

Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents

2010 TECHNICAL REPORT

Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents

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REPORT SUMMARY

Nuclear power plants report the amount of radioactivity released through permitted effluent pathways in their plant annual reports. This report provides users with a method for calculating the amount of carbon-14 (¹⁴C) generated in a light water reactor (LWR) core and released through plant gaseous effluent pathways.

Background

Improvements in nuclear power plant effluent management practices have resulted in a decrease in the concentration and a change in the distribution of gaseous radionuclides released to the environment. The latest revision of Regulatory Guide 1.21 defines a "principal nuclide" as any radionuclide whose concentration exceeds 1% of the total release, stating that the released quantity must be included in the annual radioactivity discharge report. Regulatory Guide 1.21 indicates that the ¹⁴C discharge can be estimated by sample measurements or by use of a normalized ¹⁴C source term and scaling factors based on power generation. However, the normalized source term and scaling factors were developed several decades ago, and updated research and experience exists to explore more precise methods of ¹⁴C source term and release estimation.

Objective

To present a method for calculating the amount of ¹⁴C generated in pressurized water reactor (PWR) and boiling water reactor (BWR) cores and released through plant gaseous effluent pathways.

Approach

This report reviews ¹⁴C measurements that have been made for the most part in the 1970s and 1980s, but more recently in Europe, Korea, and the United States. The primary emphasis is to provide a method for estimating ¹⁴C source terms for BWRs and PWRs based on "effective" neutron cross sections, core coolant mass, and a two- or three-energy group core neutron flux distribution. Transport of ¹⁴C in both types of reactors is discussed, and ¹⁴C generation rates for each type of reactor are calculated based on plant-specific parameters. A brief summary of ¹⁴C gaseous and liquid sampling and analysis techniques is included.

Results

The following general conclusions were developed during preparation of this report:

- A significant database on ¹⁴C generation and its transport at PWRs and BWRs exists.
- The principal production reaction leading to the release of 14 C during plant operation is the $^{17}O(n,\alpha)$ 14 C nuclear reaction in LWR coolant.

- The production of ¹⁴C from the ¹⁴N(n,p) ¹⁴C reaction also contributes to the PWR ¹⁴C source term. Ammonia is formed due to hydrazine injection for oxygen reduction during startups. Nitrogen also enters the reactor coolant system as dissolved nitrogen during boron dilution, particularly in mid-to-latter phases of the fuel cycle.
- Most of the ¹⁴C produced in a BWR is released in a gaseous form by the off-gas system, primarily in the form of ¹⁴CO₂.
- Gaseous release of ¹⁴C from the PWR (without a recombiner in the gas treatment system) will be mainly in the form of low molecular weight hydrocarbons. The remainder will be inorganic, primarily ¹⁴CO₂.
- A method was developed to allow PWR and BWR personnel to calculate a site-specific ¹⁴C source term, based on knowledge of the neutron flux distributions and coolant mass in the "active" core during the fuel cycle. Examples of this calculation method are provided.
- The technology for ¹⁴C gaseous effluent sampling and analysis is well developed and in routine use at several international utilities. Recently, a number of exploratory measurements have been made at U.S. PWRs.

EPRI Perspective

EPRI conducts research and development on nuclear power plant effluents to support industry best practices in minimizing and managing the impact of permitted radioactive releases to the community and the environment. As nuclear power plants continue to implement best practices to reduce the total radioactivity in plant effluents, other radionuclides that were not previously significant fractions of the effluent streams will need to be quantified and reported. Additionally, as stakeholders become increasingly concerned about environmental protection, more in-depth and precise knowledge of the potential impacts of nuclear power plant operations on the environment will be necessary. EPRI conducts research and development activities to provide the industry with best practices for accurately estimating the source term, transport, and release of ¹⁴C and other radionuclides from nuclear power plants. These research and development efforts will support the nuclear power industry in effectively communicating with stakeholders about nuclear power plant effluents.

Keywords

Carbon-14
Gaseous Effluents
Carbon-14 Production
Carbon-14 Transport
Carbon-14 Measurement
Carbon-14 Production Cross Sections

CONVERSION FACTORS

To Convert From	To	Multiply By
μCi	Bq	3.7E4
•	kBq	37
	MBq	3.7E-2
	GBq	3.7E-5
Ci	Bq	3.7E10
	kBq	3.7E7
	MBq	3.7E4
	GBq	37
	TBq	3.7E-2
μCi/sec	Ci/yr	31.56
	GBq/yr	1167.6
G:/ ·	G: /	0.7260
μCi/min	Ci/yr	0.5260
	GBq/yr	19.46
μCi/MW _{th} -sec	Ci/MW _{th} -yr	31.56
	GBq/MW _{th} -	1167.6
	yr kBq/MW _{th} -h	1.332E5
GBq/GW _e -yr	Ci/GW _e -yr	2.703E-2
Ci/GW _e -yr	GBq/GW _e -yr	37
Ci/G We yi	Ci/GW _{th} -yr	37 0.34 ^(a)
	22/ 3 (m j1	0.5 1
rad	gray (Gy)	1.0E-2
mrad	mGy	1.0E-2
rem	Sievert (Sv)	1.0E-2
mrem	mSv	1.0E-2

⁽a) At a thermal efficiency of 34%.

CONTENTS

1 INTRODUCTION	1-1
2 BACKGROUND	2-1
2.1 Sources of ¹⁴ C in the Environment	2-1
2.1.1 ¹⁴ C Production Reactions	2-1
2.1.2 Environmental Sources of ¹⁴ C	2-4
2.1.3 Chemical Forms of ¹⁴ C Produced in the LWR Primary Coolant	2-5
2.2 Measurement and Reporting Requirements at Nuclear Plants	2-6
2.2.1 Gaseous and Liquid Effluents	2-6
2.2.2 Regulatory Guide 1.21	2-7
2.2.3 Solid Wastes (10 CFR Part 61)	2-8
3 CARBON-14 GENERATION AND RELEASE IN BWR SYSTEMS	3-1
3.1 Overview of ¹⁴ C in the BWR	3-1
3.2 ¹⁴ C Source Term Estimations and Measurements	3-1
3.3 Site Specific BWR Source Term Estimation	3-3
3.3.1 STEP 1: Unit Specific Neutron Flux	3-4
3.3.2 STEP 2: Effective Cross-Sections and ¹⁴ C Generation Rates	3-5
3.3.2.1 Production Rate from ¹⁷ O(n,α) ¹⁴ C Reaction	3-6
3.3.2.2 Production Rate of ¹⁴ C from ¹⁴ N(n,p) ¹⁴ C Reaction	3-7
3.3.3 STEP 3: Unit Specific Coolant Mass	3-8
3.3.4 STEP 4: ¹⁴ C Source Term	3-8
3.3.4.1 ¹⁴ C Source Term from ¹⁷ O(n,α) ¹⁴ C Reaction	3-8
3.3.4.2 ¹⁴ C Source Term from ¹⁴ N(n,p) ¹⁴ C Reaction	3-8
3.3.5 Summary of ¹⁴ C Source Term Calculations	3-9
3.4 Release Pathways and Chemical Form	3-9
3.4.1 Nine Mile Point Unit-1	3-12
3.4.2 Ovster Creek	3-12

3.4.3 Fowler's Summary	3-14
3.4.4 Brunswick Units 1 and 2	3-14
3.4.5 J. A. FitzPatrick BWR	3-18
3.4.6 Nordic BWRs	3-19
3.4.7 ¹⁴ CO ₂ Adsorption on Charcoal	3-21
3.5 Effect of Chemistry on Gaseous Release Rate and Chemical Form	3-23
3.6 Effect of Fuel Failures	3-24
3.7 Summary of BWR Transport and Release	3-24
4 CARBON-14 GENERATION AND RELEASE IN PWR SYSTEMS	
4.1 Overview of ¹⁴ C in the PWR	
4.2 ¹⁴ C Source Term Estimation and Measurement	
4.3 Site Specific PWR Source Term Estimation	
4.3.1 STEP 1: Unit Specific Neutron Flux	4-6
4.3.2 STEP 2: Effective Cross-Sections and ¹⁴ C Generation Rate	4-7
4.3.2.1 Production Rate of ¹⁴ C from ¹⁷ O(n,α) ¹⁴ C Reaction	
4.3.2.2 Production Rate of ¹⁴ C from ¹⁴ N(n,p) ¹⁴ C Reaction:	
4.3.3 STEP 3: Unit Specific Coolant Mass	
4.3.4 STEP 4: ¹⁴ C Source Term	
4.3.4.1 ¹⁴ C Source Term from 17O(n,α) ¹⁴ C Reaction	
4.3.4.2 ¹⁴ C Source Term from ¹⁴ N(n,p) ¹⁴ C Reaction	
4.3.5 Summary of ¹⁴ C Source Term Calculations	
4.4 ¹⁴ C Release Pathways and Chemical Form	
4.4.1 Ginna	
4.4.2 Indian Point Unit 3	
4.4.3 Palisades and Big Rock Point	
4.4.4 Vance Studies	
4.4.5 Nordic PWRs	
4.4.6 Diablo Canyon Units 1 and 2	
4.4.7 V. C. Summer	
4.4.8 Korean PWRs	
4.5 Chemistry Effects	
4.6 Effects of Fuel Failures	
4.7 Reactor Make-up Water	
4.8 Summary of PWR ¹⁴ C Transport and Release	4-25

5 MEASUREMENT IN THE NUCLEAR POWER INDUSTRY	5-1
5.1 General	5-1
5.2 Study Methodologies	5-2
5.3 Current Methods	5-5
5.4 Commercial Sampling Systems	5-6
6 REFERENCES	6-1
A BWR ¹⁴ C SOURCE TERM CALCULATION	A-1
1.0 Cross-Section Estimation	A-1
2.0 ¹⁷ O(n,α) ¹⁴ C Reaction	A-1
2.1 Source Term Calculation	A-7
2.1.1 Moderator Region	A-7
2.1.2 Bypass Region	A-7
2.1.3 Core Production Rate	A-7
2.1.4 Site Specific Calculations	A-7
3.0 ¹⁴ N(n,p) ¹⁴ C Reaction	A-8
3.1 Source Term Calculation	A-8
3.1.1 Moderator Region	A-10
3.1.2 Bypass Region	A-10
3.1.3 Core Production Rate	A-10
4.0 Supplemental BWR Neutron Flux Data	A-10
B SITE SPECIFIC BWR SOURCE TERM CALCULATION	B-1
1.0 BWR	B-1
C PWR ¹⁴ C SOURCE TERM CALCULATION	C-1
1.0 Core Flux Energy Distribution	C-1
2.0 Effective Cross Sections	C-3
2.1 ¹⁷ O(n,α) ¹⁴ C Reaction	C-3
2.2 ¹⁴ N(n,p) ¹⁴ C Reaction:	C-5
3.0 Supplemental Neutron Flux Data	C-7
D PWR SITE SPECIFIC SOURCE TERM CALCULATION	D-1
1.0 Westinghouse – A (W-A)	D-2
2.0 Westinghouse-B (W-B)	D-4

3.0 C	ombustion Engineering-A (CE-A)	D-7
4.0 W	/estinghouse-C (W-C)	D-11
5.0 W	/estinghouse-D (W-D) and Westinghouse-E (W-E)	D-16
5.	1 Westinghouse-D (W-D)	D-17
5.2	2 Westinghouse-E (W-E)	D-19
6.0 W	/estinghouse-F (W-F)	D-22
7.0 C	ombustion Engineering-B (CE-B) Calculation	D-25
8.0 W	/estinghouse-G (W-G) Calculation	D-29
<i>E</i> PWR I	PRIMARY WATER NITROGEN CONCENTRATION	E-1
1.0 G	eneral Considerations	E-1
2.0 S	ample Calculation	E-1
3.0 E	ffects of VCT Operating Practices	E-2
Refer	rence	E-2

LIST OF FIGURES

Figure 2-1 Reaction Cross-Sections (ENDF)	2-2
Figure 3-1 ¹⁴ C Transport in the BWR	3-26
Figure 4-1 Carbon-14 Mass Balance for PWR	4-2
Figure 4-2 Ringhals Unit-3 Main Stack ¹⁴ C Release During the Years 2002-2006 (Bengtsson, 2010)	4-17
Figure 4-3 Ringhals Unit-4 Main Stack ¹⁴ C Release During the Years 2002-2006 (Bengtsson, 2008)	4-18
Figure 4-4 ¹⁴ C Transport in the PWR	
Figure A-1 ¹⁷ O(n,α) ¹⁴ C Reaction Cross-Section, Reference BWR Neutron Flux Distribution at 50 GWd/MT and ¹⁴ C Production	A-2
Figure A-2 BWR Moderator Neutron Flux vs. Fuel Exposure	A-4
Figure A-3 BWR Bypass Neutron Flux vs. Fuel Exposure	A-5
Figure A-4 ¹⁴ N(n,p) ¹⁴ C Reaction Cross-Section, Reference BWR Neutron Flux Distribution at 50 GWd/MT and Calculated ¹⁴ C Production Rate	A-9
Figure A-5 Radial Three Group Neutron Flux Distribution at Core Axial Mid-Plane, ANISN Calculation for General Electric BWR (Evans, et al.,1984)	A-12
Figure C-1 Sample of the 70 Group Neutron Spectrum at Various Burnups – 4.20 w/o Fuel (Secker (2009))	C-2
Figure C-2 ¹⁷ O(n,α) ¹⁴ C Reaction Cross-Section, Reference PWR Neutron Flux Distribution at 16 GWd/MT and Calculated ¹⁴ C Production	C-4
Figure C-3 ¹⁴ N(n,p) ¹⁴ C Reaction Cross-Section, Reference PWR Neutron Flux Distribution at 16 GWd/MT and Calculated ¹⁴ C Production	C-6
Figure C-4 Radial Three Group Neutron Flux Distribution at Core Axial Mid-Plane, ANISN Calculation for Westinghouse PWR (Evans, et al., 1984)	C-8

LIST OF TABLES

Table 2-1 Carbon-14 Production Reactions in PWRs and BWRs	2-1
Table 2-2 Reaction Cross-Sections (JEF Report 14, 1994)	2-3
Table 2-3 Basis for Reaction Cross Section Calculations	2-3
Table 2-4 Amount of Carbon in Various Exchangeable Reservoirs (Suess, 1958)	2-4
Table 2-5 Possible Chemical Forms Produced from ¹⁴ C In-Core Reactions	2-6
Table 3-1 Calculated Production Rates of Carbon-14 in Light Water Reactors (Fowler, 1976)	3-2
Table 3-2 Calculated Production Rates of ¹⁴ C in the Reactor Coolant of a 2500 MW _{th} ASEA-ATOM BWR (Magnusson, et al. 2008)	3-3
Table 3-3 Calculated Production Rates of Carbon-14 in BWR Reactor Coolant (Vance, 1995)	3-3
Table 3-4 Example of Flux Variations over BWR Fuel Cycle ^a	3-5
Table 3-5 "Effective Cross-Sections" for the ¹⁷ O(n,α) ¹⁴ C Reaction in the BWR	3-6
Table 3-6 "Effective Cross-Sections" for the ¹⁴ N(n,p) ¹⁴ C Reaction in the BWR	3-6
Table 3-7 Yields of ¹¹ C Species in Triple Distilled, Degassed Water Irradiated with 185 MeV Protons, in Per Cent of the Total Yield of ¹¹ C	3-11
Table 3-8 Oyster Creek BWR Carbon-14 Measurements (Blanchard, et al.,1976)	3-13
Table 3-9 Concentration of ¹⁴ C Released from the Oyster Creek Main Condenser Steam Jet Air Ejectors ^a	3-13
Table 3-10 Concentration of ¹⁴ C in Reactor Water at Oyster Creek	3-13
Table 3-11 BWR Carbon-14 Source Terms ^a (Nominal 1250 MW _a at 80% Capacity Factor)	3-14
Table 3-12 ¹⁴ C Measurements at Brunswick Units 1 and 2 (NUREG/CR-4245, 1985)	3-16
Table 3-13 Estimation of Brunswick Main Stack Release, µCi/MW _{th} -h	3-18
Table 3-14 Carbon-14 Gaseous Release Rate, Chemical Form and Discharge Pathways for the J. A. FitzPatrick BWR (Kunz, 1985)	3-19
Table 3-15 ¹⁴ C Production in BWR Reactor Fuel and Internals (based on FSAR for F3/O3) ^a	3-20
Table 3-16 ¹⁴ C Production and Release Pathways for Three ASEA-ATOM BWRs	3-21
Table 3-17 Concentration of ¹⁴ C in Off-Gas System at Oskarshamn Unit 3 (NWC) (Magnusson and Stenström (2005))	3-21
Table 3-18 Transport of ¹⁴ C Through the Off-Gas System at Oskarshamn Unit 3 (NWC) (Magnusson and Stenström (2005))	3-22
Table 3-19 Summary of Calculated ¹⁴ C Generation Bates at BWBs ⁸	3-24

Table 3-20 Summary of Observed Gaseous Release Rates at BWRs	3-25
Table 4-1 Carbon-14 Release Data from Operating PWR's (NUREG-0017, Rev. 1)	4-3
Table 4-2 Distribution of Carbon-14 Released in Gaseous Effluents (NUREG-0017, Rev. 1)	4-4
Table 4-3 Calculated Production of Carbon-14 in PWRs (Fowler, et al., (1976))	
Table 4-4 Calculated Production Rates of Carbon-14 in PWR Reactors (Vance, 1995)	
Table 4-5 Calculated Production Rates of ¹⁴ C in the Reactor Coolant of a 2775 MW _{th} Westinghouse PWR (Magnusson (2008))	4-5
Table 4-6 Example of Core Average Neutron Flux over PWR Fuel Cycle	4-7
Table 4-7 "Effective" Cross-Section for the ¹⁷ O(n,α) ¹⁴ C Reaction in the PWR	4-8
Table 4-8 PWR ¹⁴ C Production Rate from the ¹⁷ O(n,α) ¹⁴ C Reaction for Example Plant	
Table 4-9 "Effective Cross-Sections" for the ¹⁴ N(n,p) ¹⁴ C Reaction in the PWR	4-9
Table 4-10 PWR ¹⁴ C Production Rate from the ¹⁴ N(n,p) ¹⁴ C Reaction for Example Plant	4-9
Table 4-11 Percentage of the Total Gaseous ¹⁴ C Activity Detected in Various Compounds	4-11
Table 4-12 PWR ¹⁴ C Source Terms (Fowler, et al. (1976) (Westinghouse Design, Nominal 1250 MW _a at 80% Capacity Factor)	4-12
Table 4-13 ¹⁴ C Gaseous Release Rate, Chemical Form and Discharge Pathways at Ginna and Indian Point 3 (Kunz, 1985)	4-12
Table 4-14 Percentage of Various Compounds in ¹⁴ C Activity Detected in Release Pathways at Indian Point Unit-3	4-13
Table 4-15 Chemical Speciation of ¹⁴ C in Reactor Primary Coolant Water Samples (Vance, et al., 1995)	4-14
Table 4-16 Ringhals Unit 4 Process Water ¹⁴ C Analysis, 2005	4-15
Table 4-17 Carbon-14 Source Terms and Release Pathways at Ringhals Units 3 and 4 (Magnusson, et al. (2008))	4-16
Table 4-18 Diablo Canyon ¹⁴ C Gaseous Sampling, April 2010	
Table 4-19 Diablo Canyon Gaseous ¹⁴ C Annual Discharge Per Operating Unit During Operation	
Table 4-20 Diablo Canyon ¹⁴ C Liquid Sampling April 2010 ^a	
Table 4-21 V. C. Summer ¹⁴ C Sampling Results	
Table 4-22 Gaseous Effluent Results from Five Korean PWRs (Lee (2010))	
Table 4-23 Normalized Gaseous Effluent Release from Some KHNP Plants (D. Lee (2010))	
Table 4-24 Reactor Coolant and Spent Fuel Pool Analyses at Five Korean PWRs (D. Lee (2010))	
Table 4-25 Summary of Calculated ¹⁴ C Generation Rates in Coolant at PWRs ^a	
Table 4-26 Summary of Observed Gaseous Release Rates at PWRs	
Table A-1 Equations Utilized to Fit ¹⁷ O(n,α) ¹⁴ C Cross-Section Data	
Table A-2 "Effective Cross-Sections" for the $^{17}O(n,\alpha)^{14}C$ Reaction in the BWR	A-3
Table A-3 Summary of ¹⁷ O(n,α) ¹⁴ C Production Reaction in the BWR	A-6

Table	A-4 Reference BWR Neutron Flux (50 GWd/MT Exposure)	. A-8
Table	A-5 "Effective Cross-Sections" for the ¹⁴ N(n,p) ¹⁴ C Reaction in the BWR	. A-8
Table	A-6 Core Average Neutron Flux in Various BWRs (Lin, 1980)	A-10
Table	A-7 Approximate BWR Neutron Flux at Core Mid-Plane from Figure A-5	A-11
Table	B-1 BWR Power Station Core Flux and Reactor Coolant Mass Data	. B-1
Table	B-2 BWR Power Station Calculated ¹⁴ C Source Term	. B-2
Table	C-1 Plant Parameters	. C-1
Table	C-2 PWR Flux Distribution	. C-2
Table	C-3 $^{17}O(n,\alpha)^{14}C$ "Effective Cross-Section" Estimation in the PWR	. C-5
Table	C-4 ¹⁴ N(n,p) ¹⁴ C "Effective Cross-Section" Estimation in the PWR	. C-7
	C-5 Typical Westinghouse PWR Neutron Flux Values (Table 4-2, EPRI-009951(2004))	. C-7
	C-6 Radial Three Group Neutron Flux Distribution at Core Axial Midplane, ANISN calculation for a Typical Westinghouse PWR (see Figure C-4) (Evans, et al.(1984))	. C-7
Table	D-1 W-A Cycle 20 BOC and EOC Core Flux	. D-2
Table	D-2 W-A Average Production Rates for the ¹⁷ O(n,α) ¹⁴ C Reaction	. D-3
Table	D-3 W-A Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	. D-4
Table	D-4 W-B BOC and EOC Core Flux	. D-5
Table	D-5 W-B Average Production Rates for the ¹⁷ O(n,α) ¹⁴ C Reaction	. D-6
Table	D-6 W-B Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	. D-7
Table	D-7 CE-A System Parameters	. D-8
Table	D-8 CE-A Cycle 20 Core Flux	. D-8
Table	D-9 "Effective" Cross-Section for the $^{17}O(n,\alpha)^{14}C$ Reaction in the PWR	. D-9
Table	D-10 CE-A Cycle 20 Average Production Rates for the $^{17}O(n,\alpha)^{14}C$ Reaction	D-10
Table	D-11 "Effective" Cross-Section for the ¹⁴ N(n,p) ¹⁴ C Reaction in the PWR	D-10
Table	D-12 CE-A Cycle 20 Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	D-11
Table	D-13 W-C System Parameters	D-12
Table	D-14 W-C Cycle 14 Core Flux	D-12
Table	D-15 "Effective" Cross-Section for the $^{17}O(n,\alpha)^{14}C$ Reaction in the PWR	D-13
Table	D-16 W-C Cycle 14 Average Production Rates for the $^{17}O(n,\alpha)^{14}C$ Reaction	D-14
Table	D-17 "Effective" Cross-Section for the ¹⁴ N(n,p) ¹⁴ C Reaction in the PWR	D-14
Table	D-18 W-C Cycle 14 Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	D-15
Table	D-19 W-D Core Flux Distribution	D-16
Table	D-20 W-E Core Flux Distribution	D-16
Table	D-21 Effective Cross Sections	D-17
Table	D-22 W-D Average Production Rates for the ¹⁷ O(n,α) ¹⁴ C Reaction	D-18
Table	D-23 W-D Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	D-19
Table	D-24 W-E Average Production Rates for the ¹⁷ O(n,α) ¹⁴ C Reaction	D-20
Table	D-25 W-E Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	D-21

Table	D-26 W-F Core Flux Distribution	D-22
Table	D-27 "Effective" Cross-Section for the ¹⁷ O(n,α) ¹⁴ C Reaction in the PWR	D-22
Table	D-28 W-F Average Production Rates for the ¹⁷ O(n,α) ¹⁴ C Reaction	D-23
Table	D-29 "Effective" Cross-Section for the ¹⁴ N(n,p) ¹⁴ C Reaction in the PWR	D-24
Table	D-30 W-F Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	D-25
Table	D-31 CE-B Core Average Neutron Flux	D-25
Table	D-32 "Effective" Cross-Section for the $^{17}\text{O}(n,\alpha)^{14}\text{C}$ Reaction in the PWR	D-26
Table	D-33 CE-B Average Production Rates for the $^{17}\text{O}(n,\alpha)^{14}\text{C}$ Reaction	D-27
Table	D-34 "Effective" Cross-Section for the ¹⁴ N(n,p) ¹⁴ C Reaction in the PWR	D-27
Table	D-35 CE-B Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	D-28
Table	D-36 W-G BOL, MOL and EOL Core Flux	D-29
Table	D-37 W-G Average Production Rates for the ¹⁷ O(n,α) ¹⁴ C Reaction	D-30
Table	D-38 W-G Average Production Rates for the ¹⁴ N(n,p) ¹⁴ C Reaction	D-31

1 INTRODUCTION

Improvements in nuclear power plant effluent management practices have resulted in a decrease in the concentration and a change in the distribution of gaseous radionuclides released to the environment. As a result, carbon-14 (¹⁴C) may become a principal radionuclide for the gaseous effluent pathway. In the latest revision of Regulatory Guide 1.21 "Measuring, Evaluating, and Reporting Radioactive Material in Liquid and Gaseous Effluents and Solid Waste", the NRC has recommended that U.S. nuclear power plants evaluate whether ¹⁴C is a principal gaseous effluent, and if so, report the amount of ¹⁴C released. Regulatory Guide 1.21 describes methods acceptable to the NRC for this evaluation.

