Carbon-14 Production in Fusion Reactors

R. D. Scheele
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September 1976

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SUMMARY

Calculations based on existing composition data were performed to estimate the order of magnitude and the final location of ^{14}C in fusion reactors. These calculations indicate that approximately 8 Ci/day, formed principally by ^{14}N activation, will be produced in the UWMAK-II reference reactor (5,000 MWth). If Nb-1% Zr is used as the structural material instead of stainless steel 316 this quantity will be more than doubled. No information is available on the form of the ^{14}C produced, but reduced forms such as carbides, hydrocarbons and perhaps CO may be produced. Most of the ^{14}C may remain fixed in structural and other reactor materials until the material is reclaimed. Activation of air in the plasma chamber would be an immediate concern.

There is presently a lack of developed technology for control and fixation of ^{14}C . Systems which could be used include collection with molecular sieves or cryogenic traps followed by fixation as calcium carbonate. This assumes that CO_2 and CO and gaseous hydrocarbons are the species produced. Studies should be performed to determine the chemical form of the ^{14}C produced in the plasma, in the different materials, and its ease of removal. Transfer between components should be investigated. A review should be made of the nitrogen impurity levels in reactor component materials and methods of removal. Finally some consideration must be given to methods of fixation and storage.

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CARBON-14 PRODUCTION IN FUSION REACTORS

INTRODUCTION

With expanded efforts and continued advances in plasma physics, engineering, and materials in the fusion reactor field, more attention is being directed towards environmental aspects.

Tritium has been the isotope of major concern because of the tremendous amounts involved in fusion reactors. Metal activation products have also received considerable attention. Although the behavior of carbon-14 (^{14}C) as well as its natural production, distribution, reactions, and biological impact have been well documented, the contribution to the world's ^{14}C inventory from the nuclear industry has only recently been addressed. Leipunskii,⁽¹⁾ Pauling,⁽²⁾ and Totter⁽³⁾ reviewed in 1957 and 1958 the effects of ^{14}C and the significant contribution from weapons testing. However, it was not until a decade later that Harkness and Walton⁽⁴⁾ pointed out that by 1963 the ^{14}C inventory in the troposphere of the Northern Hemisphere had doubled while the dilution by "dead" fossil fuel carbon was only 3%. Since 1973, Bonka,⁽⁵⁾ Rublevskii,⁽⁶⁾ Magno,⁽⁷⁾ Brooks,⁽⁸⁾ and Snider⁽⁹⁾ have recently reviewed the ^{14}C production from the nuclear power industry. As a result of these studies, the sources and production rates are reasonably well established. The chemical form of the ^{14}C and the degree of release has not in most instances been experimentally determined. The behavior of CO_2 which is the eventual form of most of the ^{14}C is well known, and its relatively short turnover time of approximately 3 years⁽⁴⁾ in the biosphere and eventual irreversible deposition in the deep ocean mitigates the effect of the long ^{14}C decay half-life of 5,720 years. The weapons ^{14}C decay data presented by Brooks⁽⁸⁾ suggest an atmospheric half-life of about 10 to 20 years.

A preliminary study dealing with information requirements necessary for fusion reactor environment support statements was assembled by Battelle, Pacific Northwest Laboratory in 1975.⁽¹⁰⁾ The present report considers a single problem, ^{14}C from fusion reactors, the sources, amounts, final location, and form.

There are two basic types of fusion reactor concepts. One concept employs magnetic fields to confine the plasma while the other uses an inertially confined plasma and high energy lasers to ignite the deuterium-tritium pellets. For the latter case, the conceptual design is less well defined, and the present discussion will be directed towards the magnetically confined type. UWMAK-II will be used as the reference reactor, but data for other types will be considered. Reactor descriptions can be found in Reference 11 and specific design details are available from design documents. (12-20)

DISCUSSION

PRODUCTION CALCULATION

Carbon-14 is produced by neutron activation reactions. The (D+T) plasma reaction which is to be used in first generation reactors produces 17.6 MeV per (D+T) reaction; included in this 17.6 MeV is a 14.1 MeV neutron. Therefore, the neutron spectrum is of high energy. The spectrum expected for the Princeton Tokamak can be found in their reference document.⁽¹²⁾

Table 1 lists the neutron reactions which involve ^{14}C along with their cross-sections for thermal and 14-MeV neutrons. Reactions 5 and 9 in Table 1 can be ignored since the threshold energies are greater than 14 MeV while Reactions 7 and 8 added a negligible amount due to the low concentration of ^{15}N . Reactions 1, 3, and 4 then will be the significant production paths. Thus, the quantities of ^{14}C produced will depend on the concentration of air (or nitrogen) in gas or vacuum components and on the level of impurities (primarily nitrogen) in reactor components. It may be noted that the burnout of ^{14}C (Reaction 6) will be negligible.

