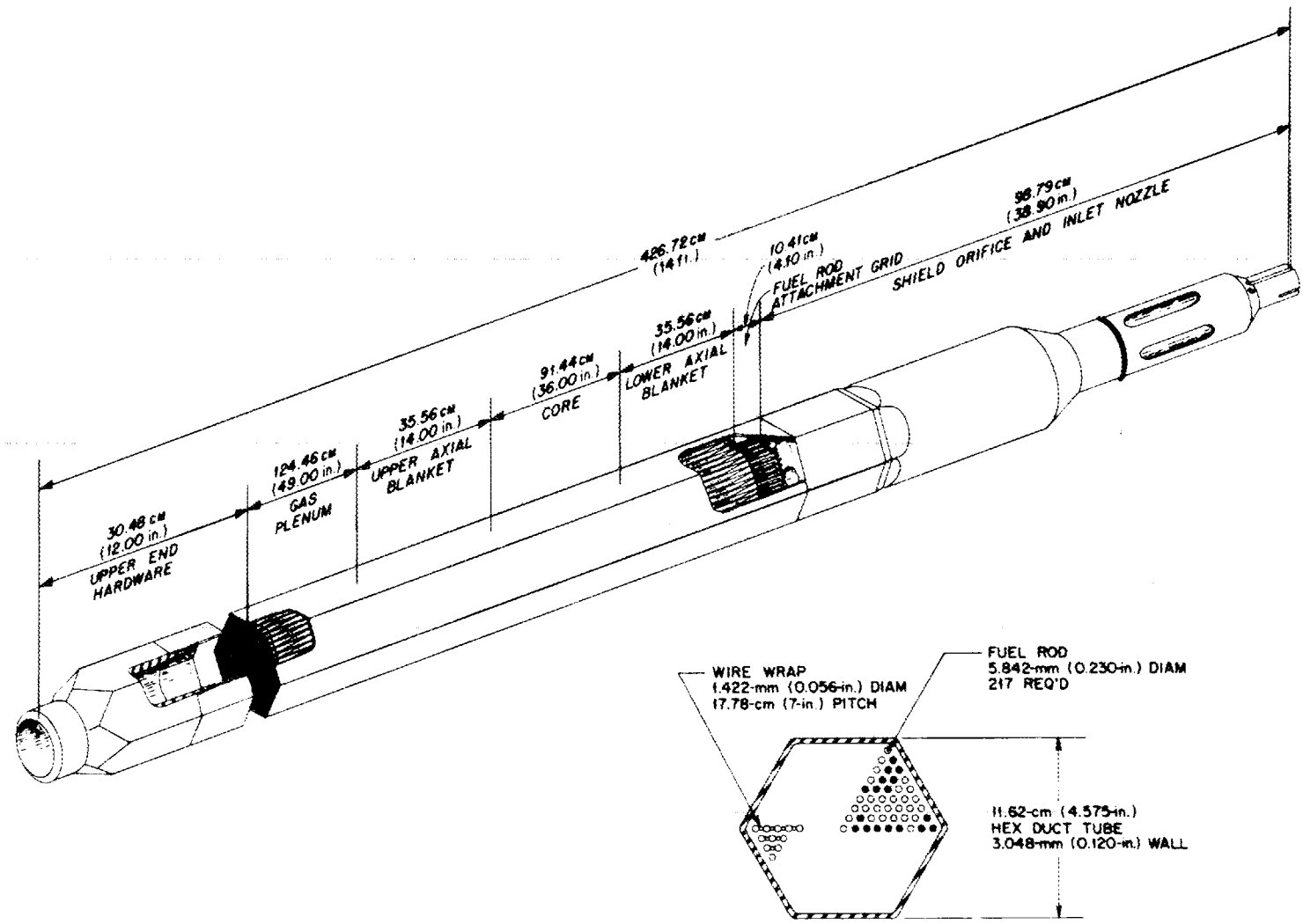
Fig. 1.  Reference CRBR core fuel assembly.