The objectives of this work were to investigate and develop methods for site-specific estimation of ¹⁴C generation and release from nuclear power plants.

The scope of the work encompassed the following:

- Review of available industry data and literature, specifically NCRP-81, ANSI 18.1 and the GALE codes, relative to estimation of ¹⁴C generation and release via liquid, gaseous and solid waste pathways at PWRs and BWRs.
- Evaluations of techniques and models used in these sources for estimating ¹⁴C generation rates and concentrations in liquid, solid and gaseous waste streams at individual plants.
- Comparisons of the database employed to develop the generation and distribution models to the currently available data base considering both US and overseas experience. Evaluate possible impacts of current chemistry practices on the applicability of the existing database.
- Identification of areas where improvements to the calculations appear possible relative to estimating ¹⁴C generation and releases.
- Evaluation of techniques to monitor and estimate ¹⁴C release rates via liquid, gaseous and solid waste streams.
- Compilation of available information on ¹⁴C releases from PWRs and BWRs.
- Comparisons of industry release rate data to estimates developed from NCRP-81, ANSI 18.1 and GALE.
- Develop guidance for site-specific carbon-14 generation and release calculations based on nuclear reactor physics and industry research and experiences.

Introduction

This report comprises five sections:

• Section 1 Introduction:

Describes carbon-14 as a principle nuclide and summarizes the objectives of this report.

• Section 2 Background:

Discusses the sources of ¹⁴C in the environment and describes the nuclear reactions leading to its production. A summary of the ¹⁴C measurement and reporting requirements is provided.

• Section 3 Carbon-14 Generation and Release in BWR Systems:

Describes the methodology to calculate or estimate the ¹⁴C production rate in the BWR. Discusses chemical forms and release pathways, and presents the results of ¹⁴C measurements at several domestic and foreign BWRs.

• Section 4 Carbon-14 Generation and Release in PWR Systems

Describes the methodology to calculate or estimate the ¹⁴C production rate in the PWR. Discusses chemical forms and release pathways, and presents the results of ¹⁴C measurements at several domestic and foreign PWRs.

• Section 5 Measurement in the Nuclear Power Industry

Describes the methodology for sampling and analysis of ¹⁴C as applied to the nuclear power industry.

2BACKGROUND

2.1 Sources of ¹⁴C in the Environment

2.1.1 14 C Production Reactions

¹⁴C has a half-life of 5700±30 years, and 100 percent of its decay is by beta emission to ¹⁴N. The beta particle end-point energy is 156.475 keV; the mean beta-energy is 49.47 keV and the decay dose is 0.0495 MeV/Bq-sec (Ajzenberg-Selove (1991)). Carbon-14 can be produced by several nuclear reactions (Table 2-1) of which only the ¹⁷O(n,α)¹⁴C and ¹⁴N(n,p)¹⁴C are of significance at light water reactors (LWR).

Table 2-1
Carbon-14 Production Reactions in PWRs and BWRs

Neutron Induced Reaction	Natural Isotopic Abundance of Target Element (%) ^a or Yield ^b
¹⁴ N(n,p) ¹⁴ C	99.632
¹⁷ O(n,α) ¹⁴ C	0.038
¹³ C(n,γ) ¹⁴ C	1.07
Ternary Fission ^b	
²³⁵ U(n,f) ¹⁴ C	1.7 atoms per 10 ⁶ fissions
²³⁹ Pu(n,f) ¹⁴ C	1.8 atoms per 10 ⁶ fissions

a. Chart of the Nuclides, 2002.

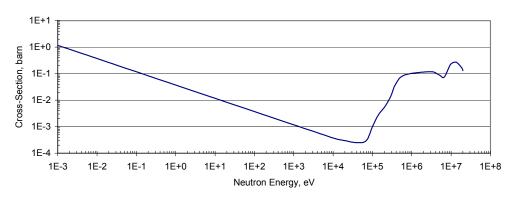
The (n,p) reaction produces 14 C by reaction of neutrons with 14 N. This nitrogen occurs as N_2 or other nitrogen species (ammonia or hydrazine) dissolved in the water, or as nitrogen impurity in the fuel or other core materials. The (n,α) reaction on 17 O produces 14 C by reaction with 17 O present in the fuel and moderator. The (n,γ) reaction on 13 C produces 14 C by reaction with organic materials in the moderator, with the carbon in B_4 C control rods, or with carbon in graphite-moderated reactors. However, the production rate for this reaction is inconsequential for modern light water reactors, i.e., the 14 C production rate per ppm of 13 C is lower than the 14 C production rate per ppm of nitrogen by a factor of approximately 1E-5. The neutron cross-sections for these three reactions are shown in Figure 2-1 (ENDF). All three nuclear reactions have a 1/v region and a significant high energy neutron cross-section. In addition to the generation of 14 C in the reactor coolant, ternary fission produces 14 C in the fuel. Also 14 C

b. Hayes, 1977.

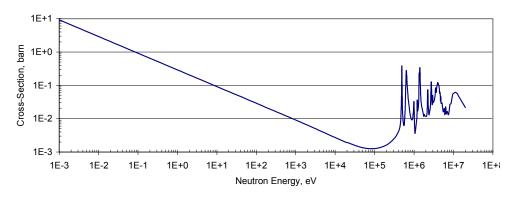
Background

produced by the $^{14}N(n,p)^{14}C$ and $^{17}O(n,\alpha)^{14}C$ reactions in the fuel. However, these reactions do not impact the reactor coolant in the absence of a fuel cladding defect.

Cross-Section for ¹⁷O(n,alpha) ¹⁴C Reaction



Cross-Section for ¹⁴N(n,p)¹⁴C Reaction



Cross-Section for ¹³C(n,gamma) ¹⁴C Reaction

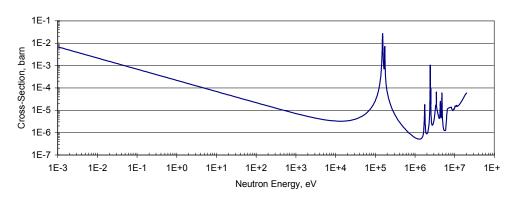


Figure 2-1
Reaction Cross-Sections (ENDF)

The following tabulation (Table 2-2) of cross-sections for the two main ^{14}C production reactions, $^{17}O(n,\alpha)^{14}C$ and $^{14}N(n,p)^{14}C$, is found in JEF Report 14 (1994).

Table 2-2 Reaction Cross-Sections (JEF Report 14, 1994)

¹⁷ O(n,α) ¹⁴ C	Cross Section, barn				
File	2200 M/s	Maxwellian Average	Resonance Integral	14-MeV	Fission Average
JEF-2.2	0.2353	0.2085	0.1059	0.2624	0.09494
ENDF/B-VI	0.2350	0.2350	0.1058	0.2624	0.09494

¹⁴ N(n,p) ¹⁴ C Cross Section, barn					
File	2200 M/s	Maxwellian Average	Resonance Integral	14-MeV	Fission Average
JEF-2.2	1.821	1.613	0.8177	0.04325	0.03547
ENDF/B-VI	1.827	1.827	0.8196	0.04447	0.03830
JENDL-3.2	1.770	1.569	0.7949	0.00420	0.03433
BROND-2	1.923	1.976	14.88	0.05404	0.03490
CENDL-2	1.906	1.737	1.091	0.04324	0.03842

These values were calculated over various regions of the neutron energy spectrum using the parameters shown in Table 2-3.

Table 2-3 Basis for Reaction Cross Section Calculations

Parameter	Type/Energy	Integration Limits
Maxwellian Spectrum Average		
Temperature (20°C)	0.0253 eV	1.0E-5 to 10 eV
Resonance Integral		
Spectrum	1/E	0.5 eV to 100 keV
Fission Spectrum Average		
²³⁵ U Watt Distribution		1 keV to 20 MeV
14 MeV Value	14.0 MeV	

The distribution of neutron energies in a reactor differs from the fission neutron spectrum due to the slowing down of neutrons in elastic and inelastic collisions with fuel, coolant and construction material. The fission spectrum for U-235 and Pu-239 can be approximated by the Watt distribution (Cullen, D. (2004)). The thermal Maxwellian cross-section is calculated at 20°C while LWRs operate between 285°C and 310°C. Neither of these distributions represents the neutron flux distribution in a BWR or PWR. However, these distributions were used to determine the Table 2-2 values for reaction cross-sections, and these reaction cross-sections have been used in the past (sometimes with temperature adjustments) to calculate the ¹⁴C source terms in both reactor types.

2.1.2 Environmental Sources of 14C

There are numerous sources of carbon-14 dioxide ($^{14}\text{CO}_2$) introduction into the environment. The largest single source is cosmic ray reactions in the upper atmosphere where atmospheric nitrogen is transmuted to ^{14}C by the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction at a rate of 2.5 atoms/sec-cm² of earth surface (Suess; 1958). Using an earth surface area of 5.1E18 cm², the production rate is estimated to be approximately 42,000 Ci/year (1.55E3 TBq/year); the total global inventory of ^{14}C is estimated to be 3.45E8 Ci (1.28E7 TBq). ^{14}C has been observed to be in secular equilibrium throughout the biosphere at a concentration of 6.89 picocuries/g C (15.3 dpm/g C). This equilibrium concentration is believed to have been essentially unchanged for at least 15,000 years prior to 1954 when large thermonuclear tests resulted in the production of sufficient amounts of additional ^{14}C to perturb the natural equilibrium.

As shown in Table 2-4, there are numerous exchangeable reservoirs for ¹⁴CO₂ introduced into the biosphere (Suess; 1958).

Table 2-4
Amount of Carbon in Various Exchangeable Reservoirs (Suess, 1958)

Exchangeable Reservoir	Carbon Content, g/cm² Earth Surface	% of Total
Atmospheric CO ₂	0.126	~1.6
Terrestrial Biosphere (living)	0.06	~0.8
Humus	0.215	~2.7
Marine Biosphere	0.002	~0.03
Dissolved Organic Matter in Sea	0.533	~6.8
Total Inorganic Carbon in Sea	6.94	~88.1
Total Exchangeable Carbon:	7.88	

The rate of exchange between these reservoirs is short compared to the average life of ¹⁴C atoms (8297 years). The average residence time for ¹⁴C in the ocean has been estimated to be 300 to 400 years; in the atmosphere, the average residence time is much shorter, i.e., 7±3 years.

Atmospheric nuclear weapon testing has been the second largest contributor of ¹⁴C to the atmosphere. It has been estimated that each megaton of total nuclear energy (fission + fusion) produces (2±1)E26 atoms of ¹⁴C if detonated in the free atmosphere and half that value if detonated at the earths surface (Machta, L., et al.; 1964). At the end of 1962, it was estimated that the total yield of nuclear weapons testing in the atmosphere was 511 megatons (406 megatons in the free atmosphere and 105 megatons at the earth's surface) (Federal; 1962). The calculated production from weapons testing is therefore 91.7E27 atoms of ¹⁴C or 9.51E6 Ci. Since that time, additional atmospheric testing has further increased the ¹⁴C inventory. To put the weapons related ¹⁴C production in perspective, one can estimate that it would require approximately sixteen thousand 3597 MW_{th} BWR/6's operating for 30 years, each generating 20.0 Ci/year, to introduce into the atmosphere the amount of ¹⁴C that has been introduced by above ground weapons testing prior to 1963.

The rate of exchange of carbon dioxide between the atmosphere and the biosphere, humus and surface ocean waters is relatively rapid. Human tissue also comes into equilibrium relatively rapidly with the ¹⁴CO₂ in the atmosphere ("effective" half-life in the body is ~35 days (Raaen, V. F., et al.; (1968)). Prior to weapons testing, the amount of ¹⁴C in the "standard man" acquired from natural sources was on the order of 0.1 μCi (3.7 kBq). The average dose rate from this source was 1.64 mrem/year (1.64E-2 mSv/year) bone and 0.71 mrem/year (7.1E-3 mSv/year) soft tissue and bone marrow (1.06 mrem/year (1.06E-2 mSv/year) whole body) (Raaen, V. F., et al.; 1968). As a result of weapons testing, the dose rate to the "standard man" from ¹⁴C has increased. Model predictions of the yearly dose due to the 14C introduced into the environs by weapons testing prior to 1963 are quite small (Machta, L. and J. H. Harley). In fact, the integrated doses delivered by the year 2000 was calculated to be only 13 mrem whole body.

Projected absorbed dose rates to man from the release of ¹⁴C from nuclear power plants over the period 1955 to 2000 were reported in NCRP Report No. 81. The yearly doses from the natural contribution of ¹⁴C in the body prior to nuclear testing was reported at 1.25 mrad per person annum. The estimated dose attributable to ¹⁴C releases from nuclear power plants is quite small.

2.1.3 Chemical Forms of ¹⁴C Produced in the LWR Primary Coolant

The stable chemical compounds of ¹⁴C produced in the primary coolant generally contain one carbon atom and are essentially limited to the following set of compounds (Table 2-5).

Table 2-5
Possible Chemical Forms Produced from ¹⁴C In-Core Reactions

Single Carbon Species			
CO ₂	Carbon Dioxide		
СО	Carbon Monoxide		
НСООН	Formic Acid		
H ₂ C=O	Formaldehyde		
CH₃OH	Methanol		
CH ₄	Methane		

Rosset (1994) calculated the redox properties of 14 C at 300°C. In the preparation of this potential-pH diagram, the authors adopted the following conditions: $P(CH_4) = 1E-8$ atm, $P(CO) = P(CO_2) = 1E-9$ atm, $[H_2CO_3]_{total} = [CH_3OH] = [H_2C=O] = [HCOOH]_{total} = 2E-12$ mol/L.

The BWR operates at an at-temperature pH (pH_T) of \sim 5.62 and at a potential between \sim 150 mV positive during operation with normal water chemistry (NWC) to hundreds of mV negative (-200 to -600 mV) depending on the location in the reactor system and the extent of hydrogen addition during operation with hydrogen water chemistry (HWC). For the BWR, H₂CO₃, HCO₂, HCO₃ and CH₃OH are indicated to be thermodynamically stable. The PWR operates at a high temperature pH between 6.9 and 7.4 and at a potential of -800 mV where HCO₂ and CH₃OH are the expected stable products. Although these types of potential-pH diagrams provide some indication of the expected chemical forms of ¹⁴C they do not factor in radiolysis reactions and chemical kinetics.

2.2 Measurement and Reporting Requirements at Nuclear Plants

2.2.1 Gaseous and Liquid Effluents

The requirements for gaseous and liquid effluent activity measurements are specified in the Radiological Effluent Technical Specifications (RETS). A typical example for a PWR is given in NUREG-1302 and for a BWR in NUREG-1302. The environmental monitoring requirements are given in NUREG-0475. The RETS include the requirements for effluent on-line radioactivity monitoring as well as for batch sampling and analysis and reporting. They specify the specific nuclides which must be analyzed and define lower levels of detection (LLD) for these nuclides. Reporting of any additional nuclides that are observed also is required. There is no specific requirement to report ¹⁴C in either gaseous or liquid effluents. Since ¹⁴C is a pure beta emitter, it is not identified in a mixture of radionuclides, and in general it has not been given significant consideration since the nuclear power plant release of ¹⁴C has a minimal impact on the world inventory of ¹⁴C. In addition, its impact on local vegetation prior to its dispersion has been given little attention. As reported at the 2010 RETS-REMP Workshop (Sandike, (2010)) the Indian Point Station (2 PWRs) is at least one of the exceptions to these general observations. Based on the work of Kunz from 1978 – 1982, they have assumed a 9.6 Curie release per GW_e -

year for Indian Point 3 (\sim 3200 MW_{th} PWR), with 2.6 Curie per year as CO₂ (26%). Using standard Regulatory Guide 1.109 dose calculations and 2.6 Curies per year per GW(e), the total annual airborne dose at the primary receptor was calculated to be 0.25 mrem per year child bone and 0.051 mrem per year child total body. No credit was taken for the fact that vegetation only is growing approximately 6 months of the year. These values are presented and discussed in their annual Reg. Guide 1.21 submittals.

The RETS were originally part of the plant technical specifications. However, the NRC issued Generic Letter 89-01 in 1991 which permitted utilities to move the RETS and environmental monitoring programs to the Offsite Dose Calculation Manual (ODCM). Most utilities have done this as the ODCM can be modified without prior NRC approval as long as the changes are reported in the Radioactive Effluent Release Report. The annual radioactivity reporting requirements and semiannual release report are specified in Regulatory Guide 1.21, Revision 2, 2009 "Measuring, Evaluating, and Reporting Radioactive Material in Liquid and Gaseous Effluents and Solid Waste". Most plant technical specifications require conformance to Regulatory Guide 1.21. Other guidance documents are Regulatory Guide 4.1, Revision 2, 2009, "Radiological Environmental Monitoring For Nuclear Power Plants", and Regulatory Guide 1.109, Revision 1, 1977, "Calculation of Annual Dose to Man From Routine Releases of Reactor Effluents For the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I".

2.2.2 Regulatory Guide 1.21

Revision 2 of Regulatory Guide 1.21 issued in June 2009 introduced the "risk-informed" principals of the Reactor Oversight Process and included a discussion of ¹⁴C as a potential principal radionuclide for effluent monitoring. The document states "radioactive effluents from commercial nuclear power plants have decreased to a point that ¹⁴C is likely to be a principal radionuclide in gaseous effluents". It further states "because the dose contribution of ¹⁴C from liquid radioactive waste is much less than that contributed by gaseous radioactive waste, evaluation of ¹⁴C in liquid radioactive waste is not required". It requests that licensees evaluate whether ¹⁴C is a "principal radionuclide" for gaseous release from their facility.

A "principal radionuclide" is determined based on its relative contribution to the:

- 1. Public dose compared to the 10CFR50 Appendix I design objectives, or
- 2. Amount of activity discharged compared to other site radionuclides.

"Under this concept, radionuclides that have either a significant activity or a significant dose contribution should be monitored in accordance with a predetermined and appropriate analytical sensitivity level (LLD) outlined in a licensee's ODCM."

The Regulatory Guide further states that the quantity of ¹⁴C in gaseous waste discharged can be estimated by:

- 1. Sample measurements,
- 2. Use of normalized ¹⁴C source term and scaling factors based on power generation (see NCRP Report 81), or
- 3. Use of the GALE code (NUREG-0016 and NUREG-0017) (Reference Sections 3.2 and 4.2)

Background

If sampling is performed, "the sampling frequency may be adjusted to that interval that allows adequate measurement and reporting of effluents." If estimating ¹⁴C in gaseous effluent releases is based on scaling factors and fission rates, a precise and detailed evaluation is not necessary. Also, it is not necessary to calculate uncertainties for ¹⁴C or to include ¹⁴C uncertainty in any subsequent calculation of overall uncertainty.

2.2.3 Solid Wastes (10 CFR Part 61)

10 CFR Part 61, "Licensing Requirements for Land Disposal of Radioactive Waste" classifies solid radioactive waste according to half-lives and quantities of activity. Class A wastes are low concentration wastes that have minimum packaging requirements and are intended to be segregated from higher activity wastes. However, they can be co-mingled with higher class waste if they meet the stability requirements of the higher classification. Wastes containing the long-lived activities listed in Table 1 of 10 CFR Part 60 are either Class A or Class C waste. There is no Class B classification for long-lived nuclides. Table 1 includes ¹⁴C in solid waste forms and as a separate category, ¹⁴C in activated metals. The Class C limiting concentrations are 8 Curies per cubic meter and 80 Curies per cubic meter, respectively. The Class A limits are one tenth of these values. For waste containing mixtures of long-lived activities the sum of the fractions rule applies. 10 CFR Part 61 also specifies manifest requirements for solid wastes shipped to a waste handler or waste disposal site. These requirements are imposed on the waste generator i.e. the nuclear power plant, and require that the nuclide species and their activities determined in fulfillment of 10 CFR Part 61 be listed on the shipment manifest. One of the purposes of these requirements is to track the quantities of long-lived activities shipped to a burial site which ultimately leads to closure of the site based on the total quantity of these longlived species.

Because ¹⁴C is a difficult to measure nuclide, 10 CFR Part 61 permits the use of scaling factors in which the ¹⁴C activity is scaled to a more readily measured nuclide. Scaling factors must be determined for each waste stream on a periodic basis, usually each fuel cycle, or when there is a significant change in the concentration of the more readily measured nuclide. Because of the difficulty of performing long lived nuclide analyses, utilities send samples to outside laboratories for analysis. Recent reviews of scaling factors have been published, e.g., Vance and Cline (Vance1995), IAEA (2004), and IAEA (2009). IAEA (2009) shows that 15 of the 18 listed countries use ¹⁴C/⁶⁰Co scaling factors to determine the ¹⁴C content of their solid radioactive wastes. The two exceptions are India which does not include ¹⁴C in its disposal requirements, and Italy which shut down all its reactors in 1987. Russia and Taiwan, were not included in this review as they are not IAEA members. Sweden no longer uses the ¹⁴C/⁶⁰Co scaling factor because of the wide variation in the ¹⁴C/⁶⁰Co ratios. Most of the solid waste ¹⁴C is found with the primary coolant ion exchange resins, and they estimate the total accumulation of ¹⁴C on these resins based on the MW_e generated over the life of the resins, the established production rate of ¹⁴C in the coolant, the integrated letdown flow, and the expected fractional removal by the resins.