The accurate determination of ^{14}C production in fusion reactors would require extensive multigroup calculations for the total neutron spectrum as the neutron absorption cross-sections for ^{13}C and nitrogen-14 (^{14}N) vary over the energy range; oxygen-17 (^{17}O) has a constant cross-section over the energy range of interest. To simplify the calculations an approximation of the average energy must be made. Based on the Princeton Tokamak,⁽¹²⁾ neutron energy varies essentially from 1 eV to 14 MeV, with an average value of 10 keV. In this energy range the neutron cross-section of ^{14}N , ignoring resonances, is relatively constant. Therefore, the cross-section, 7.7×10^{-2} b, at 14 MeV is used in the calculations. For $^{13}\text{C}(n,\gamma)^{14}\text{C}$, a cross-section of 10^{-6} b is used. The flux used in the average total flux, Figure 1 and Table 2, for a particular zone in UWMAK-II.⁽¹⁴⁾

TABLE 1. Carbon-14 Production Reactions

Reaction	Energy Threshold, MeV	Isotopic Abundance, %	Cross-Section, barns	
			Thermal ⁽²⁰⁾	14.1 Mev
1. $^{14}\text{N}(n,p)^{14}\text{C}$	0	99.634	1.81	7.7×10^{-2} (21)
2. $^{12}\text{C}(n,\gamma)^{13}\text{C}$	0	98.893	3.4×10^{-3}	
3. $^{13}\text{C}(n,\gamma)^{14}\text{C}$	0	1.107	0.9×10^{-3}	$\sim 1 \times 10^{-10}$ (22)
4. $^{17}\text{O}(n,\alpha)^{14}\text{C}$	0	0.0374	0.235	0.25 (22)
5. $^{17}\text{O}(n,^3\text{He})^{14}\text{C}$	19.85	0.204		
6. $^{14}\text{C}(n,\gamma)^{15}\text{C}$	0	N	$< 1 \times 10^{-6}$	
7. $^{15}\text{N}(n,d)^{14}\text{C}$	8.5	0.366		
8. $^{15}\text{N}(n,np)^{14}\text{C}$	10.9	0.366		
9. $^{16}\text{O}(n,^3\text{He})^{14}\text{C}$	15.52	99.759		

TABLE 2. Summary of the Neutron Environment
for UWMAK-II Solid Materials⁽¹⁴⁾

Function	Material	Distance from First Wall, cm	Neutron Flux, n/cm ² -sec		
			>13.5 Mev	>1 Mev	Total
Curtain	Carbon	-2	9.5 E13	4.4 E14	5.6 E14
Structure	316 Stainless Steel	0-89 (Blanket)	9.5 E13 to 6.8 E10	4.4 E14 to 5.6 E12	5.6 E14 to 7.1 E12
		89-189 (Shield)	6.8 E10 to 5.2 E05	5.6 E12 to 6.4 E07	7.1 E12 to 7.1 E07
		204-302 (Magnet)	$\leq 5.2 \text{ E05}$	$\leq 6.4 \text{ E07}$	$\leq 7.1 \text{ E07}$
Breeder	$\text{Li}_2\text{Al}_2\text{O}_4$	2-4.5	7.1 E13 to 6.0 E13	4.0 E14 to 3.9 E14	5.2 E14 to 4.8 E14
		23-32	1.5 E13 to 8.7 E12	2.1 E14 to 1.5 E14	2.8 E14 to 1.9 E14
Neutron Multiplier	Beryllium	5-22	5.2 E13 to 1.6 E13	3.9 E14 to 2.1 E14	4.8 E14 to 2.8 E14
Reflector	Carbon	32-70	8.7 E12 to 2.3 E11	1.5 E14 to 1.4 E13	1.9 E14 to 4.0 E13
Shielding	Lead, B ₄ C, Stainless Steel	89-189	6.8 E10 to 5.2 E05	6.4 E12 to 7.1 E07	7.1 E13 to 7.1 E07
Magnet Materials	Copper, Niobium-Titanium	204-302	$\sim 5.2 \text{ E05}$	$< 7.1 \text{ E07}$	$< 7.1 \text{ E07}$