Table 6. Data pertaining to $^{14}C$ production in the CRBR

| CRBR region | Specific power[a] $\left[\dfrac{MW(t)}{MTHM}\right]$ | Mass of HM[a,b] charged (MT) | Mass of stainless steel[a,b] (MT) | Mass ratio[b] $\left(\dfrac{MTSS}{MTHM}\right)$ | ORIGEN - calculated burnup $\left[\dfrac{MW(t)\text{-}d}{MTHM}\right]$ | Specific production of $^{14}C$ from | | |
|---|---|---|---|---|---|---|---|---|
| | | | | | | Carbon $\left(\dfrac{Ci}{g\ C}\right)$ | Nitrogen $\left(\dfrac{Ci}{g\ N}\right)$ | Oxygen $\left(\dfrac{Ci}{100\ kg\ O}\right)$[c] |
| Inner core | 113.22 | 1.4361 | 10.93 | 0.66 | 93,066 | 9.98E-9 | 1.88E-2 | 8.39E-3 |
| Outer core | 104.63 | 1.2006 | 9.11 | 0.66 | 86,005 | 6.92E-9 | 1.32E-2 | 5.48E-3 |
| Upper axial blanket | 3.482 | 1.0361 | 8.40 | 0.66 | 2,862 | 1.47E-9 | 2.85E-3 | 1.03E-3 |
| Lower axial blanket | 7.276 | 1.0361 | 7.77 | 0.66 | 5,981 | 2.66E-9 | 5.13E-3 | 1.92E-3 |
| Radial blanket | 4.302 | 3.0373 | 20.04 | 0.185 | 3,536 | 1.75E-9 | 3.39E-3 | 1.24E-3 |
| Total in reactor | | 32.3505 | 56.25 | 0.393 | | | | |
| Mass-average | 30.184 | | | | 24,811[d] | | | |

[a]See Ref. 48.

[b]The heavy metal (HM) charge is the annual charge; annually, one-third of the core and axial blankets and one-sixth of the radial blankets are replaced. The stainless-steel mass is the total in the specified region, not just the fresh steel. The mass ratio of stainless steel to heavy metal [(MTSS/MTHM), column 5)] is the sum (cladding mass + shroud mass + wire mass) between the bottom and top fuel elevations, Fig. 1, per unit mass of heavy metal. Calculations are based on the following data for core and axial blanket tubes (fuel pins, see Fig. 1): OD = 0.230 in.; ID = 0.200 in,; wire-rod spacer (running nearly coaxially with fuel pin) = 0.055 in. diam; hex face-to-face distance = 4.575 in.; hex metal thickness = 0.120 in.; fuel diameter = 0.200 in.; density of stainless steel = 8.02 g/cm$^3$; density of fuel ($UO_2$) = 9.316 (85% of theoretical 10.96 g/cm$^3$). The radial blanket fuel rod dimensions are: OD = 0.520 in.; ID = 0.490 in.; fuel diam = 0.485 in.; all other parameters are as given above.

[c]From the stoichiometry of $(U,Pu)O_2$, there are about 134 kg O/MTHM.

[d]This corresponds to 36.80 MTHM/GW(e)-yr, as used in Table 7.

Table 7. Production of $^{14}C$ in the CRBR[a]

| CRBR region | Production of $^{14}C$ in fuel from | | | | | | | | Production of $^{14}C$ from nitrogen in stainless steel | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Oxygen | | Nitrogen | | | | | | | |
| | | | Low ( 10 ppm ) | | Average (25 ppm) | | High ( 75 ppm) | | | |
| | Ci/MTHM | Ci/GW(e)-yr | Ci/MTHM | Ci/GW(e)-yr | Ci/MTHM | Ci/GW(e)-yr | Ci/MTHM | Ci/GW(e)-yr | Ci/MTHM | Ci/GW(e)-yr |
| Inner core | 1.13E-2 | 1.11E-1 | 1.88E-1 | 1.84E+0 | 4.70E-1 | 4.61E+0 | 1.41E+0 | 1.38E+1 | 1.24E+0 | 1.22E+1 |
| Outer core | 7.35E-3 | 7.80E-2 | 1.32E-1 | 1.40E+0 | 3.30E-1 | 3.50E+0 | 9.90E-1 | 1.05E+1 | 8.73E-1 | 9.27E+0 |
| Upper axial blanket | 1.39E-3 | 4.43E-1 | 2.85E-2 | 9.09E+0 | 7.12E-2 | 2.27E+1 | 2.14E-1 | 6.82E+1 | 1.88E-1 | 6.01E+1 |
| Lower axial blanket | 2.58E-3 | 3.94E-1 | 5.13E-2 | 7.83E+0 | 1.28E-1 | 1.96E+1 | 3.85E-1 | 5.87E+1 | 3.39E-1 | 5.18E+1 |
| Radial blanket | 1.67E-3 | 4.31E-1 | 3.39E-2 | 8.76E+0 | 8.48E-2 | 2.19E+1 | 2.54E-1 | 6.57E+1 | 6.27E-2 | 1.62E+1 |
| Mass-average | 3.64E-3 | 1.34E-1 | 6.65E-2 | 2.45E+0 | 1.66E-1 | 6.12E+0 | 4.99E-1 | 1.84E+1 | 3.49E-1 | 1.28E+1 |