3

CARBON-14 GENERATION AND RELEASE IN BWR SYSTEMS

3.1 Overview of ¹⁴C in the BWR

Carbon-14 is generated in the BWR coolant predominantly by the $^{17}O(n,\alpha)^{14}C$ reaction. Due to the oxidizing environment in the BWR core, the produced species are most likely $^{14}CO_2$, $^{14}CO_3$, $H^{14}CO_2H$, $H_2^{14}CO_3$, and possibly some $^{14}CH_3OH$ (see discussion in Section 2.1.3). Radiolysis reactions and sampling data suggest that the principal species transported in the steam is $^{14}CO_2$. Any transported would be drawn off at the SJAE and would be converted to $^{14}CO_2$ in the recombiner and/or in the environment. The $^{14}CO_2$ and $H^{14}CO_2H$ will partition at the steam/water interface in the reactor coolant and in the condensate. The reactor water cleanup system and condensate treatment system will remove some of the carbonate and formate. The chemical form released to the environment may not be the same as that released from the core since the steam jet air ejector (SJAE) exhaust is passed through a hydrogen/oxygen recombiner and large charcoal delay beds before being released.

3.2 ¹⁴C Source Term Estimations and Measurements

The BWR-GALE code (NUREG-0016 (1979)) assumes that ¹⁴C behaves like a noble gas fission product and is released through the main condenser offgas system. The estimated annual quantity of ¹⁴C generated and released from a BWR is 9.5 Ci/yr. The assumptions in the calculation of the BWR ¹⁴C source term were:

- 3.9E4 kg, mass of water in the reactor core
- 1.3E22 atoms of ¹⁷O/kg of natural water
- 0.80 plant capacity factor
- 1.03E-22 Ci/atom, specific activity of ¹⁴C
- 2.4E-25 cm², thermal neutron cross section for ¹⁷O, and
- 3E13 n/cm²-sec, thermal neutron flux

Note that 14 C is also produced by neutron activation of 14 N in the drywell and dissolved nitrogen in the reactor coolant. These sources are a small fraction of that produced by the 17 O(n, α) 14 C reaction and can be neglected since reactor coolant normally contains less than 0.1 ppm by weight nitrogen and the neutron flux in the drywell is low (estimated at 4E4 n/cm²-sec).

Fowler, et al., (1976) reported on the health considerations of ¹⁴C discharges from the nuclear power industry. This report was later updated by Fowler and Nelson (1979). In their 1976 report, the production rates of ¹⁴C for a reference BWR and a reference PWR were calculated and compared to results of calculations made by others (Table 3-1).

Fowler, et al. (1976) considered activation only by thermal neutrons and decreased the 2200 m/s cross-section value by a factor of 0.6 to correct for the temperature dependence of the thermal neutron cross-section and the thermal neutron spectrum. The mass of coolant in the reactor flux of his reference BWR (3579 MW_{th} MW_{th}

Table 3-1	
Calculated Production Rates of Carbon-14 in Light Water Reactors (Fowler, 1976)	

		Carbon-14 Production Rate (Ci/GW _e -yr)				
	Target	Fowler, et al. (1976)	Bonka, et al. (1974)	Hayes, et al. (1977)ª	ERDA-1535 (1975)	Kelly, et al. (1975) ^a
	O-17	4	8.4	10.9		2.7
BWR Fuel	N-14	18	12.9	21.2		10.9
	Total	22	21.3	32.1	20 ^b	13.6
DWD	O-17	8.9	9.9	11.5		
BWR	N-14	0.26	1.3			
Coolaill	Total	9.2	11.2	11.5	16	16
BWR Sum		31	32.5	43.6	36	29.6

a. The production rates presented by Hayes et al. (1977) and Kelly et al. (1975) for 1000 MW_{th} were multiplied by 3.03 (33% thermal efficiency) to roughly present the values on a per GW_e-yr basis for comparison purposes.

Davis (1977) calculated the quantities of 14 C formed in the fuel, core structural materials, and coolant in light-water cooled reactors, high temperature gas-cooled reactors and in liquid-metal cooled fast breeder reactors. The calculated value for the 17 O(n, α) 14 C reaction was 4.7 Ci/GW_e-yr for the BWR, however, using an alternative method they estimated a coolant 14 C production rate greater than 8 Ci/GW_e-yr but considerably less than 16 Ci/GW_e-yr. The production of 14 C in the fuel was calculated to be 11.5 Ci/GW_e-yr from the 14 N(n,p) 14 C reaction and 3.3 Ci/GW_e-yr from the 17 O(n, α) 14 C reaction.

An extensive sampling and analysis program was performed on the characterization of ¹⁴C in Swedish light water reactors by Magnusson, et al. 2008. The work of Magnusson involved the development of ¹⁴C analysis techniques, mapping of ¹⁴C in waste streams and measurements of ¹⁴C in environmental samples in the vicinities of several Swedish reactors. Highlights of this evaluation are discussed below.

b. Fuel and cladding production rates for ERDA-1535 (1975) were added and identified as a fuel production rate in this table.

The calculated core specific coolant production rates for a Swedish BWR are summarized in Table 3-2. The thermal neutron flux was calculated for the reactor core and considered enrichment and burn-up level for an equilibrium core. The neutron spectrum for higher energies was based on in-core fuel management calculations.

The generic production rate of 14 C in Nordic BWRs had previously been calculated to be 23-24 kBq/MW_{th}—h (Lundgren, et al. 2002). This corresponds to 593 to 619 GBq/GW_e-yr compared to 580 GBq/GW_e-yr in footnote "a" of Table 3-2.

Table 3-2 Calculated Production Rates of ¹⁴C in the Reactor Coolant of a 2500 MW_{th} ASEA-ATOM BWR (Magnusson, et al. 2008)

	Production Rate ^a (Bq/s)			
Target	Thermal	Epithermal	Fission	
¹⁷ O	8.4E3	770	6.5E3	
¹⁴ N ^b	2.7	0.25	0.10	

- a. Production rates correspond to 580 GBq/GW_e-yr (15.7 Ci/GW_e-yr). The overall uncertainty in the calculated production rates was estimated by the authors to be $\pm 20\%$.
- b. Assumes introduction of nitrogen through the BWR scram system equivalent to 100 ppm in the core bypass channel.

Vance et al. (1995) reported production rates of 540-570 GBq/GW_e-yr for BWRs. Vance's source term calculations for the BWR are summarized in Table 3-3. Vance noted that there are significant uncertainties associated with these production rates, principally in the values for the equivalent cross sections.

Table 3-3
Calculated Production Rates of Carbon-14 in BWR Reactor Coolant (Vance, 1995)

Reaction	BWR⁵
¹⁷ O(n,α) ¹⁴ C	14.5 Ci/GW(e)-yr
¹⁴ N(n,p) ¹⁴ C	0.29 Ci/GW(e)-yr/ppm
¹³ C(n,γ) ¹⁴ C	0.027 Ci/GW(e)-yr/ppm

- a. Effective cross-section for the $^{17}\text{O}(n,\alpha)^{14}\text{C}$ reaction: 0.183 b. Effective cross-section for the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction: 1.17 for the $^{13}\text{C}(n,\gamma)^{14}\text{C}$ reaction: 0.006 b.
- b. Thermal flux: 4.8E13 n/cm²-sec, BWR coolant mass exposed to flux: 33,000 kg.

3.3 Site Specific BWR Source Term Estimation

This section provides guidance for estimating carbon-14 source term based on unit specific reactor core physics and reactor design. In order to calculate the unit specific carbon-14 generation, each plant will need their best estimate of their reactor neutron flux profiles (2 or 3 energy groups), mass of coolant in the "active core", and concentration of nitrogen. With this

Carbon-14 Generation and Release In BWR Systems

information, the generation of carbon-14 from oxygen-17 and nitrogen-14 can be calculated and summed for the total carbon-14 production rate.

Given a constant neutron flux and target concentration, the rate of production of a species, N_a , in atoms per second is given by:

$$N_a = N_T \cdot \Sigma(\sigma_i \cdot \varphi_i)$$
, $i = 1$ to 2 or 3^a

a. In some cases there may be only 2 energy groups, $E \le 0.625 \text{ eV}$ and E > 0.625 eV)

where:

 N_a = Rate of production, atoms/sec

N_T = Number of target ¹⁷O or ¹⁴N target species per kg of coolant

 σ_i = "effective" neutron cross-section for each of the 2 or 3 energy groups, cm²

 φ_i = neutron flux for each of the 2 or 3 energy groups, neutron/cm²-sec

The source term of each species A_i, d/s-sec, or Bq/sec is given by:

$$A_a = N_a \cdot \lambda_a$$

Where λ_a is the decay constant of the species.

The source term in μ Ci/sec is given by:

$$A_a (\mu \text{Ci/sec}) = N_a \cdot \lambda_a / 3.7\text{E4 d/sec-}\mu \text{Ci}$$

The following methodology for estimating a site specific BWR ¹⁴C source term is recommended for consideration and was used in Appendix B to calculate the source term for one domestic BWR.

3.3.1 STEP 1: Unit Specific Neutron Flux

Develop values for core "average" neutron flux at the beginning of the cycle (BOC), mid-cycle and end of cycle (EOC) for two or three energy groups for both the moderator (in-channel) and bypass (leakage) regions of the core. For illustration purposes, the following three energy flux data are used (Table 3-4):

Table 3-4 Example of Flux Variations over BWR Fuel Cycle^a

вос		Neutron Flux (φ), n/cm²-sec	
Neutron Group	Group Energy	Moderator	Bypass
Thermal	≤0.625 eV	3.70E13	5.81E13
Intermediate	>0.625 eV - <1 MeV	1.31E14	1.26E14
Fast	≥1 MeV	4.41E13	3.91E13
Mid-Cycle		Neutron Flux (φ), n/cm²-sec	
Neutron Group	Group Energy	Moderator	Bypass
Thermal	≤0.625 eV	3.87E13	5.96E13
Intermediate	>0.625 eV - <1 MeV	1.33E14	1.28E14
Fast	≥1 MeV	4.50E13	3.98E13
EOC		Neutron Flux (φ), n/cm²-sec	
Neutron Group	Group Energy	Moderator	Bypass
Thermal	≤0.625 eV	4.14E13	6.22E13
Intermediate	>0.625 eV - <1 MeV	1.34E14	1.30E14
Fast	≥1 MeV	4.56E13	4.03E13

a. Private communication from Ken Watkins to H. Helmholz.

Not all BWRs have a core average three sectioned neutron flux distributions readily available for this calculation. Some sites will have only a two group neutron flux distribution; ≤ 0.625 eV, and ≥ 0.625 eV. The ≥ 0.625 eV value is the sum of the intermediate and fast flux.

3.3.2 STEP 2: Effective Cross-Sections and ¹⁴C Generation Rates

Use the "Effective Cross-Sections", provided in Tables 3-5 and 3-6, in the two or three neutron energy groups to calculate the source term for the two major production reactions in units of μ Ci/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction and μ Ci/sec-kg-ppm N for the $^{14}N(n,p)^{14}C$ reaction. The methodology utilized to determine the values of "effective cross-section" employed in this section is presented in Appendix A.

Table 3-5 "Effective Cross-Sections" for the $^{17}O(n,\alpha)^{14}C$ Reaction in the BWR

		"Effective Cross-Section (σ)", b	
Neutron Group	Group Energy	Moderator	Bypass
Thermal	≤0.625 eV	0.1325	0.1386
Intermediate (I)	>0.625 eV - <1 MeV	0.0238	0.0222
Fast (F)	≥1 MeV	0.1106	0.1106
I+F	>0.625 eV	0.0458	0.0432

Table 3-6
"Effective Cross-Sections" for the ¹⁴N(n,p)¹⁴C Reaction in the BWR

		"Effective Cross-Section (σ)", b	
Neutron Group	Group Energy	Moderator	Bypass
Thermal	≤0.625 eV	1.0560	1.0903
Intermediate (I)	>0.625 eV - <1 MeV	0.0384	0.0423
Fast (F)	≥1 MeV	0.0479	0.0478
I+F	>0.625 eV	0.0408	0.0437

3.3.2.1 Production Rate from $^{17}O(n,\alpha)^{14}C$ Reaction

The 14 C production rate from the 17 O(n, α) 14 C reaction is calculated using the three energy group flux distribution as follows:

$$Production \ Rate \ (\mu Ci/sec - kg \) = \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i} \bullet \phi_{i} + \sigma_{f} \bullet \phi_{f} \] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

1.27E22 atoms ¹⁷O/kg H₂O N "effective" thermal cross-section, b σ_{th} thermal neutron flux, n/cm²-sec ϕ_{th} "effective" intermediate cross-section, b σ_{i} intermediate neutron flux, n/cm²-sec = ϕ_i "effective" fast cross-section, b = $\sigma_{\rm f}$ fast neutron flux, n/cm²-sec ϕ_f conversion factor, 1.0E-24 cm²/b 1.0E-24 ¹⁴C decay constant, 3.833E-12/sec = λ conversion factor, 3.7E4 d/sec-µCi 3.7E4

The calculated production rate is as follows:

	Production Rate, μCi/sec-kg			
	Moderator Bypass			
BOC	1.697E-5	1.996E-5		
Mid-Cycle	1.746E-5	2.040E-5		
EOC	1.805E-5	2.100E-5		
Average:	1.749E-5	2.046E-5		

3.3.2.2 Production Rate of ¹⁴C from ¹⁴N(n,p)¹⁴C Reaction

The production of ¹⁴C via the ¹⁴N(n,p)¹⁴C nuclear reaction is a minor component to the overall ¹⁴C source term for domestic BWRs and can be neglected. Nevertheless, this discussion is provided for completeness.

The ¹⁴C production rate for the ¹⁴N(n,p)¹⁴C reaction for the three energy groups is calculated as follows:

Production Rate(
$$\mu$$
Ci/sec-kg-ppmN) =
$$\frac{N \bullet [\sigma_{th} \bullet \varphi_{th} + \sigma_i \bullet \varphi_i + \sigma_f \bullet \varphi_f] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

4.284E19 atoms ¹⁴N/kg-ppm N N "effective" thermal cross-section, b σ_{th} thermal neutron flux, n/cm²-sec ϕ_{th} "effective" intermediate cross-section, b σ_{i} intermediate neutron flux, n/cm²-sec "effective" fast cross-section, b $\sigma_{\rm f}$ fast neutron flux, n/cm²-sec conversion factor, 1.0E-24 cm²/b 1.0E-24 ¹⁴C decay constant, 3.833E-12/sec λ = 3.7E4 = conversion factor, 3.7E4 d/sec-µCi

The calculated production rate is as follows:

	Production Rate, μCi/sec-kg-ppm N			
	Moderator Bypass			
BOC	2.051E-7	3.131E-7		
Mid-Cycle	2.136E-7	3.209E-7		
EOC	2.266E-7	3.339E-7		
Average:	2.151E-7	3.226E-7		

3.3.3 STEP 3: Unit Specific Coolant Mass

Determine the mass of coolant in the "active" core in units of kilograms of H₂O. It is suggested that utilities use the mass of reactor coolant in the fuel cells from the "bottom" of the active core to the "top" of active core and not include the fluid flowing around the core inside the shroud. Develop mass values for both the moderator and bypass regions of the active core. These liquid masses are used to calculate the ¹⁴C source term.

NOTE: Since plant and fuel designs vary, it is necessary to develop plant specific values for the moderator and bypass regions of the core.

3.3.4 STEP 4: 14 C Source Term

Calculate the ¹⁴C source term based on the above generation rates considering plant specific values of power, in-core coolant mass and coolant nitrogen concentration.

For illustration purposes, consider a 3579 MW_{th} BWR with a moderator water mass of 12,655 kg and a bypass region water mass of 17,100 kg. The coolant nitrogen concentration is assumed to be <10 ppb.

3.3.4.1 ¹⁴C Source Term from ¹⁷O(n,a) ¹⁴C Reaction

```
12,655 kg • 1.749E-5 μCi/sec-kg + 17,100 kg • 2.046E-5 μCi/sec-kg = 0.571 μCi/sec = 18.0 Ci/yr = 0.574 μCi/MW<sub>th</sub>-h = 5.03 Ci/GW<sub>th</sub>-yr = 14.8 Ci/GW<sub>e</sub>-yr @ 34% efficiency = 21.3 kBg/MW<sub>th</sub>-h
```

3.3.4.2 ¹⁴C Source Term from ¹⁴N(n,p)¹⁴C Reaction

```
12,655 kg • 2.151E-7 μCi/sec-kg-ppm N + 17,100 kg • 3.226E-7 μCi/sec-kg-ppm N = 8.239E-3 μCi/sec-ppm N
```

For 10 ppb N in the reactor coolant during steady state operation, this reaction will produce only 0.0026 Ci/yr or $\sim 0.01\%$ of the generation rate based on the $^{17}O(n,\alpha)^{14}C$ reaction. Note that it is expected that the reactor coolant nitrogen concentration will be much less than 10 ppb during power operation. For example, at a feedwater nitrogen concentration of 100 ppb (considered a maximum value during power operation), the reactor water concentration will be <1 ppb.

3.3.5 Summary of 14 C Source Term Calculations

In conclusion, the ¹⁴C source term is the sum of the production rates from ¹⁷O and ¹⁴N. However, the nitrogen concentration in the reactor coolant is so low that the ¹⁴C generation from nitrogen can be neglected.

Calculation of the ¹⁴C production rate from ¹⁷O requires knowledge of in-core coolant mass, the two or three energy group neutron flux distribution, and the "effective" neutron cross-sections provided in this report.

3.4 Release Pathways and Chemical Form

It is known that ¹⁴C is released from the core in a volatile form such as ¹⁴CO or ¹⁴CO₂. The ¹⁴CO will be drawn off at the SJAE and will be converted to ¹⁴CO₂ in the recombiner and/or in the environment. The ¹⁴CO₂ will partition at the steam/water interface in the reactor vessel and in the condenser.

If CO₂ is the principal species produced in the BWR coolant, the fraction of the ¹⁴CO₂ carried over in the steam can be calculated based on high temperature partitioning and hydrolysis data for CO₂ and data for the dissociation of water. Cobble (1982) tabulated the high temperature free energy of formation for species of interest in the carbonate system, and Sweeton, F. H., et al. (1974) has published data on the dissociation of water at high temperatures.

From these data, the following reactions are postulated for carbon dioxide in a BWR operating at a saturated steam pressure of 7.0 M Pa (1015 psia) and 286°C (~550°F).

The hydrolysis of $CO_{2(l)}$ to $H_2CO_{3(aq)}$ is so fast that the concentration of $H_2CO_{3(aq)}$ is the concentration of $CO_{2(l)}$. The $H^+_{(aq)}$ is 2.37E-6 mol (at pH_T = 5.63). To illustrate this calculation, a concentration of $H_2CO_{3(aq)}$ of 1E-8 mol was assumed. The calculated concentration of the carbon species is tabulated below.

Species	Concentration, Molal	Percent
$[H_2CO_{3(aq)}] = [CO_{2(l)}]$	= 1.000E-8	99.6845
[HCO _{3 (aq)}]	= 3.165E-11	0.3155
[CO _{3 (aq)}]	= 4.05E-17	0.0000
Total:	= 1.003165E-8 molal	

Based on these calculations, $H_2CO_{3(aq)}$ represents 99.6845% and $[HCO_{3(aq)}]$ represents 0.3155% of the carbon species at the BWR operating temperature.

The partitioning between the liquid and gas phase is given by:

$$K = \frac{P[CO_{2(g)}]}{H_2CO_{3(aq)}} = 7.9 \text{ M Pa kg/mole}$$

$$P[CO_{2(g)}] = 7.9E-2 \text{ Pa (7.80E-7 atm)}$$

Assuming an ideal gas and saturated steam at a temperature of 285.9 °C (559.0 K), the steam CO₂ concentration will be;

 $n/V(\text{moles/liter}) = 7.80E-7 \text{ atm/}[(0.082054 \text{ l-atm/K-mole} \cdot 559.01 \text{ K})] = 1.70E-8 \text{ moles/liter}$

and the mass partition coefficient will be:

Mass Partition Coefficient =
$$\frac{(1.70E - 8 \text{ mol/L})/0.0365533 \text{kg/L}}{(1.0E - 8 \text{ M/kg})} = 46.5$$

The above value agrees very well with a more recent value of 42.6 for the mass partition coefficient (IAWPS, 1998) and 43.09 in the EPRI MULTEQ code.

For a 3578 MW_t (238/732) BWR with a core flow of 105.0E6 #/h and a steam flow of 15.4E6 #/h, the ratio of the steam to return water transport rate is

$$\frac{15.4E6 \#/h \cdot 42.6}{(105.0 - 15.4)E6 \#/h} = 7.322$$

The steam removal for a single pass of water through the core is $7.322 \cdot 100/8.322 = 88.0\%$. The time to reach equilibrium with such a high partition coefficient is only a few minutes and the measured concentration of 14 C in the reactor coolant returning from the steam/water interface is only about 13.6 % of the concentration in the steam/water mixture exiting the core.

At an estimated production rate of 2.05E3 μ Ci/h (18.0 Ci/y) for the example 3578 MW_{th} BWR, the concentration of ¹⁴C in the returning liquid from the steam/water interface should be ~5.9E-9 μ Ci/g. The observed concentrations of ¹⁴C in the BWR coolant is considerably higher (8E-8 to 4E-6 μ Ci/g) which indicates that other less volatile species of ¹⁴C are also produced.

Few fundamental studies on the reactions of hot carbon atoms in aqueous solutions have been performed. The most extensive study was made by Stenström (1970) who studied the reactions of ¹¹C atoms in aqueous solutions. (Note: The chemical fate of ¹⁴C is expected to be identical to that observed by Stenström (1970) for ¹¹C.) Numerous reaction products were observed and the yield of any one product was found to be dose dependent. Selected data from Table 8 in Stenström's thesis has been reproduced below as Table 3-7.