MATERIAL 1 IS A MIXTURE OF 90% B₄C + 10% SS AND MATERIAL 11 IS A MIXTURE OF 90% Pb + 10 % SS

FIGURE 1. A Schematic of UMMAK-II Blanket and Shield⁽¹⁴⁾

The ^{14}C produced is calculated from the product of the time, volume, concentration, and cross-section of the reacting nuclide and the neutron flux. The decay term and the ^{14}C transmutation term can be ignored because of the long half-life of ^{14}C , 5,720 y, and the very small cross section (Table 1).

Using these data and the amounts and types of materials in the various regions, the total ^{14}C production is calculated. Where data were lacking, an approximation of 100 wppm was made. The regions are discussed separately below. Impurity level data in the literature were presented in most cases as ppm. We interpreted ppm as wppm as opposed to appm and made our calculations accordingly.

Plasma

The production of ^{14}C in the plasma will be due to air leakage. This has been estimated to be 0.1% of the plasma chamber volume⁽¹³⁾ which at STP will be 3.47×10^2 moles of air. The calculated quantities of ^{14}C produced per day are presented in Table 3. The only significant producer is the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction. Production from ^{13}C and ^{17}O is insignificant in this case, and in any other case when impurity activation is the sole source. The result is due to the small cross-section of ^{13}C and the small isotopic abundance of ^{17}O , respectively.

The ^{14}C produced in this region could react with the oxygen impurity to form CO , but more likely with the residual deuterium or tritium fuel to form hydrocarbons.

Curtain

A carbon curtain has been proposed for use in UWMAK-II to protect the first wall from the energetic alpha particles and to partially attenuate the neutron flux. Carbon-14 production from ^{13}C in the curtain has been calculated to be 6.7×10^{-3} Ci/day.⁽¹⁴⁾ This number is evidently based on the thermal neutron cross-section. Calculations based on the volume of the

TABLE 3. Calculated Carbon-14 Production

<u>Region</u>	<u>Material</u>	<u>Flux,^(a) Neutrons cm² -Sec</u>	<u>Reaction</u>	<u>¹⁴C Produced, Ci/day</u>
Plasma Zone 1 and 2	Air in- leakage	5.6x10 ¹⁴ (b)	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	1.2x10 ⁻¹
		5.6x10 ¹⁴ (b)	¹³ ₆ C(n,γ) ¹⁴ ₆ C	3.5x10 ⁻¹²
		5.6x10 ¹⁴ (b)	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	3.8x10 ⁻⁵
Curtain	Carbon	5.6x10 ¹⁴ (b)	¹³ ₆ C(n,γ) ¹⁴ ₆ C	6.7x10 ⁻³ (14) 7.3x10 ⁻⁴ (c)
	90 appm Nitrogen	5.6x10 ¹⁴ (b)	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	4.9x10 ⁻³ (c)
	75 appm Oxygen	5.6x10 ¹⁴ (b)	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	5.12x10 ⁻⁶
Blanket Zone 2-10	316 Stainless Steel			
	390 appm Nitrogen	2.8x10 ¹⁴	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	2.9
	2780 appm Carbon	2.8x10 ¹⁴	¹³ ₆ C(n,γ) ¹⁴ ₆ C	2.9x10 ⁻⁶
	<700 appm Oxygen	2.8x10 ¹⁴	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	6.2x10 ⁻³
	Niobium - 1% Zirconium			
	1300 appm Nitrogen	2.8x10 ¹⁴	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	9.7
	800 appm Carbon	2.8x10 ¹⁴	¹³ ₆ C(n,γ) ¹⁴ ₆ C	8.3x10 ⁻⁷
	1800 appm Oxygen	2.8x10 ¹⁴	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	1.6x10 ⁻²
Impurity Data for Nimonic PE-16 Unavailable				
Blanket Zone 4	Li ₂ Al ₂ O ₄	5.0x10 ¹⁴	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	7.8x10 ⁻¹
	940 appm ^(d) Nitrogen	5.0x10 ¹⁴	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	1.2
	1100 appm ^(d) Carbon	5.0x10 ¹⁴	¹³ ₆ C(n,γ) ¹⁴ ₆ C	2.1x10 ⁻⁷
Zone 4-6 ⁽¹³⁾ UWMAK-I	Lithium			
	15 appm Nitrogen	2.6x10 ¹⁴ (13)	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	2.7x10 ⁻¹
	250 appm Nitrogen	2.6x10 ¹⁴ (13)	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	4.5