[a]Calculations do not include formation of $^{14}C$ in stainless steel above the top or below the bottom of the fuel.

## 6.0 COMPARISONS AND DISCUSSIONS

Calculated quantities of $^{14}$C that are or will be produced in the four types of reactors (BWR, PWR, HTGR, and LMFBR) considered in this report are summarized in Table 8 in units of Ci/GW(e)-yr. Ranges are given for all calculated values of $^{14}$C from all reactors except the HTGR. The ranges are due to variations in the nitrogen content of the fuel. Values spanning the full range of 10 to 75 ppm (by weight) are shown in Table 3, which is a summary of manufacturing data.

The Barnwell plant of Allied General Nuclear Services is designed to process about 5 MTHM/day, or 1500 MTHM/yr, of LWR fuel. Heavy metal (HM) is uranium or uranium plus plutonium charged to BWR, PWR, and LMFBR; HM is also uranium plus thorium charged to the HTGRs. The Barnwell design corresponds to about 45 GW(e)-yr. Similarly, reference HTGR- and LMFBR-fuel reprocessing plants are designed to process annually fuel that produced about 45 GW(e)-yr of energy. Using this factor as a multiplier for values listed in Table 8, it is appropriate to examine the total quantities of $^{14}$C that would be released from the various fuel reprocessing plants if equipment is not installed to collect and retain the gases containing this nuclide; it is also appropriate to examine how much will be contained within the hardware that becomes part of the high-level waste that may be shipped to a Federal repository. Light-water reactor fuel processed in 1 year in a Barnwell-sized plant will contain 400 to 2200 Ci of $^{14}$C; the hardware will contain 1400 to 2700 Ci of $^{14}$C. The calculated values for $^{14}$C in the hardware are conservatively high since they are based on the assumption that all core hardware -- not just the cladding - is in as intense a flux field as is the cladding.

Lesser quantities of $^{14}$C will be produced in LMFBR fuel. The fuel entering a reprocessing plant of 45 GW(e)-yr capacity will contain 100 to 800 Ci of $^{14}$C per year while the cladding will contain about 600 Ci of $^{14}$C per year. Quantities of this nuclide in other hardware are not included in Table 8.

The $^{14}$C content of HTGR fuel entering a 450 MTHM/yr [45 GW(e)-yr] fuel reprocessing plant in 1 yr will be about 530 Ci if the nitrogen content of the fuel is 25 ppm. Only this "median" nitrogen content is considered because the graphite probably will be the dominant source of $^{14}$C. In particular, if there is no nitrogen in the graphite, the $^{14}$C content [due solely to the $^{13}$C(n,$\gamma$)$^{14}$C reaction] of graphite entering the fuel reprocessing plant in 1 yr will be about 1660 Ci; the $^{14}$N(n,p)$^{14}$C reaction will add about 5660 Ci of $^{14}$C if the nitrogen content of the graphite is 30 ppm. The value of <200 Ci of $^{14}$C/GW(e)-yr shown in Table 8 for the HTGR corresponds to <9000 Ci entering the fuel reprocessing plant each year. These maxima include $^{14}$C in reflector blocks as well as in fuel blocks. There is no metallic hardware in an HTGR corresponding to cladding and other structural components of the LWRs and LMFBRs.