Table 3-7
Yields of "C Species in Triple Distilled, Degassed Water Irradiated with 185 MeV Protons, in Per Cent of the Total Yield of "C

Dose (rad)	¹¹ CO ₂	11 CO	H¹¹COOH	H¹¹CHO	¹¹CH₃OH	¹¹ CH₄	Not Identified
160	23 ± 1	32 ± 1	17 ± 2	21 ± 2	6 ± 2	0.19 ±0.03	0.9
170	22 ± 2	32 ± 2	20 ± 2	19 ± 3	5 ± 2	0.27 ±0.05	1.8
180	30 ± 1	34 ± 2	11 ± 1	19 ± 3	4 ± 1	0.19 ±0.04	1.3
370	34 ± 2	32 ± 2	14 ± 2	16 ± 2	4 ± 2	0.18 ±0.05	1.1
370	27 ± 1	25 ± 1	25 ± 3	14 ± 2	6 ± 2	0.23 ±0.02	2.6
860	47 ± 1	16 ± 1	16 ± 1	14 ± 2	5 ± 1	0.21 ±0.02	1.4
1600	65 ± 5	7.7 ± 2	14 ± 3	9 ± 3	3 ± 1	0.19 ±0.02	1.4
1700	76 ± 5	9.0 ± 0.5	10 ± 5	3.6 ± 0.5	1.3 ± 0.5	0.18 ±0.02	0.3
1700	39 ± 2	4.4 ± 0.5	37 ± 3	10 ± 2	4 ± 1	0.22 ±0.01	5.5
2100	62 ± 3	2.0 ± 0.4	29 ± 3	3 0.3	1.1 ± 0.4	0.18 ±0.02	1.6
3400	94 ±2	1.8 ± 0.3	2 ± 1	1.4 ± 0.3	0.2 ± 0.1	0.11 ±0.03	0.2
3400	63 ± 2	1.1 ± 0.1	29 ± 2	1.8 ± 0.4	0.8 ± 0.3	0.15 ±0.01	3.9
5200	53 ± 1	1.3 ± 0.1	40 ± 3	2.0 ± 0.5	0.7 ± 0.2	0.16 ±0.01	3.5
6800	57 ± 1	0.6 ± 0.05	39 ± 2	1.0 ± 0.2	0.5 ± 0.2	0.09 ±0.01	1.6
6800	90 ± 1	0.8 ± 0.1	8 ± 1	0.2 ± 0.1	0.13 ± 0.03	0.09 ± 0.01	0.3
13700	97 ± 0.5	0.24 ± 0.05	1.9 ± 0.5	0.3 ± 0.1	0.09 ± 0.04	0.032 ±0.003	0.03

At low doses (160 rad), CO₂ (23±1%), CO (32±1%), HCO₂H (17±2%), H₂CO (21±2%), CH₃OH (6±2%) and CH₄ (0.2±0.03%) are the observed reaction products. At higher doses (>20,000 rad), the dominant product is CO₂. In a typical BWR/6, the in-core radiation field is in the range of 1E5 rad/sec (neutron + gamma) and since in-channel transit time is on the order of 1 second, doses considerably in excess of 20,000 rads are expected. As such ¹⁴CO₂ should be the dominant reaction product released from the BWR core. However, it is worthwhile to note that the chemical form of ¹⁴C released to the environment may not be the same as that released from the reactor coolant since in the modern BWR, the SJAE exhaust is passed through a hydrogen/oxygen recombiner and large charcoal delay beds before being released.

3.4.1 Nine Mile Point Unit-1

Kunz (1976) reported on measurements made at the Nine Mile Point Unit-1 (NMP-1). Samples were taken of the process off-gas from the main condenser air ejectors. Sample aliquots of 100 to 500 cm^3 were mixed with measured amounts of carrier gases (Kr, Xe, CO₂, CO, CH₄, C₂H₆, C₃H₈ and C₄H₁₀). The carriers were separated from the sample using cryogenic and chromatographic techniques and the separated fractions were individually loaded into gas proportional tubes for counting. The total ¹⁴C in the sample was separately determined by passing another aliquot with added carriers over CuO at 800°C prior to counting. The results of these measurement were 95% CO₂, 2.5% CO and 2.5% hydrocarbons.

The amount of 14 C discharged was estimated by multiplying the activity ratio, 14 C/ 133 Xe, as measured in their samples, by the release of 133 Xe reported by the utility. The release of 14 C from NMP-1 was estimated at 8 Ci/yr. NMP-1, at that time, was rated at 1850 MW_{th}, and assuming operation at 80% capacity, the annual release is estimated at 16 Ci/3000 MW_{th} for a larger BWR.

3.4.2 Oyster Creek

D. Blanchard, et al. (1976) study at Oyster Creek BWR (1860 MW_{th}) indicated an average $^{14}\mathrm{C}$ release rate of 0.182 $\mu\mathrm{Ci/sec}$ from the air ejector and a release rate of <3E-4 $\mu\mathrm{Ci/sec}$ at the turbine gland seal condenser exhaust. The estimated amounts of $^{14}\mathrm{C}$ effluent release during the second half of 1971 through the first half of 1973 are reported in Table 3-8. At both the air ejector (Table 3-9) and turbine gland seal condenser, there was about twice the release rate of $^{14}\mathrm{C}$ as CO_2 as compared to other chemical species. The average reactor coolant $^{14}\mathrm{C}$ concentration after the steam separation was reported to be 4.0E-6 $\mu\mathrm{Ci/ml}$ (Table 3-10). The $^{14}\mathrm{C}$ in the laundry waste was 0.15 pCi/ml.

Table 3-8
Oyster Creek BWR Carbon-14 Measurements (Blanchard, et al.,1976)

Source	Annual ¹⁴ C Discharge Rate(Ci/yr)
Gaseous Releases ^a	
Main Condenser Air Ejector Off-gas	3.0
Turbine Gland Seal	0.005
Reactor Drywell	0.00096
Building Ventilation Air	1.2
Stack	9.1
Liquid Releases	
Waste Sample Tank	0.008
Laundry Drain Tank	0.0001

a. The stack sample probe is located high in the stack and all gaseous effluent, including the offgas system, are discharged to the stack.

Table 3-9 Concentration of ¹⁴C Released from the Oyster Creek Main Condenser Steam Jet Air Ejectors^a

	Concentra		
Sample Date	Non-CO ₂	CO ₂	% CO ₂
Jan. 18, 1972	(7.2±0.8)E-6	(5.5±0.8)E-6	43
Apr. 10, 1972	(3.5±0.8)E-7	(4.2±0.8)E-6	92
Apr. 12, 1972	(2.5±0.5)E-7	(2.3±0.2)E-6	90
Aug. 24, 1972	(1.8±0.8)E-7	(2.8±0.1)E-6	94
Dec. 13, 1972	(1.5±0.9)E-7	(1.5±0.4)E-6	91
Mar. 28, 1973	(1.0±0.5)E-7	(1.2±0.1)E-7	55

a. Concentration measured at beginning of the 75-min delay line, no recombiner.

Table 3-10 Concentration of ¹⁴C in Reactor Water at Oyster Creek

Sample Date	μCi/ml
Aug. 31, 1971	<1E-6
Nov. 30, 1971	1.5E-5
Mar. 14, 1972	<1E-6
Dec. 13, 1972	<1E-6

3.4.3 Fowler's Summary

Fowler, et al. (1976) summarized released source terms for the BWR and PWR utilizing data available to them from other studies or measurements. The results of these efforts yielded the values listed in Table 3-11 for the normalized BWR.

Table 3-11
BWR Carbon-14 Source Terms^a (Nominal 1250 MW_a at 80% Capacity Factor)

Source	Annual ¹⁴ C Discharge Rate (Ci/yr) or (Ci/GW _e -yr)
GASEOUS SOURCE TERMS	
Main Condenser Air Ejector Off-gas	9.0
Turbine Gland Seal	<0.015
Containment Purge	0.0088
Turbine Building Ventilation	0.030
Radwaste Facility Ventilation	0.000044
	Total: 9.0
LIQUID SOURCE TERMS	
Clean Wastes	0.029
Dirty (Low Purity) Wastes	0.013
Chemical Wastes	0.0023
Detergent Wastes	0.000075
	Total: 0.044

a. Fowler, et al. (1976).

3.4.4 Brunswick Units 1 and 2

NUREG/CR-4245 (1985) presents data obtained at Brunswick Unit 1 and 2 as a part of the In-Plant Source Term Measurement Program conducted for the Office of Nuclear Regulatory Research and Meteorology and Effluent Treatment Branch of the Office of Nuclear Reactor Regulation. This program was performed by the Idaho National Engineering Laboratory as the prime contractor. The Brunswick plant was the sixth in a series of operating LWRs to be studied and the first BWR in the series.

Two nearly identical BWRs (BWR 4, Mark 1 containment) share the Brunswick site. At the time of the measurements each unit had a generating capacity of 2436 MW_{th} and a net electrical output of 790 MW_e . Unit 1 began commercial operation in March 1977 and Unit 2 in November 1975. Both units have since been up-rated to 937 MW_e .

Liquid wastes are collected by either the floor drain collection or waste collection systems. Liquids from both units are mixed in the final collection receiver tanks and then processed for release or return to the system.

There are five gaseous release pathways to the environment from the Brunswick Station. They are the Unit 1 and Unit 2 reactor building ventilation exhausts, the Unit 1 and Unit 2 turbine building exhausts and the main stack (common to both units). The augmented gaseous radwaste system for the steam jet air ejectors was not operable during the in-plant measurement period, and the condenser air ejector was discharged to the main stack.

The measurements were performed from March 14, 1982 to November 17, 1982. Unit 2 was down for refueling from April 23 to September 30. Unit 1 was shut down from July 17 to October 17 and also has outages from June 2-5, and October 22-26. It also experienced very short shutdowns on April 20, May 6, June 18, June 28 and July 10. Samples from both liquid and gaseous process streams from both units were collected and analyzed.

The gaseous release results are given in Table 3-12. ¹⁴C releases from each reactor building during power operation were nearly the same. The release of ¹⁴C in the reactor building exhaust air was not influenced much by operation of either plant and most of the ¹⁴C released from both reactor buildings was in the oxidized form. ¹⁴C release from the Unit 2 turbine building vent was ten times that of Unit 1 prior to shutdown. The higher release may have been due to a leak in the Unit 2 steam jet air ejector which would feed the Unit 2 turbine vent. After repair and startup of the steam jet air ejector leaks, Unit 2 turbine building vent releases decreased to three times those at Unit 1. The radwaste building contributed about 1% of the ¹⁴C to the main stack releases during power operation.

Table 3-12 ¹⁴C Measurements at Brunswick Units 1 and 2 (NUREG/CR-4245, 1985)

	Unit 1 Turbi	ne Building	Unit 2 Turbi	ne Building
Date in 1982	μCi/sec	μCi/sec % Oxidized		% Oxidized
3/15 - 3/21	(1.2±0.5)E-4	n.d.	(2.4±0.1)E-2	0
3/21 - 4/01			(8±1)E-3	3
4/01 – 4/15	(9±4)E-5	18	(8±2)E-4	n.d.
4/15 – 4/22	(1.2±0.5)E-4	93	(1.2±0.5)E-4	93
4/22 - 4/23	(8±3)E-5	79	(8±3)E-5	79
4/22 - 5/09			(9±1)E-4	18
5/09 - 5/20	(2.0±0.8)E-4	100	(1.4±0.2)E-3	15
5/20 - 6//03	(1.4±0.6)E-4	100	(3.1±0.9)E-4	64
6/03 - 6/17	(1.6±0.5)E-4	100	(2.2±0.7)E-4	50
6/17 – 7/02	(1.7±0.5)E-4	95	(1.9±0.4)E-4	95
7/02 – 7/15	(8±3)E-5	45	(1.8±0.4)E-4	56
7/22 – 8/11	(2±1)E-5	29	(2.0±0.8)E-5	24
9/29 - 10/14	(1.9±0.8)E-5	100	(1.4±0.4)E-4	20
10/14 - 10/27	(4±2)E-5	87	(8±3)E-5	58
10/27 – 11/09	(3±1)E-5	93	(6±2)E-5	77
11/09 – 11-17	(2.1±0.8)E-5	100	(7±3)E-5	65

	Unit 1 React	or Building	Unit 2 React	or Building
Date in 1982	μCi/sec	% Oxidized	μCi/sec	% Oxidized
3/18 - 4/01	(5±2)E-4	100	(5±2)E-4	100
4/01 – 4/15	(6±2)E-4	100	(5±1)E-4	80
4/15 – 4/22			(8±2)E-4	69
4/15 - 5/08	(1.0±0.3)E-3	37		
4/22 - 5/08			(5±2)E-4	100
5/08 - 5/20			(7±2)E-4	96
5/09 - 5/20	(1.1±0.3)E-3	95		
5/20 - 6//03	(8±3)E-4	100	(4±1)E-4	85
6/03 – 6/17	(7±3)E-4	84	(1.0±0.4)E-4	75
6/17 – 7/02	(6±2)E-4	100	(1.5±0.6)E-4	100
7/02 – 7/15	(1.0±0.2)E-3	79	(3±1)E-4	100
7/22 – 8/06			(2.1±0.8)E-4	n.d.
7/22 – 8/11	(6±3)E-4	n.d.		
9/29 - 10/14			(2.3±0.9)E-4	100
10/10 – 10/27	(1.1±0.4)E-3	73		
10/14 – 10/27			(6±3)E-4	98
10/27 – 11/09	(1.0±0.4)E-3	97	(2.2±0.9)E-4	66
11/09 – 11-17	(5±2)E-4	100	(3.4±0.5)E-3	59

Table 3-12

14C Measurements at Brunswick Units 1 and 2 (NUREG/CR-4245, 1985) (continued)

	Radwa	aste	Main S	tack
Date in 1982	μCi/sec	% Oxidized	μCi/sec	% Oxidized
3/21 – 4/01	(1.5±0.6)E-3	n.d.		
4/01 – 4/15	(1.9±0.3)E-3	100		
4/15 – 4/22	(1.1±0.2)E-3	100		
4/19 – 4/23			(2.8±0.1)E-1	97
4/22 - 4/23	(2.6±0.4)E-3	100		
4/23 – 4/26			(5.1±0.3)E-1	100
5/09 - 5/20	(3.6±0.6)E-3	100		
6/07 - 6/16			(2.8±0.2)E-1	93
6/16 - 7/02			(3.3±0.2)E-1	100
7/02 – 7/15			(4.3±0.2)E-1	67
7/22 – 8/11			(1.0±0.2)E-2	65
9/29 - 10/14			(8.6±0.4)E-2	100
10/10 - 10/27			(2.9±0.1)E-1	100
10/27 – 11/09			(3.16±0.02)E-1	100
11/09 – 11/17			(3.5±0.2)E-1	n.d.

n. d. – not determined

The reactor buildings showed 70 to 100% oxidized ¹⁴C during power operation and 60 to 70% during shutdowns. When both units were running, ¹⁴C from the Unit 2 turbine building showed 3 to 30% oxidized form compared with 18 to 93% in Unit 1. This difference was probably a result of the steam jet air ejector leak in Unit 2.

In Table 3-13, an attempt has been made by authors of this EPRI report to estimate the individual unit main stack releases of 14 C for the Brunswick units. The estimated percent power during the sampling intervals was obtained by manually interpreting reactor power graphs presented in their report. With exception of the first data point (which has been included in the average value), the estimated μ Ci/MW_{th}-h values were reasonably consistent. This average value of 0.68 μ Ci/MW_{th}-h was calculated for the main stack release of 14.5 Ci for a single unit operating at 2436 MW_{th} for one year. For the current up-rated operating power, it reasonable to increase this value in direct proportion to the up-rated operating MW_{th}.

Table 3-13
Estimation of Brunswick Main Stack Release, μCi/MW_{.,}-h

	Measured		Estimated	l % Power	E attaca da at	Fatimata d
Measurement Date in 1982	Main Stack Release, μCi/sec	% Oxidized	Unit 1	Unit 2	Estimated Total MW _{th}	Estimated µCi/ MW _{th} -h
4/19 – 4/23	0.28±0.01	97	47	81	3118	0.32
4/23 – 4/26	0.51±0.03	100	62	59	2948	0.62
6/07 – 6/16	0.28±0.02	93	61	0	1486	0.68
6/16 – 7/02	0.33±0.02	100	66	0	1608	0.74
7/02 – 7/15	0.43±0.02	67	72	0	1754	0.88
7/22 – 8/11	0.010±0.002	65	0	0	0	
9/29 - 10/14	0.086±0.004	100	1	8	219	
10/10 – 10/27	0.29±0.01	100	25	36	1486	0.70
10/27 – 11/09	0.316±0.002	100	69	0	1681	0.68
11/09 – 11/17	0.35±0.02	n.d.	73	0	1778	0.71
					Average:	0.68

3.4.5 J. A. FitzPatrick BWR

Kunz (1985) measured the total 14 C release and chemical form of the 14 C species in the off-gas stack and buildings ventilation from the 850 MW_e J. A. FitzPatrick BWR. The off-gas stack was monitored on a continuous basis starting July 10, 1980 for a 98 week period. During this measurement period the advanced off-gas system was not in operation. A second continuous sampler was used to measure the total gaseous 14 C discharge from the building vents for 60 to 115 days per vent. The building ventilation air varied from <7E-12 to 4E-10 μ Ci/cm³. The release rates for the turbine building, reactor building, radwaste building and refuel floor were 0.05, 0.02, 0.06 and 0.25 Ci/GW_e-yr, respectively.

The results of the measurements are shown in Table 3-14. The gaseous discharge was determined to be 95% $^{14}\text{CO}_2$ and 5% hydrocarbon gases in agreement with measurements at four West German BWRs (Schwibach, et al., 1978) and six BWRs in the United States (Wahlen, 1978). The release rate was calculated to be 12.4 Ci/GW_e-yr (4.2 Ci/GW_{th}-yr at an assumed efficiency of 34%).

Two sets of primary coolant samples were taken before and after the clean-up and condensate demineralizers. No decontamination for ¹⁴C was detected for the deep-bed condensate demineralizers. This is not to say that there is no ¹⁴C, as carbonate, retained on the bed resins, rather, it can be interpreted to mean that the beds were saturated with respect to CO₂.

The first set of reactor coolant samples were taken in 1978 and the measured concentration in the coolant at the RWCU inlet was 2E-6 μ Ci/ml whereas the outlet was 2.9E-7 μ Ci/ml, a removal efficiency of ~86%. A waste water composite contained 1.2E-7 μ Ci/ml and at the estimated wastewater discharge rate, this liquid release pathway would be ~7E-5 Ci/GW_e-yr.

Table 3-14
Carbon-14 Gaseous Release Rate, Chemical Form and Discharge Pathways for the J. A. FitzPatrick BWR (Kunz, 1985)

Parameter	Value
Gaseous Release Rate	Ci/GW _e -yr
Off-gas Stack	12.0
Turbine Building Vent	0.05
Reactor Building Vent	0.02
Radwaste Building Vent	0.06
Refuel Floor Vent	0.25
Total:	12.4
Chemical Form	
¹⁴ CO ₂	95%
¹⁴ CH ₄ , ¹⁴ C ₂ H ₆ , etc.	5%
Discharge Pathway	
Building Ventilation	3%
Off-gas Venting	97%

3.4.6 Nordic BWRs

Lundgren, et al., (2002) calculated the production rate in a standard Nordic BWR to be 23-24 kBq/MW_{th}—h which is consistent with the 22 kBq/MW_{th}—h calculated by Vance (1995).

Unlike US BWRs, the design of the hydraulic scram system in the Nordic BWRs is such that there is some exposure of the CRD purge water to high pressure (72 bar) nitrogen gas where some fraction of the gas does dissolve in the purge water. This CRD purge water is directed through the fuel assembly bypass channel. Their calculations indicated that N_2 in the CRD purge water could impact the production of 14 C in the Nordic BWRs and that plant-specific evaluations were necessary to assess this impact.

The ¹⁴C production in reactor fuel and components is estimate to be 72 kBq/MW_{th}-h as shown in Table 3-15. The corrosion of in-core stainless steel materials and subsequent release of the ¹⁴C to the coolant was estimated at 0.004 kBq/MW_{th}-h. The estimated impact of the corrosion rate of the larger surface area Zircaloy is 0.55 Bq/MW_{th}-h (only 2.3% of the production in the reactor coolant).

Measured stack release of ¹⁴C in the Nordic BWR were normally equal or slightly lower than the calculated production rate. The two Olkiluoto plants showed an increased release of ¹⁴C during startup. It was suggested that the cause was additional production due to nitrogen gas in the scram system and/or accumulated inventory release from the off-gas delay system during the startup.

Their chemistry evaluation concluded that only the oxidized form of 14 C (carbon dioxide and bicarbonate) should be present in the reactor. They further stated that the interaction of the steam born 14 CO₂ with out-of-reactor surfaces may be responsible for the small percentage of methane (\sim 5%) seen in the gaseous effluent.

Table 3-15

14C Production in BWR Reactor Fuel and Internals (based on FSAR for F3/O3)^a

Component	Material	Weight (kg)	N(%)	¹⁴ C (Bq/MW _{th} -h)
Fuel	UO ₂ – fission	141400	0.0014	6.3E2
	UO ₂ – activation	141400	0.0014	2.2E4
	Zircaloy-2	34300	0.0040	1.3E4
	Zircaloy-4	25900	0.0040	1.6E4
	SIS-2333	7000	0.0040	6.4E3
	Inconel-X750	630	0.0100	6.1E2
Control Rods	SIS-2352	14196	0.0040	3.5E3
	Inconel-X750	152.1	0.0100	1.2E2
	Boron Carbide	1757.6	-	5.9E-1
Moderator Tank	SIS-2352	21000	0.0040	6.0E3
Moderator Tank Lid	SIS-2352	39000	0.0040	3.0E1
Core Grid	SIS-2352	6000	0.0040	3.5E3
Core Spray	SIS-2333	9600	0.0040	6.8E1
Guide Tubes	SIS-2333	5070	0.0040	3.1E0
			Total:	7.2E4

a. Forsmark-3 and Oskarshamn-3

A final conclusion from their study was that the operational waste showed significant scatter but represented only 1-10% (0.2 -2 kBq/MW_{th}-h) of the base-line production in the reactor coolant.

Magnusson (2008) reported on the distribution of ¹⁴C in the various waste streams at Oskarshamn 3 (O3), Forsmark 3 (F3), and Ringhals 1 (R1) for 2002-2006. Some of their results are summarized in Table 3-16.

Table 3-16

14C Production and Release Pathways for Three ASEA-ATOM BWRs

	BWR-NWC (O3) ASEA-ATOM Internal Pumps	BWR-NWC (F3) ASEA-ATOM Internal Pumps	BWR-HWC (R1) ASEA-ATOM External Pumps	
Basic Data				
Thermal Power, MWth	3,300	3,300	2,500	
Energy Production ^a , TW _e h/yr	9.17	9.65	6.24	
Calc. ¹⁴ C Production ^a , Bq/yr (Ci/yr)	6.15E11 (16.6)	6.43E11 (17.4)	4.11E11 (11.1)	
	Percent of Calculated Production			
Gaseous Waste				
Stack Release ^b	63	128	118	
Accumulated in off-gas system	2	2		
Solid Waste				
Spent Resins	0.77	0.77	0.55	
Liquid Waste				
Waste Water Tanks	0.04	0.04	0.04	
Total:	66	131	119	

a. Average value over the years 2002-2006. b. Stack releases routinely measured.

3.4.7 ¹⁴CO₂ Adsorption on Charcoal

Magnusson and Stenström (2005) measured the concentration of ¹⁴C in the Oskarshamn Unit 3 (NWC) off-gas system before and after the charcoal delay beds. The results of these analyses are reported in Table 3-17.

Table 3-17 Concentration of ¹⁴C in Off-Gas System at Oskarshamn Unit 3 (NWC) (Magnusson and Stenström (2005))

Sample Location	Organic µCi/cc	Organic Fraction, %	Total, μCi/cc
Before Charcoal Delay Beds	5.21E-7	0.6	8.68E-5
After Charcoal Delay Beds	2.88E-7	2.2	1.31E-5

The transport of ¹⁴C through the charcoal beds is compared to the calculate source term in Table 3-18.

Table 3-18
Transport of ¹⁴C Through the Off-Gas System at Oskarshamn Unit 3 (NWC) (Magnusson and Stenström (2005))

	Calculated Production Rate	Inlet – Charcoal Delay Beds	Outlet- Charcoal Delay Beds	Retention	
Species	Pr	Production or Transport, μCi/s			
Inorganic		0.557	0.0784	86	
Organic		0.0031	0.0018	43	
Sum	0.595	0.559	0.0802	86	

The mass balance between the calculated production rate, removal by solid waste and the inlet source term to the charcoal delay beds is very good, and it is evident that there is retention of ¹⁴C species on the carbon beds. The charcoal beds are operated in a mode with regular back flushing, and it difficult to compare the outflow from the beds with the calculated production rate.