TABLE 3. (contd)

Region	Material	Flux, ^(a) Neutrons cm ⁻² -Sec	Reaction	¹⁴ C Produced, Ci/day
Zone 6 UWMAK-II	40 appm Oxygen	2.6x10 ¹⁴ (13)	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	9.5x10 ⁻⁴
	58 appm Carbon(d)	2.6x10 ¹⁴ (13)	¹³ ₆ C(n,γ) ¹⁴ ₆ C	1.5x10 ⁻⁷
	Li ₂ Al ₂ O ₄	2.4x10 ¹⁴	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	1.3
	940 appm Nitrogen(d)	2.4x10 ¹⁴	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	2.1
	1100 appm Carbon(d)	2.4x10 ¹⁴	¹³ ₆ C(n,γ) ¹⁴ ₆ C	3.4x10 ⁻⁷
Zone 8 ⁽¹³⁾ UWMAK-I	Lithium			
	15 appm Nitrogen	2.0x10 ¹³ (e)	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	2.0x10 ⁻³
	250 appm Nitrogen	2.0x10 ¹³ (e)	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	3.4x10 ⁻²
	40 appm Oxygen	2.0x10 ¹³ (e)	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	7.2x10 ⁻⁶
	58 appm Carbon(d)	2.0x10 ¹³ (e)	¹³ ₆ C(n,γ) ¹⁴ ₆ C	1.2x10 ⁻⁹
Impurity Data for Flibe Unavailable (53.1% BeF ₂ , 46.9% LiF)				
Blanket Zone 5	Beryllium			
	5,400 appm Oxygen	3.8x10 ¹⁴	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	4.3x10 ⁻²
	500 appm Carbon	3.8x10 ¹⁴	¹³ ₆ C(n,γ) ¹⁴ ₆ C	2.3x10 ⁻⁷
	64 appm Nitrogen(d)	3.8x10 ¹⁴	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	4.1x10 ⁻¹
Blanket Zone 7	Graphite Carbon	1.2x10 ¹⁴	¹³ ₆ C(n,γ) ¹⁴ ₆ C	4.9x10 ⁻⁴
	90 appm Nitrogen	1.2x10 ¹⁴	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	3.0x10 ⁻²
	75 appm Oxygen(d)	1.2x10 ¹⁴	¹⁷ ₈ O(n,α) ¹⁴ ₆ C	3.2x10 ⁻⁵
Shield	Lead			
	1500 appm Nitrogen(d)	3.6x10 ¹²	¹⁴ ₇ N(n,p) ¹⁴ ₆ C	2.0x10 ⁻¹

(a) See Text.

(b) Flux in Plasma is assumed same as first wall.

(c) 1.4x10⁻⁶cm³ graphite at 2.25 g/cm³.

(d) Estimated impurity level of 100 wppm, data were unavailable.

(e) Flux calculated using distance from first wall of UWMAK-I and flux of UWMAK-II at that distance.

curtain, a curtain density of 2.25 g/cm^3 for pure graphite, the impurity data presented in Table 4, and a neutron cross-section for 10 keV neutrons give $7.3 \times 10^{-4} \text{ Ci/day}$ from ^{13}C and $4.9 \times 10^{-3} \text{ Ci/day}$ from 90 appm nitrogen. The contribution from ^{17}O will be negligible.

TABLE 4. Impurities in Graphite⁽²³⁾

<u>Element</u>	<u>Concentration, wppm^(a)</u>	<u>appm</u>
Lithium	1	2
Boron	162	180
Nitrogen	100	90
Silicon	17	10
Iron	50	10

(a) ppm has been interpreted as wppm

The ^{14}C produced in the curtain should remain there and thus would be a hazard only when the curtain is replaced or an attempt to reclaim the graphite is made. However, it must be pointed out that no experimental data are available for the behavior of ^{14}C generated in graphite.