### 6.1 Comparisons of Reactor Produced and Naturally Produced $^{14}$C

The natural rate of $^{14}$C formation in the atmosphere from cosmic-ray induced reactions and the contribution of $^{14}$C to the total radiation dose to man are valid bases for evaluating the impact of reactor-generated quantities of this nuclide. Lingenfelter[51] reported a global average production rate of 2.50±0.50 $^{14}$C atoms cm$^{-2}$ sec$^{-1}$ over the ten solar cycles prior to 1963. Reference has been made to this value by Lal and Suess[3] and in the UNSCEAR 1972 report.[54] Using 5.1E18 cm$^2$ as the earth's surface area,[55] Lingenfelter's value corresponds to (4.2±0.8)E4 Ci of $^{14}$C/yr. More recently, Light et al.[56] have calculated the average production rate from 1964 to 1971 to be 2.21±0.10 $^{14}$C atoms

Table 8. Comparison of $^{14}$C production in different types of reactors in units of Ci/GW(e)-yr[a]

| Reactor | In fuel | Cladding and core structural materials | In coolant | | Total calculated |
|---|---|---|---|---|---|
| | | | Calculated | Observed | |
| BWR | | 43.3-60.4 | 4.7 | 8 [b] | |
| Low value | 9.0 | | | | 57 |
| Median value | 17.6 | | | | 74 |
| High value | 46.3 | | | | 111 |
| PWR | | 30.5-41.6 | 5.0 | 6 | |
| Low value | 9.6 | | | | 44 |
| Median value | 18.8 | | | | 59 |
| High value | 49.5 | | | | 96 |
| HTGR | | <190 | nil | N.A.[c] | |
| Median value | 12.0 | | | | <200 |
| LMFBR | | 12.8 | nil | N.A.[c] | |
| Low value | 2.6 | | | | 15 |
| Median value | 6.3 | | | | 19 |
| High value | 18.5 | | | | 31 |

[a] Reactor parameters pertaining to these calculations based on the ORIGEN program are as follows: BWR, 18.823 MW(t)/MTU, 4 years in reactor, to 27,500 MWd/MTU; 2.6 wt % $^{235}$U; 33% thermal efficiency. PWR, 30.0 MW(t)/MTU, 3 years in reactor, to 33,000 MWd/MTU; 3.3 wt % $^{235}$U; 33% thermal efficiency. HTGR, 64 MW(t)/MTHM, 4 years in reactor, to 95,000 MWd/MTU; 38.5% thermal efficiency; see Table 5 for fuel compositions. LMFBR, 30.18 MW(t)/MTHM (mass average), 75% on-stream time for 3 years, to 24,800 MWd/MTU (mass average); 35% thermal efficiency; see Table 6 for fuel-region specifications.

[b] A value of 9.1 Ci/GW(e)-yr is presented in the following report, issued as the present report was in the final stage of preparation: R. L. Blanchard, W. L. Brinck, H. E. Kolde, H. L. Krieger, D. M. Montgomery, S. Gold, A. Martin, and B. Kahn, Radiological Surveillance Studies at the Oyster Creek BWR Nuclear Generating Station, USEPA, EPA-520/5-76-003 (June 1976).

[c] N.A. = not applicable.

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$cm^{-2}$ $sec^{-1}$. Based on projections of sunspot numbers for the remainder of the solar cycle, they also estimate that the 11-yr mean rate could be as large as $2.28\pm0.10$ $^{14}C$ atoms $cm^{-2}$ $sec^{-1}$. (The error limits on the rates apply only to the statistics of the calculation.) This value corresponds to $(3.8\pm0.2)E4$ Ci of $^{14}C/yr$. Thus, to one significant figure, the 11-yr average natural rate of production is $4.E4$ Ci of $^{14}C/yr$. On this basis, the quantity of $^{14}C$ in fuel annually entering an LWR fuel reprocessing plant with a capacity of 1500 MTHM/yr [equivalent to 45 GW(e)-yr and about fifty 1000 MW(e) reactors] is 1 to 5.5% of the natural production rate; corresponding values for $^{14}C$ entering an LMFBR fuel reprocessing plant are 0.3 to 2.0% of the natural production rate. The 1660 (from graphite only) to 9000 (from graphite, oxygen, 25 ppm of nitrogen in fuel, and 30 ppm of nitrogen in all graphite) Ci of $^{14}C$ annually entering the HTGR fuel reprocessing plant, of the same 45 GW(e)-yr equivalent capacity, corresponds to 4 to 22% of the natural rate of production of this nuclide.