Siriwardane, et al. (2001) performed gas adsorption studies (15% CO_2 , 82% N_2 , 3% O_2 and water vapor) on activated carbon at 25°C over an applied pressure range of 0 to 300 psig. CO_2 adsorption at 1 atm total pressure of the gas mixture yielded a value of ~1.5 mole CO_2 adsorbed per kilogram of activated carbon.

In the charcoal beds of a BWR, the ambient CO_2 concentration is $\sim 0.035\%$ and the estimated adsorption value is 3.5E-3 mole CO_2 /kg C (0.078 cc STP/g). A BWR releasing 15 Ci 14 C /yr as CO_2 via the off-gas system with an air in-leakage of 25 SCFM will have a 14 C concentration of $\sim 2.6E3~\mu$ Ci/mole CO_2 . One large domestic BWR has a guard bed containing 1280 lbs activated carbon and five beds each containing 29,600 lbs activated carbon each for a total of 149,280 lbs (67,712 kg). At equilibrium, these beds could have an inventory of about 0.6 Ci of 14 C or more should the beds be maintained at lower temperatures and/or at lower air-in-leakage.

Most of this ¹⁴CO₂ is not believed to be tightly held on the activated carbon. The delay time can be estimated using the relationship:

$$t_D = M \cdot K_d/F$$

where:

t_D = Delay time, minutes M = Mass of charcoal, g F = Flowrate, cc/min

K_d = Dynamic adsorption coefficient, cc/gram

 $t_D = 6.77E7 \text{ g} \cdot 0.078 \text{ cc/g} / (25 \text{ ft}^3/\text{min} \cdot 28.3E3 \text{ cc/ft}^3) = 7.5 \text{ min}$

The dynamic adsorption coefficient in the above equation is based on measurements at high pressure extrapolated to zero applied pressure, and the above discussion is presented as an example, but needs further investigation.

3.5 Effect of Chemistry on Gaseous Release Rate and Chemical Form

The vast majority of data on ¹⁴C transport in the BWR is for reactors operating on normal water chemistry (NWC). In this case, coolant oxygen concentrations are elevated throughout the recirculation system and in the core. As a result, the electrochemical potential (ECP) is in the range of+100 to +250 MV (SHE). To decrease the ECP to <-230 mV and mitigate stress corrosion cracking (SCC) of sensitized stainless steels, hydrogen is injected into the feedwater to decrease the reactor water oxygen concentration. This mode of operation is referred to as hydrogen water chemistry (HWC).

Since main steam line radiation levels increase as much as a factor of 5 due to increased ¹⁶N steam transport when sufficient hydrogen is injected to mitigate IGSCC in the bottom head region and in the shroud, General Electric has developed several processes for depositing noble metals on system surfaces. In the presence of a sufficient noble metal surface concentration, the hydrogen injection rate can be reduced by a factor of 5 to 10 since the hydrogen/oxygen reaction is catalyzed by the noble metal and a local hydrogen/oxygen molar ratio only slightly above two is required to reduce the local ECP to the range of -500 to -700 mV. These processes, NMCA (Noble Metal Chemical Addition) or OLNC (On-Line Noble Chemistry), have been applied at most US BWRs. Following application of one of these processes, the mode of operation is referred to as HWC-NMCA. No data have been published on the impact of NMCA on the production and chemical distribution of ¹⁴C species.

Note that during HWC or HWC-NMCA operation, the coolant remains oxidizing in upper regions of the core and in the separator/dryer region. As a result, the form of ¹⁴C transported from the vessel via the steam is expected to remain in primarily a highly oxidized state.

The chemical form exiting the core and remaining in the reactor coolant may be in a more reduced state and more ¹⁴C may be collected on the RWCU resins with a corresponding decrease in steam transport. However, the chemical forms exiting the off-gas treatment system are not expected to change due to the oxidizing environment in the off-gas system hydrogen recombiner.

Since increased shutdown dose rates can occur with adoption of HWC or HWC-NMCA, zinc is injected into the feedwater to increase the reactor water zinc concentration. As a result, general corrosion rates of system materials are decreased. In addition, zinc competes with ⁶⁰Co for incorporation into surface oxides. Both effects lead to decreases in the shutdown dose rates. No impact is expected on the distribution of carbon chemical species as a consequence of zinc addition to the feedwater of the BWR, since the presence of zinc does not impact on coolant oxidant concentrations and the change in reactor coolant pH_T is insignificant.

3.6 Effect of Fuel Failures

The 14 C production rate in the fuel is approximately 25 Ci/GWe-yr (see Table 3-1). With approximately 50,000 fuel pins in the reactor core, a fuel pin will have an inventory of approximately 1000 μ Ci of 14 C after two GW_e-yrs of operation. A fuel defect instantly releasing 10% of this inventory (a very high estimate of the release) into the reactor coolant would increase the 14 C concentration by approximately 3E-7 μ Ci/g. A steady state release of 10% of the production rate in a single defective fuel rod would be \sim 2E-6 μ Ci/sec. Either input is a small fraction of the 14 C production rate in the reactor coolant and can be neglected as a source term during normal operation.

3.7 Summary of BWR Transport and Release

Calculated values of 14 C generation rates and observed release rates via gaseous release pathways are summarized in Table 3-19 and 3-20, respectively. Values are shown in the units reported by each researcher and have also been normalized to a standard release rate unit of Ci/GW_{th}-year. A thermal efficiency of 34% has been assumed to convert from MW_e to MW_{th}.

Table 3-19 Summary of Calculated ¹⁴C Generation Rates at BWRs^a

Reference	Unit	Ci/yr	Ci/GW _{th} -yr	Ci/GW _e -yr	GBq/GW _e -yr
NUREG-0016 (1979)	BWR-GALE Code	9.5			
Bonka (1974)	-		3.81	11.2	414
Kelly (1975)	1000 MW _e		5.44	16	592
ERDA-1535 (1975)	-			16	592
Fowler (1976)	3579 MW _{th} BWR/6		3.1	9.2	
Hayes (1977)	1000 MW _e		3.91	11.5	426
Davis (1977)	-			>8 but <16	296-592
Vance (1995)	-		4.96-5.24	14.6-15.4	540-570
Lundgren (2002) ^b	Nordic BWRs		5.45-5.69	16.0-16.7	592-619
Magnusson (2008)	R1 (HWC) 2500 MW _{th}		5.30	15.6	577
	F3 (NWC) 3300 MW _{th}		5.37	15.8	585
	O3 (NWC) 3300 MW _{th}		5.41	15.9	588
This study (see Appendix B)	BWR, 3458 MW _{th}	13.7	3.95	11.6	430
	Average (1995-201	0 values):	5.1±0.6		

a. Bold is reported or calculated data. Italics are converted bold data assuming 34% thermal efficiency.

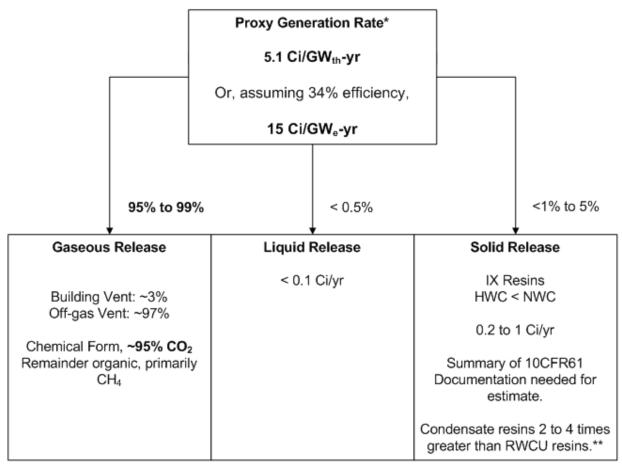
b. Lundgren's value: 23-24 kBq/MW_{th}-h.

Table 3-20 Summary of Observed Gaseous Release Rates at BWRs

		Gaseous Release Rates	
Reference	Unit	Ci/yr	Ci/GW _e -yr
Kunz (1976)	Nine Mile Point 1, 1850 MW _{th}	8	
Blanchard (1976)	Oyster Creek	13.3	
Fowler (1976)	3579 MW _{th} BWR/6 at 80% capacity		9.0
Evaluation of NUREG/CR-4245 (1985)	Brunswick, BWR/4 2436 MW _{th}	14.5	
Kunz (1985)	FitzPatrick, 850 MW _e		12.4
Magnusson (2008)	Oskarshamn 3, 3300 MW _{th}	10.8	
Magnusson (2008)	Forsmark 3, 3300 MW _{th}	22.6	
Magnusson (2008)	Ringhals 1, 2500 MW _{th}	13.1	

Based upon the observations and calculations summarized above and in Table 3-19, ¹⁴C transport in the BWR is schematically summarized below in Figure 3-1. This information can be used to estimate the amount of total generated carbon-14 that is released via gaseous effluent as carbon dioxide.

A proxy value for the BWR (5.1 ± 0.6 Ci/GW_{th}-yr) was developed based upon the average of the reported source term data over the time period of 1995 through 2010. This value can be used to estimate a carbon-14 source term for plants that do not have access to the data needed to calculate a site-specific source term.



^{*} A unit specific calculation can be conducted per the guidance provided in Section 3.3 or the proxy value provided can be used to estimate the generation of 14 C. The uncertainty on the proxy value is $\pm 15\%$.

Figure 3-1

14C Transport in the BWR

^{**}The RWCU ion exchange resins are expected to contain ¹⁴C as carbonate/bicarbonate in the inorganic form and formate in the organic form. The condensate resin is expected to be primarily in the carbonate/bicarbonate form.

4

CARBON-14 GENERATION AND RELEASE IN PWR SYSTEMS

4.1 Overview of ¹⁴C in the PWR

Carbon-14 is produced in the reactor coolant during power operation, and its production rate increases during the fuel cycle due to increasing neutron flux and ingress of nitrogen. Since the PWR operates with a reducing chemistry, most, if not all, of the ¹⁴C species initially produced are organic and contain only a single carbon atom. Possible species include methane (¹⁴CH₄), methanol (¹⁴CH₃OH), formaldehyde (H₂¹⁴C=O or the *gem*-diol H₂¹⁴C(OH)₂ (Dong and Dasgupta, 1988)), and formic acid (H¹⁴COOH). In theory, the only ionic species produced will be formic acid, and some or all of the formic acid will be removed by the letdown demineralizers. Formaldehyde is soluble in water and may partially be chemisorbed on the ion exchange resin. A quasi-equilibrium is established in the coolant between the initially produced species and other possible species in the reactor coolant.

The most chemically reduced species and probably the most prevalent species is $^{14}\text{CH}_4$ which partitions itself between the reactor coolant liquid and gas phases in the VCT and pressurizer. A calculation performed by the authors indicates that $\sim\!43\%$ of the methane is dissolved in the reactor coolant, $\sim\!50\%$ is present in the VCT vapor space and $\sim\!7\%$ in the pressurizer vapor space.

Analyses of pressurized PWR reactor coolant samples shows that the 14 C species are essentially 100% organic, and \sim 50% of the coolant activity is a volatile species (most likley methane).

The 14 C production rate in a large 4-loop Westinghouse PWR is $\sim 20~\mu$ Ci/min (see Section 4.3.4.1). However, the concentration of 14 C in the primary coolant rarely builds up to greater than 8E-4 μ Ci/g (coolant inventory of $\sim 185~m$ Ci). The total (gas phase plus liquid phase) concentration of 14 C measured in the Ringhals 4 RCS (Magnusson, et al. 2005) RCS was approximately 8E-4 μ Ci/g of which $\sim 50\%$ was in the gas phase. In general, lower concentrations (1-5 E-4 μ Ci/g) have been reported and may be a result of not obtaining a representative sample for analysis. In the absence of release or removal pathways, the 14 C in the reactor coolant would build up to 8E-4 μ Ci/g in less than two weeks of operation. The release and removal pathways from the primary coolant include VCT venting, boron dilution, inventory buildup on the letdown demineralizers and filters and known and unknown reactor coolant leakage. At some point a quasi, although not steady state, equilibrium is reached. Letdown system removal is approximately 1 Ci/yr. At a letdown flow rate of 75 gpm this removal represents a removal rate of approximately 1.9 μ Ci/min leaving another $\sim 18~\mu$ Ci/min being released from the RCS by other loss mechanisms.

The source term must balance the system inventory, the gaseous release, the liquid release and the solid release. The following type of mass balance is considered reasonable for a PWR.

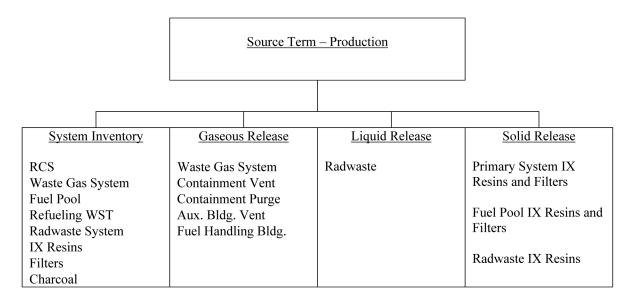


Figure 4-1 Carbon-14 Mass Balance for PWR

The ¹⁴C source term can be estimated using the calculation methodology presented below. The basis of this methodology is presented in Appendix C. Appendix D provides site-specific calculations for a number of domestic PWRs (2 Combustion Engineering (CE) units and 7 Westinghouse (W) units). These sites provided neutron flux and reactor coolant mass data for evaluation. Unfortunately, data were not available for the B&W reactor source term calculation.

Carbon-14 is pervasive in the PWR systems, as is tritium, and any location or system that contains tritium most likely also will contain ¹⁴C in some chemical form. Measurements of ¹⁴C concentrations in the various liquid systems have been performed, and some of the reported data are included in this report. As a general rule, ¹⁴C in the primary coolant is essentially all organic with a fair fraction as a gaseous species. Any time the RCS liquid or gas is exposed to an oxidizing environment, a slow transformation from an organic to an inorganic chemical form can occur. This is especially significant during the forced oxidation during the shutdown evolution and during refueling outages.

Dissolved nitrogen gas and ammonia in the RCS will contribute to the ¹⁴C source term. The dissolved nitrogen can become significant in the latter stages of the fuel cycle due to the introduction of increased quantities of non-borated water for boron dilution. Continuous venting of the VCT can reduce the RCS ¹⁴C concentration of gaseous activity and the impact of dissolved nitrogen during the latter phases of the fuel cycle (see Appendix E).

Activation of the nitrogen gas in the PWR containment is only a very minor contributor to the ¹⁴C source term due to the low neutron flux in containment.

4.2 ¹⁴C Source Term Estimation and Measurement

In the PWR-GALE Code, (NUREG-0017 (1985)), "The annual quantity of ¹⁴C released from a pressurized water reactor is 7.3 Ci/yr. It is assumed that most of the ¹⁴C will form volatile compounds that will be released from the waste gas processing system and from the containment and auxiliary building atmospheres to the environment."

The database for the NUREG-0017 release estimate of 7.3 Ci/yr is presented in Table 4-1. The basis for the distribution of ¹⁴C release in the gaseous effluents is presented in Table 4-2. No explanation has been found by the authors for the very high releases from Connecticut Yankee or the very low releases from Yankee Rowe.

Table 4-1 Carbon-14 Release Data from Operating PWR's (NUREG-0017, Rev. 1)

	Year					
Plant*	1975	1976	1977	1978	Ci/Yr-Unit	
Conn. Yankee	44	40	30	70	46	
Yankee Rowe	1.6	0.13	0.24	0.33	0.58	
Plant**		Α	rea		Ci/Yr-Unit	
Turkey Point 3/4	Auxiliary Bu	uilding			2.4	
(NUREG/CR-1629)	Containme	nt			0.075	
	Waste Gas	Processing Sy	stem		0.82	
	Spent Fuel	Area			0.38	
	•			Total:	3.7	
Fort Calhoun	Fuel Pool a	ınd Auxiliary Bu	ıilding		0.30	
(NUREG/CR-0140)	Waste Gas	Processing Sy	stem		0.81	
	Containme	nt Building***			0.78	
	•	-		Total:	1.9	
Zion Units 1 & 2	Containme	nt Building			1.8	
(NUREG/CR-0715)	Fuel Handli	ing and Auxilia	y Building		1.4	
	Waste Gas	Processing Sy	stem		0.062	
	<u>.</u>			Total:	3.3	
Prairie Island 1/2	Containme	nt Building			0.016	
(NUREG/CR-4397)	Fuel Handli	ing and Auxilia	y Building		3.3	
Waste Gas Processing S	System				0.25	
-				Total:	3.6	
Rancho Seco	Containme	Containment Building				
(NUREG/CR-2348)	Fuel Handli	ing and Auxilia		1.85		
	Waste Gas	Processing Sy	stem		0.85	
	•			Total:	3.6	
				Average:	7.3	

^{* -} Based on semi-annual release reports.

^{** -} Based on in-plant source term measurements.

^{*** -} NUREG CR-0140 states that the sampler organic oxidizer was inoperable and the containment measurements could be low by a factor of 10.

Table 4-2
Distribution of Carbon-14 Released in Gaseous Effluents (NUREG-0017, Rev. 1)

Plant	Plant Areas: Containment	Auxiliary Building and Fuel Handling	Waste Gas Processing System
Turkey Point 3 & 4	2%	75%	23%
Fort Calhoun	41%	16%	43%
Zion 1 & 2	55%	43%	2%
Rancho Seco	25%	51%	24%
Prairie Island 1/2	0.5%	92.5%	7%
Average:	22.6%	61.0%	16.4%

NUREG-0017 (1985)) indicates that "¹⁴C reacts to form volatile compounds (principally CH₄. C₂H₆ and CO₂) that are collected in the waste gas processing system through degassing of the primary coolant and released to the environment via the plant vent". It also indicates that "¹⁴C is released from the containment and auxiliary building vent as a result of leakage of primary coolant into the containment and auxiliary building atmospheres".

Fowler, et al., (1976) calculated the production rate of ¹⁴C for a reference PWR and compared their results with calculations made by others (Table 4-3). They considered activation only by thermal neutrons but adjusted the 2200 m/s cross-section values by a factor of 0.6 to correct for the temperature dependence of the thermal neutron cross-section and the thermal neutron spectrum. The mass of coolant in the reactor flux of his reference PWR (3473 MW_t Combustion Engineering) was 1.37E4 kg.

Table 4-3
Calculated Production of Carbon-14 in PWRs (Fowler, et al., (1976))

		Carbon-14 Production Rate (Ci/GW _e -yr)					
	Target	Fowler, et al. (1976)	Bonka, et al. (1974)	Hayes, et al. (1977)ª	ERDA-1535 (1975)	Kelly, et al. (1975)ª	
	O-17	4	7.1	4.0		2.7	
PWR Fuel	N-14	18	12.2	7.6		10.9	
	Total	22	19.3	11.6	17 ^b	13.6	
PWR	O-17	3.2	9.8	3.3			
Coolant	N-14	0.09	1.3	0.1			
Coolailt	Total	3.3	11.1	3.4	6	6	
PWR Sum		25	30.4	15	23	19.6	

a. The production rates presented by Hayes et al. (1977) and Kelly et al. (1975) for a 1000 MW_{th} PWR were multiplied by 3.03 (33% thermal efficiency) to convert their values to a per GW_e -yr basis.

b. Fuel and cladding production rates for ERDA-1535 (1975) were added and identified as a fuel production rate in this table.

Davis (1977) calculated the quantities of 14 C formed in the fuel, core structural materials, and coolant in light-water cooled reactors, high temperature gas-cooled reactors and liquid-metal cooled fast breeder reactors. The calculated value for the 17 O(n, α) 14 C reaction in the primary coolant of a PWR was 5.0 Ci/GW_e-yr.

Vance et al. (1995) indicated that previously reported production rates ranged from 270-410 GBq/GW_e-yr (7.3-11.1 Ci/GW_e-yr) for PWRs. Vance's source term calculations for the PWR are summarized in Table 4-4. He noted there are significant uncertainties associated with these production rates, principally in the values for the equivalent cross sections.

Table 4-4
Calculated Production Rates of Carbon-14 in PWR Reactors (Vance, 1995)

Reaction	Production Rate ^b
¹⁷ O(n,α) ¹⁴ C	6.0 Ci/GW(e)-yr
¹⁴ N(n,p) ¹⁴ C	0.12 Ci/GW(e)-yr/ppm
¹³ C(n,γ) ¹⁴ C	0.011 Ci/GW(e)-yr/ppm

- a. Effective cross-section for the $^{17}O(n,\alpha)^{14}C$ reaction: 0.183 b. Effective cross-section for the $^{14}N(n,p)^{14}C$ reaction: 1.17 b. Effective cross-section for the $^{13}C(n,\gamma)^{14}C$ reaction: 0.006 b.
- b. Thermal flux: 4.8E13 n/cm²-sec, PWR coolant mass exposed to flux: 13,400 kg.

Magnusson (2008) performed an extensive program to characterize ¹⁴C in Swedish light water reactors. The work involved the development of ¹⁴C analysis techniques, mapping of ¹⁴C in waste streams and measurements of ¹⁴C in environmental samples in the vicinities of several Swedish reactors.

The calculated core specific coolant production rate for a Swedish PWR is summarized in Table 4-5. The thermal neutron flux was calculated and considered enrichment and burn-up for an equilibrium core. The neutron spectrum for higher energies was based on in-core fuel management calculations. Their calculation indicated a production rate of 8.8 Ci/yr for this 2775 MW_{th} Westinghouse PWR.

Table 4-5
Calculated Production Rates of ¹⁴C in the Reactor Coolant of a 2775 MW_{th} Westinghouse PWR (Magnusson (2008))

	Production Rate ^a (Bq/s)				
Target	Thermal Epithermal Fission				
¹⁷ O	3.4E3	1.1E3	5.8E3		
¹⁴ N ^b	47	16	4.0		

- a. Production rate corresponds to 350 GBq/GW $_e$ -yr (9.5 Ci/GW $_e$ -yr). The overall uncertainty in the calculated production rate was estimated by the authors to be $\pm 20\%$.
- b. Assumes a nitrogen concentration of 5.4 ppm in the reactor coolant.

4.3 Site Specific PWR Source Term Estimation

This section provides guidance for estimating carbon-14 source term based on unit specific reactor core physics and reactor design. In order to calculate the unit specific carbon-14 generation, each plant will need their best estimate of their reactor neutron flux profiles (2 or 3 energy groups), mass of coolant in the "active core", and concentration of nitrogen. With this information, the generation of carbon-14 from oxygen-17 and nitrogen-14 can be calculated and summed for the total carbon-14 production rate.

Given a constant neutron flux and target concentration, the rate of production of a species, N_a ,, in atoms per second is given by:

$$N_a = N_T \cdot \Sigma(\sigma_i \cdot \phi_i)$$
, $i = 1$ to 2 or 3^a
a. In some cases there may be only 2 energy groups,

 $E \le 0.625 \text{ eV} \text{ and } E > 0.625 \text{ eV}$

where:

N_a = Rate of production, atoms/sec

N_T = Number of target ¹⁷O or ¹⁴N target species per kg of coolant

 σ_i = "effective" neutron cross-section for each of the 2 or 3 energy groups, cm²

 φ_i = neutron flux for each of the 2 or 3 energy groups, neutron/cm²-sec

The source term of each species A_i, d/s-sec, or Bq/sec is given by:

$$A_a = N_a \cdot \lambda_a$$

Where λ_a is the decay constant of the species.

The source term in µCi/sec is given by:

$$A_a (\mu \text{Ci/sec}) = N_a \cdot \lambda_a / 3.7\text{E4 d/sec-}\mu \text{Ci}$$

The following methodology for estimating a site specific PWR ¹⁴C source term is recommended for consideration and was used in Appendix D to calculate the source terms for a number of domestic PWRs.