Blanket Structural Material

A number of structural materials are being considered for use in fusion reactors including stainless steels, nimonic PE-16, niobium, vanadium, and molybdenum.⁽¹⁰⁾ The first three may be used in a first generation reactor. However, the availability and technology of niobium is such that it may have limited application in early units.

The compositions of stainless steel-316 and niobium-1% zirconium are presented in Tables 5 and 6. We were unable to locate the oxygen, nitrogen, and carbon content of nimonic PE-16 (43% nickel, 39% iron, 18% chromium). Impurity levels appear to be around 100 wppm for all metals of this type, so the substitution of PE-16 for stainless steel should make little difference in the ^{14}C yields.

TABLE 5. Typical Chemical Analysis of Commercial 316 SST⁽¹⁴⁾

Element	wppm	vppm	g-Atom/MT	appm
Aluminum	500	1000	18.5	1,030
Arsenic	300	230	4.0	220
Boron	10	52	0.9	50
Cobalt	500	500	8.5	470
Chromium	180,000	195,000	3,461.5	192,000
Copper	1,000	900	15.7	870
Iron	626,000	626,000	11 200.0	670,000
Manganese	20,000	20,000	364.0	20,200
Molybdenum	20,000	12,000	208.5	11,500
Niobium	500	300	5.4	300
Nickel	140,000	134,000	2,380.0	132,000
Phosphorus	200	400	6.5	360
Sulfur	100	200	3.1	170
Silicon	7,000	14,000	249.2	13,800
Tantalum	200	60	1.1	60
Titanium	100	100	2.1	120
Vanadium	2,000	2,200	39.3	2,180
Carbon	600	2,800	50.0	2,780
Nitrogen	100	400	7.1	390
Oxygen	<200	<700	< 12.5	<700

TABLE 6. Composition of Commercial Nb-1% Zr Alloy^(15,16)

Element	Content,		Element	Content,	
	wppm(a)	appm		wppm(a)	appm
Tungsten	300	150	Oxygen	300	1,800
Molybdenum	100	100	Nitrogen	200	1,300
Tantalum	1,000	500	Hydrogen	20	1,900
Hafnium	100	50	Carbon	100	800
Zirconium	10,000	10,000	Iron	50	80
Niobium	985,000	991,000	Nickel	20	30

(a) ppm interpreted as wppm

Carbon-14 production for these materials in the blanket and in other locations is given in Table 3. Nitrogen is again the only major producer of the carbon radioisotope, 2.9 Ci/day in stainless and 9.7 Ci/day in niobium-1% zirconium.

It would be expected that ^{14}C produced in structural materials would exist as a carbide and be relatively immobile. The possibility of transfer by extraction with molten lithium or molten salts such as "flibe" (lithium, beryllium, fluoride), assuming that contact is possible, can be predicted from consideration of thermodynamic stabilities and solubilities. Some data for compounds of interest are listed in Table 7. The question of carbon transfer (as well as nitrogen, oxygen and other elements) between phases is of extreme importance in corrosion control and has been extensively examined. Beskorovainyi⁽²⁴⁾ has reviewed the carbon-molten lithium-metal system and the general compatibility problem is discussed by Smith and Natesan.⁽²⁵⁾ Over the temperature range of interest the stability of the nitride increases in the order lithium, molybdenum, chromium, vanadium, niobium, yttrium, titanium, zirconium and for the carbide, lithium, yttrium, molybdenum, chromium, vanadium, titanium, zirconium and niobium. Nitrogen transfer before activation may be minimal, and it would appear ^{14}C (and nitrogen) that finds its way into a lithium stream could be scavenged by zirconium or yttrium. It should be reemphasized that the data of Table 7 indicate stabilities of the pure compounds. For the practical case, solubility and diffusion data as well as chemical rate data are extremely important.