## 6.2 Worldwide and Local Radiation Doses from Reactor-Produced $^{14}C$

World population radiation doses from all forms of radiation and from naturally produced $^{14}C$ provide a second form of comparison of the effects of discharge of this nuclide from fuel reprocessing plants. World-wide dose rates to gonads, bone-lining cells, and bone marrow due to internal and external irradiation from all natural sources in "normal" areas are about 90 mrad/yr (Table 20 of ref. 54, UNSCEAR 1972). Oakley[57] reports a gonadal dose equivalent to the population of the United States from all natural sources of 88 mrem/yr. The contribution of $^{14}C$ to this total is about 0.7 to 0.8 mrad/yr.[54] Other values of the contribution of $^{14}C$ to the total have been as high as 1.6 mrem/yr.[13,58] Thus, based on the percentages listed above and a nominal 1 mrem/yr due to natural $^{14}C$, after this nuclide becomes uniformly distributed over the earth, additional radiation doses due to $^{14}C$ will be in the range 0.004 to 0.06 mrem/yr for discharges from an LWR fuel reprocessing plant of capacity equivalent to 45 GW(e)-yr; corresponding incremental doses due to $^{14}C$ discharges from equivalent LMFBR and HTGR fuel reprocessing plants will be in the range 0.0004 to 0.023 mrem/yr and 0.035 to 0.19 mrem/yr, respectively.

Potential radiological impacts of annual releases of 5000 Ci of $^{14}C$ on the population out to 50 miles from a fuel reprocessing plant have been analyzed by Killough et al.[59] Three techniques for reducing these local population doses were: (1) use of a discharge stack up to 1000 ft tall; (2) heating of the discharged gas to obtain a large effect of buoyancy to increase the effective stack height; and (3) use of nocturnal, rather than continuous, emission in order to minimize the availability of the discharged $^{14}C$ for uptake by vegetation. Using meteorological data for the Oak Ridge, Tennessee, area and a 300-ft stack, the total-body dose of a population of $10^6$ people within the 50-mile radius was 110 person-rem/yr; the average individual dose was 0.107 mrem/yr, and the maximum dose to "fence-post man" (who spends all his time at 1.5 miles from the stack and eats food grown only at this location) was 240 mrem/yr.

## 6.3 Other Predictions of $^{14}C$ Formation Rates

Table 9 summarizes predictions of $^{14}C$ formation rates in BWR and PWR fuels presented in this and other reports.[60-64] Calculated formation rates in BWR fuels range from 13.6 to 22 Ci/GW(e)-yr. In the BWR coolant, from the $^{17}O(n,\alpha)$ reaction only, the range is 4.7 to 9.9 Ci/GW(e)-yr.

Table 9. Comparisons of some estimates of $^{14}$C production rates[a] in LWRs
(values are in Ci of $^{14}$C/GW(e)-yr)

| Reactor type | Region of $^{14}$C formation | Parent nuclide | Source of information | | | | |
|---|---|---|---|---|---|---|---|
| | | | Bonka et al.[b] | Kelly et al.[c] | NUREG[d] | Fowler et al.[e] | This report[f] |
| BWR | Fuel | $^{14}$N | 12.9 | 10.9 | NC[g] | 18. | 11.5 |
| | | $^{17}$O | 8.4 | 2.7 | NC | 4. | 3.3 |
| | | $^{14}$N + $^{17}$O | 21.3 | 13.6 | NC | 22. | 14.8 |
| | Coolant | $^{14}$N | 1.3 | NC | NC | 0.26 | NC |
| | | $^{17}$O | 9.9 | NC | 9.5 | 8.9 | 4.7 |
| PWR | Fuel | $^{14}$N | 12.2 | 10.9 | NC | 18. | 12.2 |
| | | $^{17}$O | 7.1 | 2.7 | NC | 4. | 3.5 |
| | | $^{14}$N + $^{17}$O | 19.3 | 13.6 | NC | 22. | 15.7 |
| | Coolant | $^{14}$N | 1.28 | NC | NC | 0.09 | NC |
| | | $^{17}$O | 9.8 | NC | 8 | 3.2 | 5.0 |