4.3.1 STEP 1: Unit Specific Neutron Flux

Develop values for core "average" neutron flux at the beginning of the cycle (BOC), mid-cycle and end of cycle (EOC) for three energy groups. For illustration purposes, the following flux data are used:

Table 4-6
Example of Core Average Neutron Flux over PWR Fuel Cycle

BOC					
Neutron Group	Group Energy	Neutron Flux (φ), n/cm ² -sec			
Thermal	≤0.625 eV	3.55E13			
Intermediate	>0.625 eV - <1 MeV	2.86E14			
Fast	≥1 MeV	6.57E13			
Mid-Cycle					
Neutron Group	Group Energy	Neutron Flux (φ), n/cm²-sec			
Thermal	≤0.625 eV	3.57E13			
Intermediate	>0.625 eV - <1 MeV	2.24E14			
Fast	≥1 MeV 6.54E13				
EOC					
Neutron Group	Group Energy	Neutron Flux (φ), n/cm²-sec			
Thermal	≤0.625 eV	3.97E13			
Intermediate	>0.625 eV - <1 MeV	2.27E14			
Fast	≥1 MeV	6.68E13			

Note that not all PWRs have a core average three group neutron flux distribution readily available for this calculation. Several sites noted in Appendix D only had a two group neutron flux distribution; \leq 0.625 eV and >0.625 eV, the >0.625 eV being the sum of the intermediate and fast flux.

4.3.2 STEP 2: Effective Cross-Sections and 14 C Generation Rate

Use the "Effective Cross-Sections" in the two or three neutron flux distributions to calculate the source term for the two major production reactions in units of μ Ci/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction (Table 4-7) and μ Ci/sec-kg-ppm N for the $^{14}N(n,p)^{14}C$ reaction (Table 4-8). The methodology for determining the values of "effective cross-section" employed in this section is presented in Appendix C.

4.3.2.1 Production Rate of ¹⁴C from ¹⁷O(n,α)¹⁴C Reaction

For the $^{17}O(n,\alpha)^{14}C$ reaction, the "effective" cross-sections in the four neutron energy groups are shown in Table 4-7.

Table 4-7 "Effective" Cross-Section for the $^{17}O(n,\alpha)^{14}C$ Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", Barns
Thermal	≤0.625 eV	0.121
Intermediate (I)	>0.625 eV - <1 MeV	0.0291
Fast (F)	≥1 MeV	0.1124
I+F	>0.625 eV	0.0479

The 14 C production rate from the 17 O(n, α) 14 C reaction is calculated for the three group flux distribution as follows:

$$Production\ Rate\ (\mu Ci/sec - kg) = \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i} \bullet \phi_{i} + \sigma_{f} \bullet \phi_{f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

1.27E22 atoms $^{17}O/kg$ H_2O N "effective" thermal cross-section, b σ_{th} thermal neutron flux, n/cm²-sec ϕ_{th} "effective" intermediate cross-section, b Intermediate neutron flux, n/cm²-sec ϕ_i "effective" fast cross-section, b $\sigma_{\rm f}$ fast neutron flux, n/cm²-sec ϕ_f Conversion factor, 1.0E-24 cm²/b 1.0E-24 ¹⁴C decay constant, 3.833E-12/sec Conversion factor, 3.7E4 d/sec-µCi 3.7E4

The calculated ¹⁴C production rate using the above data is reported in Table 4-8.

Table 4-8
PWR ¹⁴C Production Rate from the ¹⁷O(n,α)¹⁴C Reaction for Example Plant

Production Rate, μCi/sec-kg	
BOC	2.544E-5
Mid-Cycle	2.392E-5
EOC	2.489E-5
Average:	2.475E-5

4.3.2.2 Production Rate of ¹⁴C from ¹⁴N(n,p)¹⁴C Reaction:

For the ¹⁴N(n,p)¹⁴C reaction the "effective" cross-sections in the four neutron energy groups are shown in Table 4

Table 4-9 "Effective Cross-Sections" for the ¹⁴N(n,p)¹⁴C Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", Barns
Thermal	≤0.625 eV	0.951
Intermediate (I)	>0.625 eV - <1 MeV	0.0379
Fast (F)	≥1 MeV	0.0436
I+F	>0.625 eV	0.0392

Use the following equation to calculate the production rate for the $^{14}N(n,p)^{14}C$ reaction:

Production Rate (
$$\mu$$
Ci/sec - kg - ppm - N) =
$$\frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i} \bullet \phi_{i} + \sigma_{f} \bullet \phi_{f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

4.284E19 atoms ¹⁴N/kg-ppm N N "effective" thermal cross-section, b σ_{th} thermal neutron flux, n/cm²-sec ϕ_{th} "effective" intermediate cross-section, b σ_i Intermediate neutron flux, n/cm²-sec ϕ_i "effective" fast cross-section, b = $\sigma_{\rm f}$ fast neutron flux, n/cm²-sec ϕ_f conversion factor, 1.0E-24 cm²/b 1.0E-24 ¹⁴C decay constant, 3.833E-12/sec λ conversion factor, 3.7E4 d/sec-µCi 3.7E4 =

The calculated production rate using the above data is reported in Table 4-10.

Table 4-10 PWR ¹⁴C Production Rate from the ¹⁴N(n,p) ¹⁴C Reaction for Example Plant

Production Rate, μCi/sec-kg-ppm I			
BOC	2.101E-7		
Mid-Cycle	2.011E-7		
EOC	2.188E-7		
Average:	2.100E-7		

4.3.3 STEP 3: Unit Specific Coolant Mass

Determine the mass of coolant in the "active" core in kilograms H_2O (kg H_2O). It is suggested that utilities use the mass of reactor coolant in the fuel cells from the "bottom" of the active core to the "top" of active core. The volume of the coolant in the "active core" is unique to the fuel vendor's design. The core average temperature and pressure should be used to convert the liquid volume to mass. This mass is used to calculate the ^{14}C source term. In this example, a 14,100 kg value is assumed.

4.3.4 STEP 4: 14 C Source Term

Calculate the ¹⁴C source term based on the above generation rates considering plant specific values of power, mass of liquid in the "active" core and coolant nitrogen concentration.

For illustration purposes, consider a 1178 MW_e (\sim 3549 MW_{th}) Westinghouse PWR with an estimated coolant mass in the core flux of 14,100 kg and 1.0 ppm nitrogen in the reactor coolant.

4.3.4.1 ¹⁴C Source Term from 17O(n, α) ¹⁴C Reaction

The production rate for this reaction would be:

```
\begin{array}{l} 2.475 \hbox{E-5 $\mu$Ci/sec-kg} \bullet 14{,}100 \ kg &= 0.349 \ \mu$Ci/sec \\ &= 11.01 \ Ci/yr \\ &= 9.35 \ Ci/GW_e\mbox{-yr} \\ &= 3.10 \ Ci/GW_{th}\mbox{-yr} \\ &= 0.354 \ \mu$Ci/MW_{th}\mbox{-h} \\ &= 13.1 \ kBq/MW_{th}\mbox{-h} \\ &= 346 \ GBq/GW_e\mbox{-yr} \end{array}
```

It has been the general practice to express the production rates in Ci/GW_e -yr. However, a less ambiguous unit would be Ci/GW_{th} -yr. In any conversion between units, use gross MW_e .

4.3.4.2 ¹⁴C Source Term from ¹⁴N(n,p)¹⁴C Reaction

The production rate for this reaction at 1.0 ppm nitrogen would be:

```
1.0 ppm N • 2.100E-7 \muCi/sec-kg-ppm N • 14,100 kg = 2.961E-3 \muCi/sec
```

Guidance for calculating nitrogen concentrations in reactor coolant is provided in Appendix E. If it is assumed that this reactor operated with $\sim 12 \% N_2$ in the VCT and 0.53 ppm NH₃, the annual production rate would be ~ 0.5 Ci/yr from this nuclear reaction pathway.

4.3.5 Summary of 14 C Source Term Calculations

In conclusion, the carbon-14 source term is the sum of the production rates from oxygen-17 and nitrogen-14. Calculations of these production rates require knowledge of in-core coolant mass, the two or three energy groups neutron flux distribution, and the "effective" neutron cross-sections provided in this report.

4.4 ¹⁴C Release Pathways and Chemical Form

Kahn, et al. (1974) made measurements of the release from the gas waste disposal system at Haddam Neck (Connecticut Yankee). Their results revealed that virtually all of the ¹⁴C release was in a non-CO₂ form.

Kunz, et al., 1974 measured the chemical form of ¹⁴C in the decay tank gas and containment air at three operating PWRs. Their results are given in Table 4-11. The results for the gas decay tanks are the averages from several analyses while the results for containment air are for single samples.

Table 4-11
Percentage of the Total Gaseous ¹⁴C Activity Detected in Various Compounds

		Decay Tank	Containment		
Compound	Ginna	Indian Point 1	Indian Point 2	Ginna	Indian Point 2
CH ₄	66.9	74.3	84.9	57.0	58.6
C ₂ H ₆	22.4	7.5	3.5	37.5	36.0
C ₃ H ₈	2.4	7.2	2.7	3.9	2.3
C ₄ H ₁₀	0.9	10.0	2.7	n.d.	0.5
CO ₂	4.6	0.3	3.4	1.8	2.6
СО	0.4	n.d.	0.3	n.d.	n.d.

n.d. - not detected

Over 80% of the total gaseous 14 C release was in the form of low molecular weight hydrocarbons (CH₄ and C₂H₆). The CO₂ and CO fraction was less than 5%. From the above data and concentration measurements, Kunz, et al., (1974) calculated a gaseous release of ~6 Ci/GW_e-yr. They further commented that people living 1 km from the site could potentially double their 14 C body burden if all of the gaseous releases were as CO₂. Since less than 5% of the releases were as CO₂ or CO, and the releases were not at ground level, the actual increase in the 14 C body burden would be considerably less.

Table 4-12 PWR 14 C Source Terms (Fowler, et al. (1976) (Westinghouse Design, Nominal 1250 MW $_{\rm e}$ at 80% Capacity Factor)

Source	Annual ¹⁴ C Discharge Rate (Ci/yr) or (Ci/GW _e -y)		
GASEOUS SOURCE TERMS			% of Total
Gaseous Waste Disposal System		3.8	74.5
Condenser Air Ejector Off-gas		0.11	2.2
Steam Generator Blowdown Tank Vent		4.5E-4	0.0088
Turbine Gland Seal		9.2E-7	0.000018
Fuel Handling Building Ventilation		0.69	13.5
Containment Purge		0.52	10.2
Auxiliary Building Ventilation		8.0E-4	0.016
Turbine Building Ventilation		8.7E-6	0.00017
	Total:	5.1	
LIQUID SOURCE TERMS			
CVCS (Boron Recycle System)		2.7E-3	
Liquid Waste Disposal System		1.9E-3	
Steam Generator Blowdown		8.3E-4	
Turbine Drains		1.3E-5	
	Total:	5.4E-3	

Kunz (1985) measured total ¹⁴C release and chemical form of ¹⁴C at the 490 MW_e R. E. Ginna PWR and the 1,000 MW_e Indian Point Unit 3 PWR. Results are provided in Table 4-13.

Table 4-13 ¹⁴C Gaseous Release Rate, Chemical Form and Discharge Pathways at Ginna and Indian Point 3 (Kunz, 1985)

Parameter	R. E. Ginna	Indian Point 3
Total Gaseous Release Rate, Ci/GW(e)-yr	11.6	9.6
Chemical Form		
¹⁴ CO ₂	10%	26%
¹⁴ CH ₄ , ¹⁴ C ₂ H ₆ , etc.	90%	74%
Discharge Pathway		
Gas Decay Tanks	42%	7%
Containment Venting	23%	78%
Auxiliary Building Venting	35%	15%

4.4.1 Ginna

At Ginna, samplers for total ¹⁴C were placed on the main plant vent and the containment vent. When the containment was not being vented, the containment vent sampler was set up to measure only ¹⁴CO₂ at the plant vent location.

The average 14 C concentration in the nine decay tank samples collected between 1973 and 1981 was 1E-3 μ Ci/cm³. The decay tanks were vented 31 times during the 88 week test period. The annual release was ~1.5 Ci/y. The chemical composition of 14 C in the decay tanks was 74% 14 CH₄, 16% 14 C₂H₆, 6% 14 C₃H₈ and 14 C₄H₁₀ and 4% 14 CO₂.

Sampling of the reactor coolant for 14 C indicated that there was no detectable removal by the letdown demineralizers. Four samples of reactor coolant ranged in concentration from 0.78E-4 to 1.3E-4 μ Ci/ml with an average of 1.1E-4 μ Ci/ml. Approximately 0.008 Ci/yr was released to containment via a primary coolant leak.

A waste water composite sample also was analyzed for total 14 C. The concentration was 5.1E-7 μ Ci/ml. If it is assumed that all of the 14 C was collected in the evaporator bottoms, a total of 0.002 Ci/yr would be shipped to the burial site.

4.4.2 Indian Point Unit 3

At Indian Point Unit-3, all gases were discharged through the plant vent. Continuous samplers were used to measure total 14 C and 14 CO₂ during a 98-week period that started in early August 1980. The total gaseous 14 C release was estimated to be 9.6 Ci/GW(e)-yr while the release rate for 14 CO₂ was estimated at 2.5 Ci/GW(e)-yr, or 26% of the total gaseous 14 C release. The 14 CO₂ release rate was higher when the plant was not operating.

Indian Point 3 results of grab sample analysis of gaseous decay tanks, containment air and the plant vent when neither the decay tanks or containment were being vented are shown in Table 4-14.

Table 4-14
Percentage of Various Compounds in ¹⁴C Activity Detected in Release Pathways at Indian Point Unit-3

	% of ¹⁴ C Chemical Compounds					
¹⁴C Compound	Decay Tank ^a Containment Air ^b Plant Vent ^c					
CH₄	62	60	46			
C ₂ H ₆ , C ₃ H ₈ and C ₄ H ₁₀	29	32	20			
CO ₂	9	8	34			

- a. Average of four samples collected between 1976 and 1982.
- b. Average of three samples collected between 1978 and 1982.
- c. Average of three samples collected while neither the gas decay tanks nor containment was being vented.

Sampling of the reactor coolant at Indian Point 3 for 14 C indicated that there was no detectable removal by the letdown demineralizers. Three samples of reactor coolant ranged in concentration from 5.6E-5 to 7.5E-5 μ Ci/ml with an average of 6.6E-5 μ Ci/ml. Approximately 0.07 Ci/yr was released to containment via a primary coolant leak.

4.4.3 Palisades and Big Rock Point

Martin (1986) sampled major waste streams at the Palisades PWR and the Big Rock Point BWR to determine the quantities of ¹⁴C in the waste streams. The largest amounts of ¹⁴C were found in the resins and filters used for RWCU. From these data the annual amounts of ¹⁴C in wastes from nominal PWRs and BWRs were estimated to be 4.7 and 0.5 Ci/GW(e)-yr, respectively.

4.4.4 Vance Studies

Vance, et al. (1995) reported measurements of reactor coolant ¹⁴C concentrations (Table 4-15). The data indicate that a significant fraction of the ¹⁴C species in the reactor coolant is organic. However, it is not clear whether pressurized samples were taken to retain the gaseous ¹⁴C species.

Table 4-15 Chemical Speciation of ¹⁴C in Reactor Primary Coolant Water Samples (Vance, et al., 1995)

Plant	Sample Date	Total 14C μCi/cc	Inorganic 14C μCi/cc	% Organic 14C
В	1/29/92	1.13E-4	1.65E-5	85.4
В	2/05/92	1.41E-4	7.51E-6	94.7
С	4/15/92	1.83E-4	2.80E-5	84.7
G	7/11/92	8.05E-5	3.40E-5	57.8

4.4.5 Nordic PWRs

Magnusson (2008) reported on ¹⁴C release rates in various waste streams at Ringhals 3 (R3) and Ringhals 4 (R4) for 2002-2006. Some results are summarized in Tables 4-16 and 4-17.

Table 4-16 Ringhals Unit 4 Process Water ¹⁴C Analysis, 2005

		Gas P	hase	Liquid Phase			
		Inorganic	Organic	Inorganic	Organic	Total	%
Syste	m		Activity	Concentratio	n, μCi/g		Organic
RCS before IX	Jun-27	2.43E-6	3.62E-4	1.59E-6	3.27E-4	6.93E-4	99.4
	Jun-27	2.92E-6	3.49E-4	9.73E-7	3.30E-4	6.83E-4	99.4
	Jul-5	3.08E-6	4.89E-4	1.49E-6	3.65E-4	8.59E-4	99.4
	Jul-7	1.11E-5	3.95E-4	4.32E-7	3.54E-4	7.61E-4	98.4
RCS after IX	Jun-27	4.97E-6	3.30E-4	4.78E-6	3.16E-4	6.56E-4	98.5
	Jul-5	4.46E-6	3.08E-4	4.32E-7	3.35E-4	6.48E-4	99.2
	Jul-5	<3.0E-8	4.89E-4	4.59E-8	3.54E-4	8.43E-4	100.0
SFP before IX	Jul-20			<9.2E-9	1.16E-7	<1.25E-7	>92.8
SFP after IX	Jul-20			1.62E-8	6.76E-8	8.38E-8	80.6
Waste Water Ta	nk			3.65E-6	1.51E-6	5.16E-6	29.4

Measurements on the release pathways (Table 4-17) are in reasonable agreement with the calculated source term.

Table 4-17 Carbon-14 Source Terms and Release Pathways at Ringhals Units 3 and 4 (Magnusson, et al. (2008))

	PWR (R3) Westinghouse 3 - Loop	PWR (R4) Westinghouse 3 - Loop
Basic Data		
Thermal Power, MW _{th}	2,775	2,775
Energy Production ^a , TW _e h/yr	7.35	7.24
Calc. ¹⁴ C Production ^a , Bq/yr	2.83E11	2.84E11
	Percent of Calcu	lated Production
Gaseous Waste		
Stack Release	86°	70 ^b
Solid Waste		
Spent Resins	9.8	9.0
Filter Cartridges	0.02	0.02
Deposited on Steam Generator Tubes	0.04	0.04
Liquid Waste		
Waste Water Tanks	0.26	0.26
Ejector Condensate		0.08
Accumulation in RWST	<0.02	<0.02
Accumulation in Spent Fuel Pool	0.01	0.01
Total:	97	80-87

a. Average value over the years 2002-2006.

Several conclusions based on the Table 4-16 measurements are as follows:

- The ¹⁴C chemical species in the reactor coolant are essentially all organic.
- Slightly over 50% of the organic ¹⁴C in the reactor coolant is in the gas phase.
- The inlet and outlet of the letdown demineralizers have similar ¹⁴C concentrations although there is evidence of some removal by the demineralizers.
- The spent fuel pool contains some inorganic ¹⁴C.

b. Minimum value. More correct value estimated to be 78%

c. Stack releases routinely measured.

d. Primary to secondary leak rate 7 kg/h (average for period measured)

The Ringhals stack release data (Figures 4-2 and 4-3) show that the ¹⁴C release rate normally increases during a fuel cycle and that a significant fraction of the total ¹⁴C release occurs during the refueling outage. The Unit-3 main stack release during the four year period of continuous monitoring averaged 88.2% organic, whereas, the organic fraction was 70.8% for Unit-4 for the same period. Also, for this period of observation, which includes releases during refueling outages, Unit-3 released an average 6.51 Ci/yr and Unit-4 released an average of 5.60 Ci/yr.

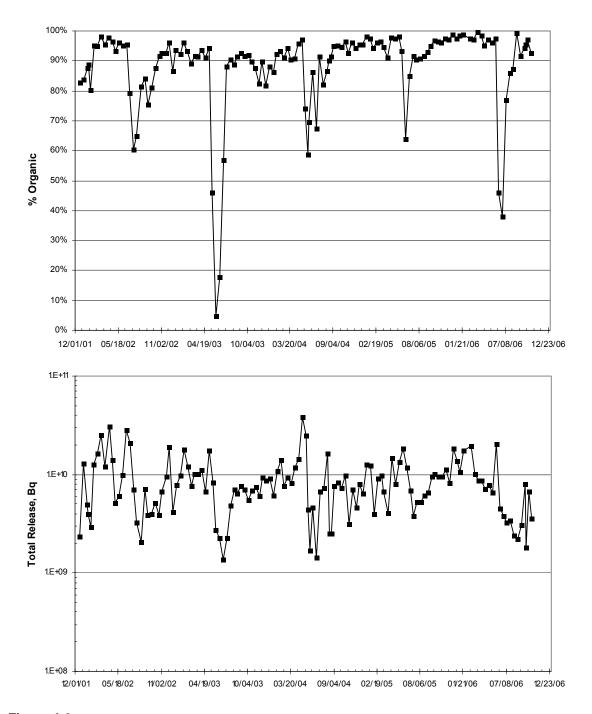
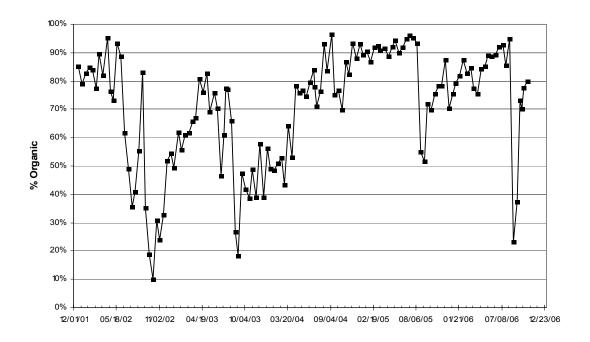


Figure 4-2 Ringhals Unit-3 Main Stack ¹⁴C Release During the Years 2002-2006 (Bengtsson, 2010)



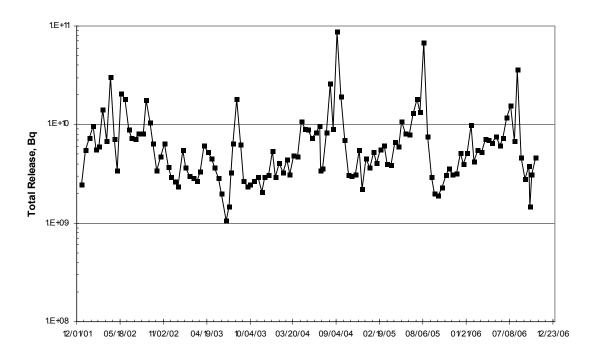


Figure 4-3 Ringhals Unit-4 Main Stack ¹⁴C Release During the Years 2002-2006 (Bengtsson, 2008)

4.4.6 Diablo Canyon Units 1 and 2

The concentrations of ¹⁴C in the gaseous waste and gaseous process streams at Diablo Canyon (Knemeyer, Wright and Cortina (2010)) were measured by GEL Laboratories, LLC (GEL) in April 2010. Results of these measurements are shown in Table 4-18.

Table 4-18
Diablo Canyon ¹⁴C Gaseous Sampling, April 2010

	¹⁴ CO ₂	¹⁴ CH ₄	Particulate	Ratio
	Activity Concentration, pCi/L			CH ₄ /CO ₂
Unit-1				
CTMT Atmosphere	39.60	440.40	0.00	11.1
Plant Vent	0.18	2.12	-0.01	11.8
Waste Gas Header	3,810	2,736,190	-	718.2
Unit-2				
CTMT Atmosphere	41.2	1,538.80	-0.02	37.3
Plant Vent	1.31	0.74	0.00	0.56
Waste Gas Header	4,700	435,300	-	92.6
Gas Decay Tank 2-1	20,600	1,839,400	-	89.3

With the exception of the Unit-2 plant vent, ¹⁴CH₄ was the dominant species.