Tritium Breeding Material

All of the conceptual reactors use lithium to breed tritium. Molten lithium, $\text{Li}_2\text{Al}_2\text{O}_4$, and molten "flibe" (a eutectic mixture of LiF , BeF_2) are the prime candidates at this time. The impurity data for lithium are found in Table 8. Impurity data for $\text{Li}_2\text{Al}_2\text{O}_4$ and "flibe" are unavailable so calculations were performed using 100 wppm each for nitrogen, oxygen and carbon impurity levels.

TABLE 7. Free Energies of Formation of Oxides, Nitrides, Carbides, and Fluorides^(a) (ΔG° , Kcal/gm atom, oxygen, nitrogen, carbon, and iron)

	Reference	500°K	1000°K	1500°K
Al_2O_3	26	-120.9	-108.4	-95
Li_2O	25	-128.0	-111.6	-95.2
ZrO_2	25	-119.2	-108.0	-87.8
TiO	25	-111.6	-101.0	-90.4
BeO	26	-131.4	-119.8	(-109.2)
V_2O_5	25	-103.4	- 79.7	- 56
Ta_2O_5	26	- 87.0	- 76.7	- 67.1
FeO	26	- 55.3	- 47.4	- 39.1
Mn_2O_3	26	- 64.0	- 53.8	- 43.6
NbO	25	- 89.6	- 78.9	- 68.2
Cr_2O_3	25	- 80.0	- 69.7	- 59.4
Y_2O_3	26	-139.5	-127.3	-115.5
NiO	25	- 46.1	- 35.7	- 25.6
CO_2	27	- 47.2	- 47.3	- 47.4
H_2O	27	- 52.4	- 46.0	- 39.3
TiC	25	- 42.6	- 41.5	- 40.3
ZrC	25	- 43.0	- 41.9	- 40.8
V_2C	25	- 34.6	- 34.1	- 33.6
Nb_2C	25	- 45.5	- 45.0	- 44.5
TaC	26	- 37.8	- 36.4	- 34.3
Cr_7C_3	28	- 19	- 20	(- 22)
$Cr_{23}C_6$	28	- 23	- 26	- 28
YC_2	28	- 12.9	- 14.0	- 15.3
GdC_2	28	- 14.9	- 15.5	- 16.8
Mn_7C_3	28	0	- 6	- 12
Fe_3C	26	+ 3.8	+ 0.8	- 0.65
Ni_3C	28	+ 7	+ 5	+ 4
Al_4C_3	28	-12.0	-12.3	-11.5
Mo_2C	26	+ 2.9	-10.3	-23.4
Li_2C_2	26	- 4.25	- 0.70	+ 2.85

TABLE 7. (contd)

	Reference	500°K	1000°K	1500°K
Be ₂ C	27	-20.7	-19.0	-17.2
YN	28	-61.0	-50.9	-40.3
Nb ₂ N	25	-54.0	-44.9	-31.8
TiN	25	-69.4	-58.3	-47.3
ZrN	25	-75.9	-64.8	-53.7
AlN	26	-51.0	(-40)	(-25)
Be ₃ N ₂	26	-56.6	(-47.2)	(-38.9)
Li ₃ N	25	-28.0	- 10.0	+ 10.2
Cr ₂ N	25	-18.3	- 12.4	- 6.4
Mn ₅ N ₂	26	-15.1	(- 5)	--
Fe ₄ N	26	+3.1	+ 9.5	+ 14
VN	25	-30.4	-20.4	- 10.4
Mo ₂ N	25	- 8.9	- 1.3	+ 6.3
TaN	26	-47.9	-40.5	- 32.3
BeF ₂	27	-113.3	-104.3	-96.5 (liq)
AlF ₃	27	-109.8	-99.6	-89.3
ZrF ₄	27	-104.1	-94.6	-87.2 @ 1400°K
YF ₃	29	-123	-115	-107
MnF ₂	26	(- 86.5)	(- 78.5)	(- 73.2)
TiF ₄	27	- 89.6	- 81.6	- 74.1
NiF ₂	26	(- 70)	(- 62)	(-54.2)
CrF ₂	29	- 83	- 75	- 67
LiF	27	-136.1	-123.7	-115.5
TaF ₂	29	(- 82)	(- 75)	(- 67)
NbF ₅	29	(- 62)	(- 59)	(- 55)
VF ₂	29	(- 90)	(- 83)	(- 75)
MoF ₆	27	- 55.8	- 49.7	---
MoF ₅	29	(- 60)	(- 58)	(- 54)
FeF ₂	27	- 75.9	- 67.9	- 59.9

(a) Data are for the normal state at the temperature indicated.