[a]Based on 20 ppm nitrogen (by weight) in the $UO_2$ except for Bonka et al.,[60] whose basis is not given.
[b]Ref. 60.
[c]Ref. 61.
[d]Parameters in ref. 62 for the BWR and in ref. 63 for the PWR correspond to about 0.9 GW(e)-yr. Thus, values in this column, which are taken from these references, should be increased about 10%.
[e]Ref. 64.
[f]Calculations pertaining to $^{14}$C produced in the BWR cooling water are based on the assumption that there is no void volume in the core due to steam.
[g]NC means not calculated.

Corresponding values in PWR fuels also range from 13.6 to 22 Ci/GW(e)-yr, and in PWR coolant they range from 3.2 to 9.8 Ci/GW(e)-yr. Carbon-14 formation rates in cooling water from the $^{14}N(n,p)$ reaction are small and uncertain, since data on concentrations of nitrogen are nearly nonexistent. When the uncertainties in cross-section data are combined with the varying choices of other nuclear parameters used by these different authors, it is perhaps not unexpected that the largest values are about twice the smallest.

Bonka et al.[60] give $^{14}C$ production rates from nitrogen in the fuel and coolant of LWRs. These authors list the 2200-m/sec cross sections for the $^{13}C(n,\gamma)^{14}C$, $^{14}N(n,p)^{14}C$, and $^{17}O(n,\alpha)^{14}C$ reactions without stating whether they used these or cross sections collapsed according to reactor fluxes. They also do not indicate the nitrogen content of the fuel or cooling water. Thus, it is not possible to comment on the agreements and differences between the values of Bonka et al.[60] and those of other authors listed in Table 9.

Kelly et al.[61] give $^{14}C$ production rates 5 to 23% lower than values in this report (Table 9). These authors also present only the 2200-m/sec cross sections for reactions 1, 2, and 5; they do not discuss collapsing cross-section data in terms of the fluxes of specific reactors. Again, no comparison can be made between their model reactors and those of this report.

The U.S. Nuclear Regulatory Commission (NRC) has presented an estimate of 9.2 Ci of $^{14}C$/yr formed in the cooling water of a BWR[62] and of 8 Ci/yr in the cooling water of a PWR.[63] Both values are based only on the $^{17}O(n,\alpha)^{14}C$ reaction; formation of $^{14}C$ from the $^{14}N(n,p)$ reaction is considered to contribute only a small fraction of 1 Ci/yr because of the low concentration of $^{14}N$ in the reactor coolant (less than 1 ppm by weight). The calculational procedure of the NRC reports includes use of an average flux of 3.0E+13 neutrons $cm^{-2}$ $sec^{-1}$ and a thermal neutron cross section for $^{17}O$ of 0.24 b for both BWR and PWR; the masses of water in the reactor cores are 39 and 33 MT, respectively. The product of flux and cross section corresponds to 7.2E-12 atoms of $^{14}C$ per second per atom of $^{17}O$.

Fowler et al.[64] wrote a technical note partly to elicit comments concerning EPA calculations of $^{14}C$ source terms and the radiological impact of this nuclide. The EPA has already published[65] proposed standards pertaining to releases of $^{85}Kr$, $^{129}I$, and certain long-lived transuranic nuclides from nuclear power operations; no standard pertaining to $^{14}C$ was proposed, because the knowledge base available (in 1975) was considered inadequate for such a proposal. Calculations in the technical note are based on assumptions of a flux of 5.0E+13 neutrons $cm^{-2}$ $sec^{-1}$, an effective cross section of 1.1 b for the $^{14}N(n,p)^{14}C$ reaction, and an effective cross section of 0.14 b for the $^{17}O(n,\alpha)C^{14}$ reaction, for both the BWR and the PWR. This choice of flux and cross sections corresponds to 5.5E-11 atoms of $^{14}C$ per second per atom of nitrogen, and 7.0E-12 atoms of $^{14}C$ per second per atom of $^{17}O$, respectively, for both the BWR and the PWR. These authors[64] also calculated a source term for $^{14}C$ formation from 1 ppm of nitrogen dissolved in the cooling water. This use of 1 ppm is arbitrary since essentially no data are available on this concentration at operating reactors, as discussed in Sect. 3.3. The calculations with 1 ppm of nitrogen were made because similar sample calculations had been made in draft regulatory guides.[66,67] However, such calculations are not made in refs. 62 and 63 which were developed from these drafts.