Knemeyer, Wright and Cortina (2010) estimated the gaseous release per operating unit as shown in Table 4-19. This estimate was based on the April 2010 measurements and documented plant operational releases.

Table 4-19
Diablo Canyon Gaseous ¹⁴C Annual Discharge Per Operating Unit During Operation

Release Point	¹⁴C, Ci
Plant Vent	~5.4
Containment	~0.2
Gas Decay Tanks	~1.2
Total:	~6.8
	~1.9 as CO ₂

Sampling for ¹⁴C in various plant process streams was also performed in April 2010 with the samples sent to GEL. The results of the GEL analyses are shown in Table 4-20.

Table 4-20 Diablo Canyon ¹⁴C Liquid Sampling April 2010^a

	Unit-1	Unit-2
System	μC	i/ml
Reactor Coolant	2.24E-4	1.78E-4
Liquid Holdup Tanks	1.91E-4	-
Refueling Water Storage Tank	1.31E-6	2.26E-6
Spent Fuel Pool	2.78E-7	3.26E-7
Primary Water Storage Tank	3.93E-7	2.24E-7
Liquid Radwaste	1.36E-6	-

a. Samples shipped to GEL; full bottles, no preservation, no refrigeration.

A three year average of liquid radwaste release volume from both units is 1.5E6 gallons/year. The yearly site release is estimated at ~ 8 mCi of 14 C. To put this value in perspective, the total liquid activity discharged in 2009 of other nuclides was ~ 43 mCi. The liquid discharge of 14 C represents a small fraction of the ~ 6.8 Ci/yr of 14 C in the gaseous discharge.

The reactor coolant samples are likely lower than the actual concentration in the RCS since the samples were not taken under pressure and a major fraction of the ¹⁴C in the gaseous form in the RCS liquid probably escaped during sampling.

4.4.7 V. C. Summer

Roberts, (2010) reported on 14 C measurements at V. C. Summer, a 3-loop Westinghouse PWR with an output of 966 MW_e (2900 MW_{th}). The unit went on line on January 1, 1984 and is owned by South Carolina Electric & gas Company.

Measurements of ¹⁴C were made at the main plant vent utilizing the effluent monitor, at the reactor building vent using the process monitor, in the waste gas system and at an off-site control location. The results of these measurements are provided in Table 4-21.

Table 4-21 V. C. Summer ¹⁴C Sampling Results

	Total ¹⁴C	¹⁴ C as CO ₂	% as ¹⁴CO₂		
Sampled Location	Concentration, μCi/ml				
Main Plant Vent	1.39E-9	<3.78E-11	<2.7		
Reactor Building	7.32E-7	4.65E-8	6.4		
Waste Gas System	5.11E-3	3.2E-4	6.3		
Off-Site Control Location ^a	<7.64E-11				

a. Environmental laboratory approximately 2.6 miles from site.

4.4.8 Korean PWRs

Lee (2010) presented results from a multiyear measurement program of ¹⁴C at five Korean PWRs. Gaseous sampling was done with NaOH bubblers, using a catalytic converter for the conversion of non-CO₂ into CO₂. Typical sampling periods were 4 weeks. LSC counting of a BaCO₃ precipitate was utilized for quantification. The apparatus used for sampling is described in Section 5 of this report. His results are summarized in Table 4-22 and 4-23.

Lee (2010) indicated that year-to-year as well as plant-to-plant variations were so large that definitive trends could not be identified. The inorganic fraction of the gaseous release was much higher than expected. At several units, the release from the fuel building dominated the gaseous release.

Table 4-22 Gaseous Effluent Results from Five Korean PWRs (Lee (2010))

Reactor(s)	Sampling Dates	Sampling Location	Released Activity, Ci	Percent Organic
Kori Unit 1	7/13/06 - 12/28/06	Plant Stack	0.81	17.0
$W-600~MW_{\scriptscriptstyle e}$	12/28/06 - 8/1/07	Plant Stack	2.33	47.7
	1/31/08 — 11/13/08	Plant Stack	0.43	25.5
		Weighted Aver	rage % Organicª:	38.1
Kori Unit 3	9/15/06 - 12/28/06	Fuel Bldg.	1.06	0.2
$W-950~MW_{_{ m e}}$		Auxuliary Bldg.	0.10	39.0
		Radwaste Bldg.	0.58	80.1
	12/28/06 - 1/4/08	Fuel Bldg.	3.03	3.0
		Auxuliary Bldg.	0.33	29.1
		Radwaste Bldg.	1.05	60.4
		Containment Bldg.	0.04	96.7
	1/4/08 - 10/15/08	Fuel Bldg.	3.68	3.2
		Auxuliary Bldg.	0.74	51.9
		Radwaste Bldg.	1.65	72.2
		Containment Bldg.	0.42	85.0
		Weighted Average % Org	anicª:	27.0
Yonggwang	7/4/06 — 1/4/07	Fuel Bldg.	1.66	16.8
Unit 5		Primary Aux. Bldg.	0.22	72.5
CE - 1000 MW _e		Sec. Aux. Bldg. 0.55		73.6
		Radwaste Bldg.	0.31	94.5
	1/4/07 - 1/8/08	Fuel Bldg.	0.65	4.3
		Primary Aux. Bldg.	0.16	48.9
		Sec. Aux. Bldg.	0.24	41.6
		Radwaste Bldg.	0.37	83.2
		Containment Bldg.	0.20	97.3
	1/8/08 - 10/17/08	Fuel Bldg.	0.50	13.1
		Primary Aux. Bldg.	0.11	58.9
		Sec. Aux. Bldg.	0.12	40.1
		Radwaste Bldg.	0.91	90.1
		Containment Bldg.	0.35	88.0
		Weighted Average % Organic ^a :		49.6
Ulchin Units 1 & 2	7/14/06 — 12/27/06	Plant Stack	2.25	53.5
Framatome	12/27/06 - 8/3/07	Plant Stack	8.19	40.0
2 @ 950 MWe each	1/3/08 - 10/18/08	Plant Stack	5.73	43.4
		Weighted Ave	rage % Organicª	43.1

a. Our calculation.

Weekly reactor coolant and spent fuel pool samples also were collected using pre-evacuated 20 ml glass vials with septum. Five samples were combined prior to 14 C extraction and analysis. A three step procedure was utilized: gas phase extraction, acid stripping and wet oxidation using $K_2S_2O_8$ plus AgNO₃. Results are shown in Table 4-24.

Table 4-23 Normalized Gaseous Effluent Release from Some KHNP Plants (D. Lee (2010))

Plant	Rated Power (MW _e)	Year	Gaseous ¹⁴ C Release (Ci/GW _e -yr)	% Organic
Kori Unit 1	600	2007	7.1	47.7
		2008	1.0	25.4
Kori Unit 3	950	2007	5.0	19.5
		2008	9.5	31.5
Yonggwang Unit 5	1000	2007	1.8	43.7
		2008	2.8	65.6
Ulchin Units 1&2	2*950	2007	4.6	39.9
		2008	4.1	43.4

Table 4-24
Reactor Coolant and Spent Fuel Pool Analyses at Five Korean PWRs (D. Lee (2010))

	Total μCi/g	Fraction Gaseous	% Organic
RCS Analysis			
Kori 1	1.67E-4	0.17	91.8
Kori 3	1.09E-4	0.60	74.6
Yonggwang 5	2.20E-4	0.35	90.0
Ulchin 1	4.34E-4	0.50	90.1
Ulchin 2	3.07E-4	0.39	87.3
Spent Fuel Pool Analysis			
Kori 1	4.26E-6	0.74	21.4
Kori 3	1.32E-5	0.88	15.0
Yonggwang 5	6.83E-6	0.66	31.0
Ulchin 1	3.54E-5	0.75	14.3
Ulchin 2	3.27E-5	0.84	8.6

Most of the ¹⁴C in the primary coolant was in the organic form. The predominant chemical form in the spent fuel pool was inorganic.

4.5 Chemistry Effects

Primary coolant chemistry control in PWRs is relatively similar from plant to plant. Boron is used as the chemical shim, and lithium is added to increase the at-temperature pH (pH_T) to 6.9 or above to reduce general corrosion rates and corrosion product deposition on the fuel. Hydrogen is added to reduce oxygen to an effectively non-detectable level thereby minimizing the risk of stress corrosion cracking. The electrochemical potential (ECP) is in the range of -800 mV. At this potential, the primary 14 C compounds are expected to be low molecular weight organics and HCO₂⁻.

Since hydrazine is added to the primary system to reduce oxygen concentrations below 100 ppb at temperatures above $\sim 100^{\circ}$ C during startups, ammonia will also be present in the primary coolant during power operation due to the decomposition of hydrazine. Ammonia can also be formed from nitrogen which enters the system with makeup water. Ammonia concentrations in the primary coolant can be measured by ion chromatography. The nitrogen gas concentration can be calculated from measurements of the gas phase concentration in the volume control tank (Reference Appendix E).

Zinc is now being added to the primary system as zinc acetate at approximately half of the operating US PWRs to reduce shutdown dose rates and materials corrosion rates. The zinc concentration is controlled in the range of \sim 5 to \sim 30ppb. This concentration of zinc does not impact on the pH_T of the primary coolant. Formation of ¹⁴C from the carbon present in the acetate is minimal (Reference Section 2.1.1).

4.6 Effects of Fuel Failures

The 14 C production rate in the fuel is approximately 20 Ci/GW_e-yr (see Table 4-3). With approximately 50,000 fuel pins in the reactor core, a fuel pin will have an inventory of approximately 800 μ Ci of 14 C after two GW_e-yrs of operation. A fuel defect instantly releasing 10% of this inventory (a very high estimate of the release) into the reactor coolant would increase the 14 C concentration by approximately 3E-7 μ Ci/g. A steady state release of 10% of the production rate in a single defective fuel rod would be \sim 1E-6 μ Ci/sec. Either input is a very small fraction of the 14 C production rate in the reactor coolant and can be neglected as a possible source term during normal operation.

4.7 Reactor Make-up Water

Large dilution water make-ups are made during mid-life to end of core life (EOL) operation. As the reactor fuel is used, boron is removed from the reactor coolant system in increasing amounts via dilution with non-borated water to maintain reactor power. There are three periods of low dilution during the fuel cycle: (1) early in core life, (2) near EOL when a deborating

demineralizer is used in place of dilutions to lower the RCS boron, and (3) during reactor coast down. The water used for this dilution comes from the reactor water make-up storage tank (RWST) or equivalent and will contain dissolved nitrogen at concentrations between 15 and 25 ppm depending upon the cover gas composition and tank water temperature.

A reactor coolant concentration of 1 ppm nitrogen (as N_2) in a 4-loop Westinghouse PWR will have ~40 moles of nitrogen in the RCS system with ~21% in the reactor coolant, ~74% in the VCT gas phase and ~5% in the PZR gas phase. To illustrate the impact of makeup water addition for boron dilution, consider a 10,000 gallon/day dilution of RWST water containing 20 ppm nitrogen. This dilution will introduce ~27 moles of nitrogen a day in to the RCS which is equivalent to a potential increase in the RCS liquid nitrogen concentration of 0.6 ppm. As a consequence of this dilution, the source term of 14 C will tend to increase sharply during the EOL operation unless compensating steps are taken to reduce the nitrogen concentration in the VCT during this high dilution period.

4.8 Summary of PWR ¹⁴C Transport and Release

Calculated values of 14 C generation rates and observed release rates via gaseous release pathways are summarized in Tables 4-25 and 4-26, respectively. Values are shown in the units reported by each researcher and have also been normalized to a standard release rate unit of Ci/GW_{th} -year and Bq/GW_{th} -year. A thermal efficiency of 34% has been assumed to convert from MW_e to MW_{th} .

The calculated source term for the Westinghouse units (Appendix D) averaged 0.387 ± 0.025 $\mu\text{Ci/MW}_{th}$ -h (3.4 ± 0.2 Ci/GW $_{th}$ -yr or 10.0 ± 0.6 Ci/GW $_{e}$ -yr), whereas, the CE units were somewhat higher at 0.445 ± 0.032 $\mu\text{Ci/MW}_{th}$ -h (3.9 ± 0.3 Ci/GW $_{th}$ -yr or 11.5 ± 0.8 Ci/GW $_{e}$ -yr). There is reasonably good agreement between this program's efforts and the calculations performed by Magnusson (2008) for Ringhals 3 and 4.

Based upon the observations reported in this section and the calculations summarized above, ¹⁴C transport in the PWR can be schematically summarized as shown in Figure 4-4. This information can be used to estimate the amount of total generated carbon-14 that is released via gaseous effluent as carbon dioxide or organic carbon. The inorganic fraction of the gaseous releases will depend on the plants operating mode. In the absence of a recombiner in the waste gas treatment system, the inorganic fraction of the gaseous release has been observed to be as low as 5% during normal operation but this fraction rises significantly during the shutdown evolutions and refueling activities.

Table 4-25 Summary of Calculated ¹⁴C Generation Rates in Coolant at PWRs^a

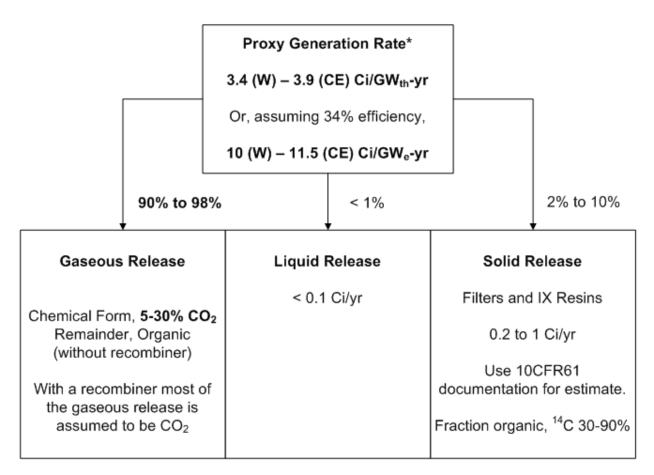
Reference	Unit	μCi/MW _t -h	Ci/yr	Ci/GW _e -yr	GBq/GW _e -yr
Fowler et al (1976)				3.3	
Bonka (1974)	-			11.1	
Kelly (1975)	1000 MW _e			6	
ERDA-1535 (1975)	-			6	
Hayes (1977)	1000 MW _e			3.4	
Davis (1977)	-			5	
Vance (1995)				6.12	270-410
Magnusson (2008)	Ringhals-3 2775 MW _{th}	0.403	9.80	10.4	385
Magnusson (2008)	Ringhals-4 2775 MW _{th}	0.411	10.0	10.6	392
This study (see Appendix D) ^b	W-A, 4-Loop 3216 MW _{th}	0.357	10.1	9.20	340
This study (see Appendix D) ^b	W-B, 4-Loop 3188 MW _{th}	0.360	10.1	9.28	343
This study (see Appendix D) ^b	W-C, 4-Loop 3650 MW _{th}	0.432	13.8	11.1	411
This study (see Appendix D) ^b	W-D, 2-Loop 1540 MW _{th}	0.387	5.23	10.0	370
This study (see Appendix D) ^b	W-E, 2-Loop 1540 MW _{th}	0.387	5.23	9.98	369
This study (see Appendix D) ^b	W-F, 4-Loop 3455 MW _{th}	0.396	12.0	10.2	377
This study (see Appendix D) ^b	W-G, 4-Loop 3626 MW _{th}	0.387	12.3	9.98	369
This study (see Appendix D) ^b	CE-A 2700 MW _{th}	0.467	11.1	12.0	444
This study (see Appendix D) ^b	CE-B 3716 MW _{th}	0.422	13.7	10.9	403

a. Bold is reported data. Italics are converted bold data assuming 34% thermal efficiency.

b. At reactor coolant nitrogen concentration of 0 ppm (e.g. only the $^{17}O(n,\alpha)^{14}C$ reaction considered).

Table 4-26 Summary of Observed Gaseous Release Rates at PWRs

		Gaseous Release Rates		Gaseous Relea	
Reference	Unit	Ci/yr	Ci/GW _e -yr		
NUREG0017 (Rev 1)	Conn Yankee	46			
NUREG0017 (Rev 1)	Yankee Rowe	0.58			
NUREG/CR-1629	Turkey Point 3/4	3.7			
NUREG/CR-0140	Ft Calhoun	1.9			
NUREG/CR-0715	Zion 1 and 2	3.3			
NUREG/CR-4397	Prairie Island 1/2	3.6			
NUREG/CR-2348	Rancho Seco	3.6			
Kunz, 1985	R. E. Ginna		11.6		
Kunz, 1985	Indian Point 3		9.6		



^{*} A unit specific calculation can be conducted per the guidance provided in Section 4.3 or the proxy value provided can be used to estimate the generation of 14 C. The uncertainty on the proxy value is $\pm 15\%$.

Figure 4-4

14C Transport in the PWR

5

MEASUREMENT IN THE NUCLEAR POWER INDUSTRY

5.1 General

Historically, most of the early reactor site ¹⁴C measurements were made using proportional gas flow counters. However, most nuclear stations now have liquid scintillation counters for tritium measurements, and improvements in the instruments and methodology make this the preferred instrument for ¹⁴C measurement. There is also the possibility of using accelerator mass spectrometry (AMS), but this is an expensive and complex methodology developed primarily for archeological dating purposes as it uses very small samples, and reduces the minimum measurable ¹⁴C to ¹²C ratio by at least a factor of ten.

Reactor site ¹⁴C measurements have been performed on gaseous effluents, liquid streams, solid wastes and environmental samples. The latter includes atmosphere, water, milk, plant, and animal tissue. Irrespective of the source, almost all determinations involve counting the ¹⁴C activity in the form of carbon dioxide or as a carbonate or carbamate salt. The usual gaseous effluent and atmospheric ¹⁴C sampling and analysis approach involves passing the sample through a particulate filter followed by a dilute sulfuric acid bubbler to remove tritium in the form of HTO and then a sodium hydroxide bubbler to capture ¹⁴C in the form of carbon dioxide (CO₂) as sodium carbonate. The gaseous effluent from this bubbler is passed over a high temperature catalytic bed with sufficient oxygen to convert organic compounds and carbon monoxide (CO) to carbon dioxide. The effluent from the catalytic converter then goes through a second sodium hydroxide scrubber to remove the carbon dioxide formed in the catalytic bed. Note that ¹⁴C in the carbon dioxide form is more significant than the organic or carbon monoxide fraction relative to dose, and this methodology separates the two fractions.

The normal PWR waste gas decay tank contains insufficient oxygen for the catalytic converter to convert organic compounds to CO_2 . The usual procedure is to capture the sample in a gas cylinder and to take it to a laboratory where it is diluted with air or oxygen and then processed through a catalytic converter. It is recommended that the sample be diluted with at least 20 times its volume to assure that the H_2 concentration is maintained less than 5%.

An alternate technique is the use of barium nitrate in the place of sodium hydroxide with precipitation of the CO_2 as barium carbonate provided there is enough CO_2 to exceed the $BaCO_2$ solubility. (Solubility product 7.0E-9 (moles/liter)² at room temperature). The barium carbonate precipitate can be suspended in the scintillation cocktail and counted in this form. Another alternative is to eliminate the caustic scrubber and to absorb the CO_2 in an organic solvent such as ethanolamine that can be transferred directly to the scintillation cocktail.

Other considerations include the radiochemical purity of the sample, e.g., are iodine radio-nuclides or the decay products of noble gases collected in the bubbler solution. If so, the bubbler contents can be acidified and nitrogen gas used to transfer the resulting CO_2 to a new absorbing bubbler.

It is suggested that atmospheric samples in the plant environs be large enough to quantify ¹⁴C at the natural background level of approximately 1.2 pCi/m³ of air, or 0.044 Bq/m³ of air (Sejkora, 2010). Air is 0.034% by volume CO₂. This corresponds to 0.67 g of CO₂ per cubic meter of air. Olivier, et. al (2005) compared the liquid scintillation counting sensitivity of direct counting of a Na₂CO₃ solution mixed with the cocktail and precipitating the CO₂ with CaCl₂ and suspending the precipitate in the cocktail. The detection limit for the NaCO₃ solution was 0.3 Bq/m³ and for suspended CaCO₃ 0.04 Bq/m³. CaCO₃. Large samples of 100 cubic meters or more of air can result in quantities of inert CO₂ that exceed the capacity of the scintillation cocktail, and an analytical procedure has been developed to convert the CO₂ absorbed in the caustic bubbler to benzene which has a much higher cocktail solubility (Woo, et al., 1999).

5.2 Study Methodologies

Some of the earliest reported ¹⁴C measurements were those made by Kunz, Mahoney, and Miller at the Ginna, and a Indian Point 1 and 2 PWRs (Kunz, 1974). Samples of the gas decay tanks and containment air were analyzed for ¹⁴C. One to five liter samples of containment air and 10 ml aliquots of decay tank air were mixed with measured amounts of Ar, Kr, Xe, H₂, CO₂, CO, CH₄, C₂H₆, C₃H₈, and C₄H₁₀ carrier gases. Various steps were used to separate the noble gas, tritium, and carbon compounds, but ultimately the carbon compounds were fractionated by use of a molecular sieve column. These fractions were then loaded into a gas proportional counter tube to determine the ¹⁴C activity of each chemical form. Pulse height analysis permitted simultaneous analysis of the tritium and ¹⁴C fractions. The results for both the gas decay tank and containment atmosphere samples showed that in all cases over 80% of the ¹⁴C activity was in the methane and ethane fractions with the majority as methane. The CO₂ fraction ranged from 4.6% to 1.8% except for the Indian Point-1 decay tank with 0.3% CO₂. In all samples, the CO fractions ranged from 0.4% to non-detectable.

In 1976, Kunz, et al. (Kunz, 1976) reported on ¹⁴C measurements at the Nine Mile Point 1 BWR. Offgas samples ranging in volume from 100 to 500 cm³ were taken from the main condenser SJAE discharge. These samples were mixed with measured amounts of carrier gases, Kr, Xe, CO, CO₂, CH₄, C₂H₆, C₃H₈, and C₄H₁₀. The carrier gases then were fractionated by a combination of cryogenic and gas chromatographic techniques. The individual fractions were loaded into a gas proportional counter to determine the ¹⁴C activity in each fraction. The total ¹⁴C activity was also determined by passing another aliquot of offgas together with the carrier gases over an 800°C CuO bed to oxidize the carbon compounds to CO₂ prior to counting. The activity distributions showed that 95% of the off gas activity was in the form of CO₂, 2.5% as CO, and 2.5% as organics. (Note that these measurements predate the retrofit of an offgas system recombiner).

In 1974, Hays and MacMurdo (Hayes, 1977) measured the ¹⁴C in the stack release and spent resins at the Savannah River heavy water reactor (HWPR). The stack exhaust measurements were made by flowing a sample stream of exhaust air at approximately 200 cc/min through a column containing a commercially available solid form CO₂ absorber, (MallcosorbTM). Following sampling, the absorbed CO₂ was released in a closed gas circulating system by adding HCl to a water solution of the absorbent. The CO₂ was then reabsorbed in a NaOH solution, which was heated, and BaCl₂ was then added to precipitate BaCO₃. The precipitate was filtered, dried, weighed and checked for residual gamma activity. If gamma activity was detected, the BaCO₃ was dissolved in acid, and the resulting CO₂ was absorbed in a NaOH solution and reprecipitated as BaCO₃. The volume of air sampled was calculated based on the weight of the BaCO₃ recovered assuming a nominal 315 ppm CO₂ in air. The volume obtained agreed within 5% of the value obtained from the sample flow meter. The BaCO₃ was dissolved slowly with HCl, and the released CO₂ was scrubbed from the air stream using a spinning band column and a countercurrent-flow of liquid scintillation solution. The ¹⁴C was determined by liquid scintillation counting, but no details were given regarding the liquid scintillation solution or the counting process.