TABLE 8. Typical Impurities Found in High-Purity Lithium^(15,16)

Element	Content,		Element	Content,	
	wppm(a)	appm		wppm(a)	appm
Silicon	5-50	1-12	Nickel	100	10
Oxygen	100	40	Chromium	100	10
Nitrogen	30-500	15-250	Tantalum	100	4
Calcium	100	20	Fluorine ^(b)	3,000	1,100
Sodium	100	30	Chlorine ^(b)	3,000	600
Potassium	100	20			
Iron	100	10			

(a) ppm interpreted as wppm

(b) Fluorine and chlorine content arises from proposed use of LiF-LiCl eutectic to extract tritium from blanket of RTPR.

Carbon-14 production from the oxygen constituent in $\text{Li}_2\text{Al}_2\text{O}_4$ amounts to 0.78 Ci/day in Zone 4 (Figure 1) and 1.3 Ci/day in Zone 6 (Figure 1). Based on 940 appm nitrogen in the $\text{Li}_2\text{Al}_2\text{O}_4$, 1.2 and 2.1 Ci/day will be produced in Zones 4 and 6, respectively.

For the lithium coolant-breeder in Zones 4 through 6 (Figure 2) of UWMAK-I the computed amount ranges from 0.27 to 4.5 Ci/day for the minimum and maximum nitrogen impurity levels. For Zone 8, an estimated flux for the same location in UWMAK-II was used, values ranging from 2.0×10^{-3} to 3.4×10^{-2} Ci/day are obtained. No calculations were performed for "flibe". Again, the ^{14}C production will depend on the nitrogen concentration of the material. Table 3 presents the calculated data for breeder materials.

FIGURE 2. University of Wisconsin CTR Blanket, Shield and Magnet Structure for 5000 MWth System, UWMK-1(13)

The fate of the ^{14}C produced in the breeder material is again dependent on the physical state, and the thermodynamics of the system (Table 7). The chemical form of the carbon produced in the $\text{Li}_2\text{Al}_2\text{O}_4$ is unknown; potentially, the most stable compound is Al_2C_3 , but it may never form under the conditions existing. In the $\text{Li}_2\text{Al}_2\text{O}_4$ case the ^{14}C problem may emerge during blanket processing. The molten lithium or "flibe" systems may lose the carbon to structural materials as it is circulated or the lithium may lose it to the proposed purification system, a zirconium or yttrium getter.⁽¹³⁾ In either of the latter cases, ^{14}C may only be a problem when the material is removed and reclaimed.

Beryllium

Beryllium is to be used as a neutron multiplier in the blanket to increase the tritium breeding. UWMAC-II has a zone of solid beryllium metal; Table 9 presents the composition of commercial-grade beryllium. Since no nitrogen content was located in the beryllium, calculations are based on 60 appm nitrogen.

TABLE 9. Chemical Composition Limits for Commercial Grade Beryllium QMV(30)

Element	wppm	appm
Oxygen	4,500 - 9,600	2,500 - 5,400
Iron	800 - 1700	100 - 300
Aluminum	300 - 600	100 - 200
Magnesium	100 - 500	40 - 200
Nickel	200 - 800	30 - 100
Chromium	100 - 300	20 - 500
Silicon	500 - 800	200 - 300
Boron	2	2
Carbon	700	500

The amount of ^{14}C produced is presented in Table 3. From 60 appm nitrogen 0.41 Ci/day are produced. This number may be low as impurities in beryllium appear to be higher than the normal impurity levels in other metals. The oxygen level is high enough to contribute 0.04 Ci/day. The ^{14}C produced should remain fixed in the metal in its lifetime in the reactor. Its fate outside of the reactor depends on the treatment of the spent beryllium.

Coolant

If the blanket material is not used as the coolant, an alternative is helium. The possibilities for ^{14}C production in this gas are greatly minimized by the high degree of purity, 2 appb, attainable.⁽³¹⁾ It is a relatively simple process to remove nitrogen from helium. However, the helium coolant specified for a typical HTGR reactor shows 0.7 appm nitrogen.⁽³²⁾

Moderator-Reflector

Graphite is typically chosen as the moderator-reflector material; however, stainless steel was chosen for UWMak-I. Impurity content is presented in Table 4.