Calculations in this report are based on parameters listed in footnote a of Table 8 and in Sect. 3.1. From the effective fission cross sections (p. 72, Table A-1, of ref. 1), the ORIGEN code calculates average fluxes of 2.07E+13 and 2.92E+13 neutrons $cm^{-2}$ $sec^{-1}$ for BWR and PWR, respectively. However, the initial and final fluxes for the BWR are 2.00E+13 and 2.26E+13, and initial and final fluxes for the PWR are 2.58E+13 and 3.45E+13 neutrons $cm^{-2}$ $sec^{-1}$. The average formation rates for a BWR are, therefore, 3.06E-11 atoms of $^{14}C$ formed per second per atom of $^{14}N$

present and 3.79E-12 atoms of $^{14}C$ formed per second per atom of $^{17}O$ present; corresponding values for a PWR are 4.32E-11 and 5.34E-12. Thus, the $^{14}C$ formation rates calculated in this report for the $^{14}N(n,p)$ reaction are only 55% (for the BWR) and 79% (for the PWR) as large as values presented by Fowler et al.[64] Carbon-14 formation for the $^{17}O(n,\alpha)$ reaction rates in this report are only 53% (for the BWR) and 74% (for the PWR) as large as values in refs. 62 and 63; they are only 54% (for the BWR) and 76% (for the PWR) as large as values in ref. 64.

Cross sections listed in Table 1 are the current best estimates for application to the steady state of reactor operations (after the first few reloads). The most recent (1974) revisions (soon to be incorporated in the ORIGEN library) of $^{14}N$ cross sections for use in the ENDF/B-IV library[20] were presented by Young, Foster, and Hale,[68] largely from an earlier review by Young and Foster.[69] Croff[70] has used this revision and the XSDRNPM computer program[71] to obtain a one-group value of 1.45 b for the effective thermal cross section for the $^{14}N(n,p)^{14}C$ reaction for LWRs. This is very close to the value 1.48 b used in this report.

## 6.4 Comparison with Releases from Russian Reactors

Rublevskii et al.[72] have presented data, listed in Table 10, on measured releases of $^{14}C$ from five Russian reactors. These authors combined their data with Spinrad's[73] projections concerning world-wide installed nuclear power to estimate the magnitude of $^{14}C$ discharges to the year 2010. Neglecting the small Obninsk and ARBUS reactors, the data in Table 10 show releases *at the reactor stations* of 200 to 800 Ci of $^{14}C$/GW(e)-yr. These values are far in excess of the 6 Ci/GW(e)-yr reported by Kunz et al.[11] for the Ginna, Indian Point 1, and Indian Point 2 PWRs, and of the 8 Ci/GW(e)-yr for the BWR at Nine Mile Point.[12] The reported releases of $^{14}C$ from Russian reactors are thus seen to be about of 10 to 100 times greater than corresponding releases from the four-mentioned American reactors. Such a discrepancy implies that Rublevskii et al.[72] have grossly overestimated the potential releases of $^{14}C$ from non-Russian nuclear reactors, and that a need exists for an analysis of the origin of $^{14}C$ formation in the Russian reactors. This overestimation appears in their conclusions that the daily production rates of $^{14}C$ in water-cooled, graphite moderated reactors and in water-cooled, water moderated reactors (LWRs) are 0.75 and 0.25 mCi/MW(t), respectively. The latter value corresponds to about 300 Ci/GW(e)-yr, which is 40 to 50 times greater than was observed by Kunz et al.[11,12] Apparently, a detailed description is not now available. However, on visits to Russian nuclear stations, Lewin[74] was advised that nitrogen gas is used to blanket the graphite of the pressure-tube reactors, such as those at Beloyarsk and Sosnovyi Bor (near Leningrad).[75,76] In addition, a pressurized water reactor VVER-210 at Novovoronezh[75] (Table 10) has been reported[77] to use nitrogen gas for pressurization; finally, hydrazine and ammonium hydroxide are used in the primary cooling water to minimize radiolytic oxygen formation, and for corrosion and pH control. Later PWRs constructed at Novovoronezh do not use nitrogen pressurization; instead, steam is heated electrically by a method similar to that used in the PWRs in the United States.[34-36, 40]

## 6.5 Reducing the Releases of $^{14}C$

Releases of $^{14}C$ can be reduced by reducing the amount that is formed in nuclear reactors, by collecting it at the reactor station and at the fuel reprocessing plant and converting most of it to solid form for permanent retention, or by a combination of these methods. Snider and Kaye[79] have

Table 10. Carbon-14 entering the atmosphere with gaseous wastes from some Russian reactors[a]

| Reactor type | Rated thermal power [MW(t)] | Power rating during studies [MW(t)] | $^{14}C$ discharged $\left(\dfrac{mCi}{day}\right)$ | $^{14}C$ discharged $\left[\dfrac{Ci}{GW(e)-yr}\right]$ [b] |
|---|---|---|---|---|
| Water-cooled, graphite moderated APS[c], USSR Academy of Science, Obninsk[d] | 30 | 12 | 9 ± 3 | 900 ± 300 |
| Water-cooled, graphite moderated (AMB), Beloyarsk APS [d,e] | 285 | 210 | 140 ± 50 | 800 ± 300 |
| Water-cooled, water moderated (VVER-210), Novovoronezh APS[c] (PWR)[e] | 760 | 740 | 120 ± 30 | 200 ± 50 |
| Water-cooled, water moderated (VK-50), (Boiling water test reactor) Ulyanovsk APS[c] | 150 | 90 | 30 ± 10 | 400 ± 130 |
| Organic moderated and cooled test reactor (ARBUS) | 5 | 5 | 0.6 ± 0.2 | 150 ± 50 |

[a]See ref. 72.

[b]Based on an assumed thermal-to-electrical efficiency of 30%, as used in ref. 72.

[c]APS = atomic power station.

[d]A pressure-tube reactor of which the two 1000 MW(e) units at Sosnovyi Bor (near Leningrad) are the most modern counterparts.

[e]Equivalent to a United States pressurized water reactor.

recently analyzed many process options and the effects on the environmental impact of $^{14}C$ releases. Reducing the quantity of $^{14}C$ formed requires that the nitride nitrogen impurity content of the fuel be reduced, and that air be removed from each fuel rod in a vacuum degassing step before the second end of the rod is closed by welding. Such reduction to a maximum of 10 ppm of nitrogen by weight is a goal that one fuel manufacturer (1 of Table 3) has already achieved and that two fuel manufacturers (2 and 3 of Table 3) could achieve without much technical or economic impact, but which the other two could not easily achieve. When the nitrogen content is reduced to 5.7 ppm (Sect. 3.1), the quantity of $^{14}C$ formed from the $^{17}O(n,\alpha)$ reaction equals that formed from the $^{14}N(n,p)$ reaction in LWR fuels.

Retaining carbon dioxide in nuclear fuel reprocessing plants is another alternative now being investigated for minimizing discharges of $^{14}C$ to the environment. The fluorocarbon absorption process,[80] now in the pilot plant stage of development for the recovery of krypton from the off-gas of LWR and LMFBR-fuel reprocessing plants, also collects $CO_2$ in the fluorocarbon solvent. The $CO_2$ so collected could be discharged into a slurry of $Ca(OH)_2$[81] and converted to $CaCO_3$ for permanent storage. Similarly, the KALC process[82,83] (Krypton Absorption in Liquid Carbon Dioxide) to recover and retain krypton in the carbon dioxide gas stream of an HTGR fuel reprocessing plant is also in the pilot plant stage of development. The $^{14}C$-containing carbon dioxide of this process could also be converted[14] to $CaCO_3$.

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