In UWMak-II, 4.9×10^{-8} and 2.99×10^{-2} Ci/day (Table 3) of ^{14}C will be produced from carbon and 90 appm nitrogen in the graphite. As was noted earlier, unless a large quantity is present, its contribution will be negligible.

Shield

The materials which are to be used in the radiation shields are lead and boron carbide. No impurity data were located for either. The flux in this region is much lower, $10^{12} \frac{\text{neutrons}}{\text{cm}^2\text{-sec}}$, and the production rate would be quite small. For lead containing 1,500 appm nitrogen, 1.3×10^{-2} Ci/day would be produced in UWMak-II (Table 3). Over long periods of time ^{14}C levels in the shield materials could become significant.

CARBON-14 REMOVAL AND FIXATION

As mentioned above, ^{14}C produced in graphite, in metals, and in other solids is probably not very mobile and may largely remain in place. Some will migrate to interfaces and be transported by liquid or gas phases to regions of higher stability (see Table 7). No information is available on the chemical forms that may exist; presumably they would include carbon, metal carbides, CO, CO_2 and hydrocarbons. Reducing conditions exist in nearly all the cases examined, hence CO and CO_2 production should be small. If hydrogen is present, the tendency in the high radiation field will be toward the production of simple saturated hydrocarbons. It is interesting to note that the ^{14}C in cover gas from a boiling water reactor ($\text{H}_2\text{-N}_2$ atmosphere) was over 80% CH_4 and C_2H_6 .⁽³³⁾

Removal methods may be unique for each reactor component, e.g., cryogenic trapping for plasma ash, and perhaps catalytic oxidation to CO_2 . For helium purification, molecular sieve or chemical trapping may be used. Some of the metal carbides are quite stable and may fix the ^{14}C until some processing step is performed. However, it should be noted that they are not in general thermodynamically stable with respect to reactions such as $\text{MC} + \text{O}_2 \rightarrow \text{MO} + \text{CO}_2$ and $\text{MO} + \text{H}_2\text{O} \rightarrow \text{MO} + \text{CH}_4$ and even (in some cases) $\text{MC} + \text{H}_2 \rightarrow \text{MH} + \text{CH}_4$.

If environmental release of ^{14}C is not permitted, conversion to a solid such as calcium or barium carbonate has been proposed as an alternative. The carbonate would be transferred to a repository for storage.⁽³⁴⁾

CONCLUSIONS AND RECOMMENDATIONS

Our calculations for ^{14}C production from ^{14}N , ^{17}O , and ^{13}C neutron reactions in a 5,000 MWth toroidal fusion reactor indicate that ~ 8 Ci/day, principally from ^{14}N if stainless steel is used as the structural material; the use of Nb-1% Zr will double this quantity. This can be compared to the natural global production rate from cosmic rays of 6 kg/y or 78 Ci/day.⁽⁸⁾ Of the ^{14}C produced, only 0.12 Ci/day produced by activation of air in-leakage into the plasma may be of immediate environmental concern. The remainder will be fixed in the structural or other reactor materials until this material is reclaimed, i.e., until sufficient time has passed for depletion of the other activated elements.

The ^{14}C produced by activation of air which has leaked into the plasma chamber would be removed in the ash treatment and could subsequently leak or be released to the environment. Further consideration of the behavior of ^{14}C in the plasma is indicated. In general, the chemical form of the ^{14}C produced is not known. Reducing conditions generally prevail leading to carbides and perhaps hydrocarbons. Experimental studies should be made to identify species and determine ease of removal and treatment. It should be noted that a wide range of materials is involved: gases, non-metals, liquid and solid metals, and liquid and solid salts. The chemistry of carbon in each is quite different.

The impurity level in materials is very important, especially that of nitrogen. Missing data should be found and methods of minimizing the nitrogen content should be examined as should the migration of nitrogen between components. Methods of conversion of ^{14}C forms to CO_2 or other form for fixation should also be evaluated along with methods of fixing the resulting compound for storage. Finally, the production data listed are only estimates and could be refined by using multigroup calculations along with better nitrogen impurity data.

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