Kunz later published the results of extensive ¹⁴C sampling and analysis programs at the Fitzpatrick BWR and at the Indian Point-3 and Ginna PWRs (Kunz, 1985). Continuous samplers were installed on the gaseous activity discharge points and the ventilation vents to measure total ¹⁴C and ¹⁴CO₂ in the effluent gases. The continuous samplers used a 100 cm³/min diaphragm pump to draw the sample through a 600°C catalytic bed of palladium and platinum on alumina that oxidized all reduced forms of carbon to CO₂. The gas then flowed through a solid drying agent such as Drierite™ (anhydrous CaSO₄) to remove water vapor, including HTO, and then through a cartridge containing 25 g of 8 to 20 mesh Ascarite™ (NaOH on a solid silicate support) to absorb the CO₂. After sampling, the cartridge was removed, the contents acidified and helium carrier gas used to transport the released CO₂ to a liquid nitrogen cold trap which liquefied the CO₂. The volume of liquid CO₂ was measured, usually 660 cm³ for a 2-week sampling period, and a 50 cm³ aliquot of the liquid CO₂ was purified by a gas chromatography and loaded into a gas proportional counter.

In addition to the continuous samples, grab samples were taken at these release points. Carrier gases were added, and the samples analyzed for chemical species and their activity using gas chromatography and internal gas counting as previously described.

In 1995 Vance and Cline, and their respective associates, published an EPRI report (Vance 1995) characterizing nuclear power plant ¹⁴C In addition to reviewing Kunz's work, they also published the primary coolant ¹⁴C concentration they measured at three BWRs and three PWRs. The analysis procedure used 300 ml samples which were acidified with H₂SO₄ and CO₂ free air used to sweep the dissolved CO₂ into a Ca(OH)₂ trap precipitating CaCO₂. The CaCO₂ was further purified and the ¹⁴C activity measured by liquid scintillation counting. The total inorganic and organic ¹⁴C activity was determined by refluxing the coolant sample at 90°C with K₂S₂O₈ and AgNO₃ catalyst to oxidize all carbon forms to CO₂ which was analyzed as described. The organic fraction was determined by the difference in the results of the two procedures.

Woo, et.al. (Woo, 1999) measured the 14 C release at the Wolsong nuclear reactor site which has a total of 6 CANDU reactors. At CANDU heavy water reactors, 14 C production is significantly higher than in US light water reactors, but their sampling and analysis techniques are applicable. For environmental air samples, the air was drawn through a bubbler containing 200 ml of 2N NaOH at a sample flow of about 400 ml/min. Sample periods were 2-4 weeks resulting in a sample volume of 8 to 16 m³ of air. The absorption of CO_2 in the bubbler was found to be more than 99.5%. A diagram of the sampler is shown in Figure 5-1. The first bubbler was filled with 0.5N H₂SO₄ to remove tritium in the form of HTO. The second bubbler was filled with 2N NaOH to trap N as N as N and N are effluent from this bubbler went through a 600°C tube furnace filled with a platinum and palladium catalyst to oxidize organic carbon to N as N and N are effluent was then passed through a bubbler filled with N NaOH to absorb the N as N and N are effluent was then passed through a bubbler filled with N and N absorb the N as N and N are effluent was then passed through a bubbler filled with N and N as N as N and N are efficiency exceeded 99.2%. (Note that any tritium present as HT will be oxidized to HTO in the tube furnace. Inclusion of an N bubbler at the furnace outlet would address this possible issue.)

Woo, et.al. (1999) also described the process of using open, plastic trays filled with 600 ml of $2\underline{M}$ NaOH to measure environmental, atmospheric $^{14}\text{CO}_2$ levels. The trays were exposed to the atmosphere for 2 to 4 weeks, and then taken to the laboratory for determination of the amount of sodium carbonate and ^{14}C . Results were reported in terms of the ^{14}C specific activity, Bq ^{14}C per gram of carbon as CO_2 . The difference in specific activity of the active and passive sampling methods was 5% maximum with the passive method results generally lower than the active ones.

For ground water sampling they found that closed loop, nitrogen sparging of an acidified sample to remove the CO₂ was more effective than BaCO₃ precipitation. The nitrogen sparge would also be expected to extract most low molecular weight organic forms.

Biological samples were rinsed with 1% hydrochloric acid to remove surface contaminants and then oven dried. Tissue samples were freeze-dried and ground to a powder. The samples were then combusted, and the resulting CO_2 transferred to a bubbler containing 150 ml of 3M NaOH solution.

In all cases the 14 C activity was determined by liquid scintillation counting. A mixture of NH₄Cl and BaCl₂ was added to the NaOH bubbler solution to precipitate BaCO₃. Samples resulting in significant quantities of BaCO₃ were analyzed by suspending the precipitate in 12 ml of "Instagel XF" (Perkin Elmer "Insta-Gel Plus") plus 6 ml of water. Small quantities of precipitate were acidified and the resulting CO₂ absorbed in 10 ml of "Carbsorb E" plus 10 ml of "Pemafluor V".

The authors also describe the benzene synthesis method that converted the CO₂ from the BaCO₃ to benzene that then was added directly to the scintillation cocktail. This method was used infrequently.

5.3 Current Methods

Essentially all ¹⁴C measurements involve trapping of CO₂. In the case of BWRs, at least 95% of the gaseous ¹⁴C is released as CO₂. At most PWRs, a large fraction of the ¹⁴C release is in an organic form, and to determine the total ¹⁴C activity these compounds can be oxidized to CO₂ using a high temperature catalytic converter. Carbon activity in the form of carbonates can be released from liquids and resins by addition of hydrochloric acid and nitrogen carrier gas. In addition, strong oxidizers such as potassium peroxydisulfate can be used to oxidize any non-carbonate liquid compounds to CO₂. Vegetation and animal tissue as well as resins can be oxidized in combustion units to convert any carbon compounds to CO₂.

The released CO₂ can be trapped in a bubbler containing NaOH or KOH and later precipitated as BaCO₃. If it is an atmospheric sample, it can be filtered, washed, dried and weighed to determine the chemical yield based on the known atmospheric CO₂ concentration, and the sample volume. If further radiochemical purification is needed, the precipitate can be dissolved in a closed system using HCl or H₂SO₄, and nitrogen carrier gas used to transfer the resulting CO₂ to another caustic containing bubbler. At this point, there are several choices. There are organic CO₂ absorbers such as ethanolamine that form carbamates that are miscible with LSC scintillation phosphors. Alternately, the BaCO₃ can be suspended in the scintillation cocktail. The choice of methodology depends primarily on the amount of inert CO₂ and the absorbing media.

BWR gaseous effluent sampling is straightforward. Most of the activity is released by the offgas treatment system, and it is in the form of CO_2 as a result of having been passed through the offgas system recombiners.

At an assumed ¹⁴C production rate of 15 Ci/yr and 25 scfm air inleakage, the offgas system discharge concentration is 80 d/m per cm³. Depending on the particular plant, the discharge may be through a dedicated vent or be released with various combinations of ventilation exhaust varying from 30,000 to 400,000 cfm. At 400,000 cfm the ¹⁴C activity would be 0.093 d/m liter, and roughly 100 liters would be required to achieve a 10 d/m sample. Assuming air is 0.035% by volume CO₂, the atmospheric concentration of CO₂ is 0.0168 millimoles per liter, and a 100 liter sample would contain 1.68 millimoles of CO₂ and would require 16.8 ml of 0.1 M absorber.

Perkin Elmer (PE) has published trapping capacity of various absorbents, which shows for 0.1 molar hydroxide solution 0.05 moles of CO_2 Na or K hydroxide solution can be absorbed per data and the background specific activity of ^{14}C is 15 d/m per gram of carbon, the atmospheric concentration of CO_2 is 0.0168 millimoles per liter, and the natural occurring ^{14}C activity is 6.48E-5 d/m per liter.

5.4 Commercial Sampling Systems

Three commercially available gas effluent samplers that include combustion capability have been identified. These are the Mound Technical Solutions Model MRB500C14 [M1], also available through F&J as Model MRB500C14 [S-1], the Overhoff Technology Corporation TASC, and the SDEC HAGUE 7000-C14 [S-2]. They are similar in that they both consist of four to six gas bubblers in series with a high temperature catalytic combustion tube in the middle of the string of bubblers. Both units include electronic flow control monitors and oven temperature controls. The primary difference is the Mound and Overhoff units have two sets of three standard scintillation counter ready vials (20 ml) intended to be filled with ethanolamine to absorb the CO₂; whereas, the Hague 7000 uses four 250 ml vials intended to be filled with sodium hydroxide. The cascaded bubblers provide assurance that all the ¹⁴C has been trapped. It appears that the Mound and Overhoff units are intended for higher activity gas streams as the bubbler sizes are small, but require less sample manipulation prior to counting. The SDEC sampler will accept much larger quantities of CO₂ and is more suitable for environmental monitoring.

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A BWR ¹⁴C SOURCE TERM CALCULATION

1.0 Cross-Section Estimation

The "effective cross-sections" for the two dominant nuclear reactions producing ¹⁴C in the BWR were calculated from detailed reaction cross-section and neutron flux data as a function of neutron energy.

The cross-section data were obtained from ENDF files. The neutron flux in both the moderator and bypass regions was obtained for GE9 8x8 fuel with a large water rod via a private communication between one of the authors and K. Watson of Transware Inc. The neutron flux data were for burn-ups of 0 to 50 MWD/MTU. A basic tenet of this effort was that although the fluxes in the energy groups of interest (\leq 0.625 eV, >0.625 eV to <1 MeV, \geq 1 MeV and >0.625 eV) vary with burn-up, the general shape of the flux distribution does not, and thus the "effective cross-section" should only be a mild function of burn-up.

The production rate for the $^{17}O(n,\alpha)^{14}C$ reaction was obtained by multiplying the average cross-section in an energy interval by the average neutron flux in the same energy interval for each of the energy intervals provided in the neutron flux distribution. In the case of the BWR flux distribution, there were 97 energy intervals ranging from 2.53E-4 eV to 10 MeV. The production curve was integrated over the desired energy range, and the integral was divided by the total neutron flux in that interval to calculate the "effective cross-section." This calculation was performed at intervals of 10 MWD/MTU with the average value of the cross-section over the range of 10 to 40 MWD/MTU used to calculate the value of the "effective" cross-section.

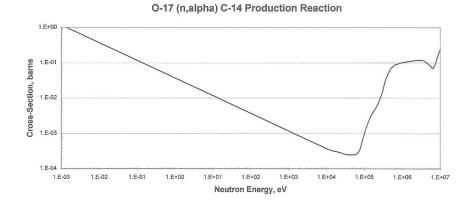
For the $^{14}N(n,p)^{14}C$ reaction, which is only of minor importance in the BWR, the calculation was performed for only the 50 MWD/MTU flux data.

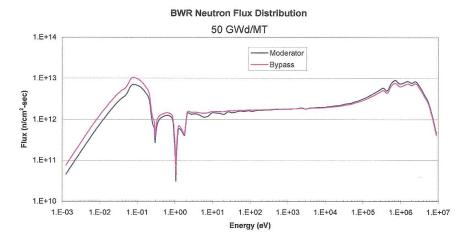
2.0 $^{17}O(n,\alpha)^{14}C$ Reaction

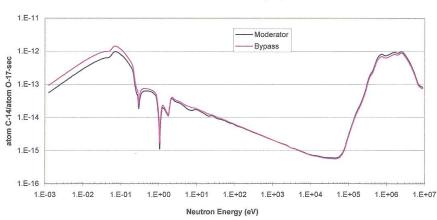
The $^{17}O(n,\alpha)^{14}C$ reaction cross-section, the reference BWR neutron flux distribution at 50 GWd/MT and the ^{14}C production by this nuclear reaction are shown in Figure A-1.

To utilize this cross-section data in source term calculations, the digital data were fit to a number of equations to cover the energy/cross-section spectrum. The equations used in the evaluation of the source term are summarized in Table A-1.

The flux in each of the neutron groups is a function of fuel exposure as shown in Figures A-2 and A-3.







BWR C-14 Production from O-17(n,alpha)C-14 Reaction

Figure A-1 $^{17}\text{O}(\text{n},\alpha)^{14}\text{C}$ Reaction Cross-Section, Reference BWR Neutron Flux Distribution at 50 GWd/MT and ^{14}C Production

Table A-1 Equations Utilized to Fit $^{17}O(n,a)^{14}C$ Cross-Section Data

Eqn.	Neutron Energy Range (eV)	Equation
1	1.00E-5 to 1.00E4	$\sigma = 3.7379E-2 \cdot E_n^{-0.50}$
2	1.00E4 to 1.00E5	$\sigma = 5.3265E-23 \cdot E_n^4 - 8.7788E-18 \cdot E_n^3 + 5.9072E-13 \cdot E_n^2 - 1.9754E-8 \cdot E_n + 5.2051E-4$
3	1.00E5 to 4.00E5	$\sigma = -1.5294E-28 \cdot E_n^5 + 1.7661E-22 \cdot E_n^4 - 7.5316E-17 \cdot E_n^3 + 1.5100E-11 \cdot E_n^2 - 1.3876E-6 \cdot E_n + 4.7976E-2$
4	4.00E5 to 1.00E6	σ = -4.5446E-25·E _n ⁴ + 1.7645E-18·E _n ³ – 2.5311E-12·E _n ² + 1.6284E-6·E _n - 3.0425E-1
5	1.00E6 to 6.17E6	$\sigma = 2.4466E-28 \cdot E_n^4 - 2.9005E-21 \cdot E_n^3 + 6.5493E-15 \cdot E_n^2 + 9.3313E-9 \cdot E_n + 8.9792E-2$
6	6.17E6 to 1.17E7	$\sigma = 6.9362E-28 \cdot E_n^4 - 2.7631E-20 \cdot E_n^3 + 4.0062E-13 \cdot E_n^2 - 2.4635E-6 \cdot E_n^2 + 5.5042$
7	1.17E7 to 2.00E7	$\sigma = -1.3435E-28 \cdot E_n^4 + 8.9157E-21 \cdot E_n^3 - 2.2040E-13 \cdot E_n^2 + 2.3840E-6 \cdot E_n^2 - 9.2185$

The "effective cross-sections" and production calculations for the $^{17}O(n,\alpha)^{14}C$ reaction utilizing the 0, 10, 20, 30, 40 and 50 GWD/MT BWR neutron flux distributions are summarized in Table A-3.

The recommended "effective cross-sections" for the $^{17}O(n,\alpha)^{14}C$ reaction in the BWR are summarized in Table A-2.

Table A-2 "Effective Cross-Sections" for the $^{17}\text{O}(\text{n}, a)^{14}\text{C}$ Reaction in the BWR

		"Effective Cross-Section (σ)", b		
Neutron Group	Group Energy	Moderator Bypass		
Thermal	≤0.625 eV	0.1325	0.1386	
Intermediate (I)	>0.625 eV - <1 MeV	0.0238	0.0222	
Fast (F)	≥1 MeV	0.1106	0.1106	
I+F	>0.625 eV	0.0458	0.0432	

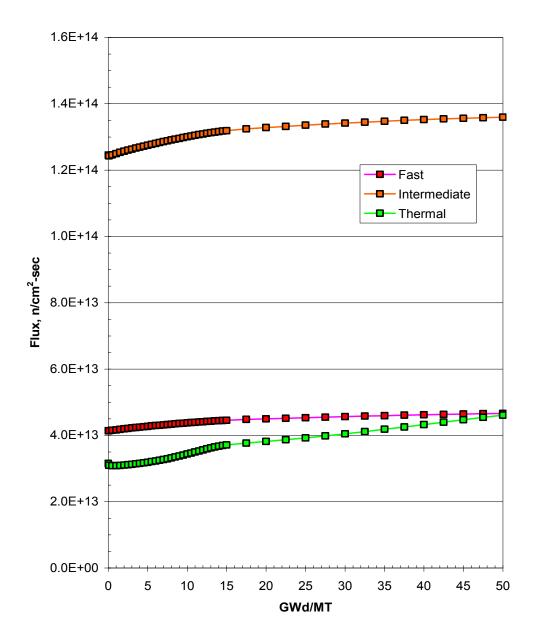


Figure A-2 BWR Moderator Neutron Flux vs. Fuel Exposure

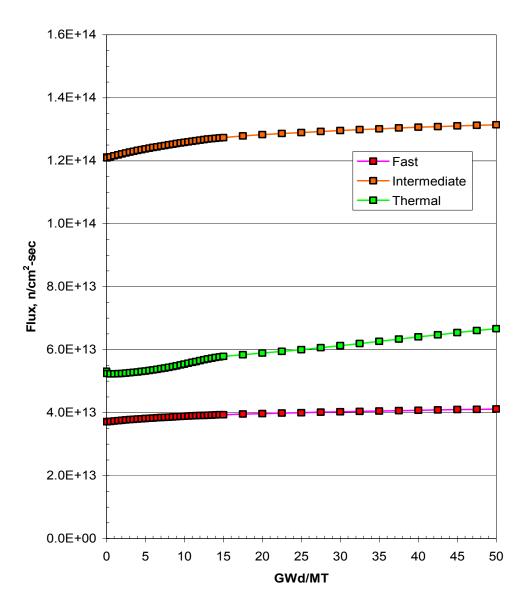


Figure A-3 BWR Bypass Neutron Flux vs. Fuel Exposure

Table A-3 Summary of $^{^{17}}\text{O}(\text{n},\alpha)^{^{14}}\text{C}$ Production Reaction in the BWR

	Neutron Flux, n/cm²-sec					
	Moderator			Bypass		
GWd/MT Exposure	Thermal	Inter- mediate	Fast	Thermal	Inter- mediate	Fast
0	3.16E13	1.24E14	4.14E13	5.31E13	1.21E14	3.71E13
10	3.44E13	1.30E14	4.38E13	5.54E13	1.26E14	3.88E13
20	3.82E13	1.33E14	4.50E13	5.89E13	1.28E14	3.97E13
30	4.05E13	1.34E14	4.57E13	6.13E13	1.30E14	4.03E13
40	4.33E13	1.35E14	4.62E13	6.40E13	1.31E14	4.07E13
50	4.61E13	1.36E14	4.66E13	6.67E13	1.31E14	4.11E13
	"Effective" Cross-Section, barn					
	Moderator		Bypass			
GWd/MT Exposure	Thermal	Inter- mediate	Fast	Thermal	Inter- mediate	Fast
0	0.1284	0.0239	0.1107	0.1374	0.0224	0.1106
10	0.1298	0.0239	0.1106	0.1377	0.0223	0.1106
20	0.1321	0.0239	0.1106	0.1385	0.0223	0.1106
30	0.1334	0.0238	0.1106	0.1389	0.0222	0.1106
40	0.1345	0.0238	0.1106	0.1394	0.0222	0.1106
50	0.1355	0.0238	0.1106	0.1399	0.0222	0.1106
			Production Ra	ate, µCi/sec-kg		
	N	loderator Regio	on	Bypass Region		
GWd/MT Exposure	Thermal	Inter- mediate	Fast	Thermal	Inter- mediate	Fast
0	5.33E-6	3.92E-6	6.03E-6	9.60E-6	3.57E-6	5.41E-6
10	5.87E-6	4.09E-6	6.37E-6	1.00E-5	3.69E-6	5.65E-6
20	6.63E-6	4.17E-6	6.55E-6	1.07E-5	3.76E-6	5.78E-6
30	7.10E-6	4.21E-6	6.64E-6	1.12E-5	3.79E-6	5.86E-6
40	7.66E-6	4.23E-6	6.72E-6	1.17E-5	3.82E-6	5.93E-6
50	8.22E-6	4.25E-6	6.78E-6	1.23E-5	3.84E-6	5.99E-6
	Total Production Rate, μCi/sec-kg					
GWd/MT Exposure	Moderator Region		Bypass Region			
0	1.53E-5		1.86E-5			
10	1.63E-5		1.93E-5			
20	1.74E-5		2.02E-5			
30	1.80E-5		2.09E-5			
40	1.86E-5		2.15E-5			
50	1.93E-5		2.21E-5			

2.1 Source Term Calculation

Using the flux and cross-section data in Table A-3, the calculated ¹⁴C production per kilogram of water at an exposure of 30 GWd/MT is illustrated below. The number of ¹⁷O atoms per kilogram of water is 1.27E22.

2.1.1 Moderator Region

$$\frac{1.27E22 \bullet (4.05E13 \bullet 0.1334 + 1.34E14 \bullet 0.0238 + 4.57E13 \bullet 0.1106) \bullet 1E - 24 \bullet ln2}{3.7E4 \bullet 5730 \bullet 365.25 \bullet 24 \bullet 3600}$$
$$= 1.80E - 5 \mu Ci / sec - kg$$

Production in the moderator region is \sim 40% from thermal neutrons, \sim 24% from intermediate neutrons and \sim 37% from fast neutrons.

2.1.2 Bypass Region

Production in the bypass region is \sim 54% from thermal neutrons, \sim 18% from intermediate neutrons and \sim 28% from fast neutrons.

2.1.3 Core Production Rate

The production rate at a 3579 MW_{th} GE-BWR with 748 fuel assemblies, 12,655 kg of water in the moderator region and 17,100 kg of water in the bypass or leakage region, is as follows:

```
\begin{array}{ll} 1.80\text{E-}5*12,\!655 + \ 2.08\text{E-}5*17,\!100 = \ 0.573\ \mu\text{Ci/sec} \\ &= \ 18.1\ \text{Ci/yr} \\ &= \ 14.9\ \text{Ci/GW(e)-yr}\ (@34\%\ \text{efficiency}) \\ &= \ 21.3\ k\text{Bq/MW}_{\text{th}}\text{-h} \\ &= \ 0.576\ \mu\text{Ci/MW}_{\text{th}}\text{-h} \\ &= \ 5.05\ \text{Ci/GW}_{\text{th}}\text{-yr} \end{array}
```

This calculation was repeated over the range of the BWR fuel flux/exposure data with the results reported in Table A-3.

2.1.4 Site Specific Calculations

For site specific calculations, "effective cross-sections" for the $^{17}O(n,\alpha)^{14}C$ reaction given in Table A-2 are recommended.

3.0 ¹⁴N(n,p)¹⁴C Reaction

The ¹⁴N(n,p)¹⁴C reaction cross-section, the reference BWR neutron flux distribution (GE9 Fuel, 8x8 with large center water rod) at 50 GWd/MT and the ¹⁴C production by this nuclear reaction are shown in Figure A-4. The flux distribution (three reference energy groups) used in this calculation is provided in Table A-4. The "effective cross-sections" for the ¹⁴N(n,p)¹⁴C reaction utilizing the reference 50 GWd/MT BWR neutron flux distribution are summarized in Table A-5.

Table A-4
Reference BWR Neutron Flux (50 GWd/MT Exposure)

		Neutron Flux, n/cm²-sec		
Neutron Group	Group Energy	Moderator	Bypass	
Thermal	≤0.625 eV	4.61E13	6.67E13	
Intermediate (I)	>0.625 eV - <1 MeV	1.36E14	1.31E14	
Fast (F)	≥1 MeV	4.66E13	4.11E13	
I+F	>0.625 eV	1.83E14	1.72E14	

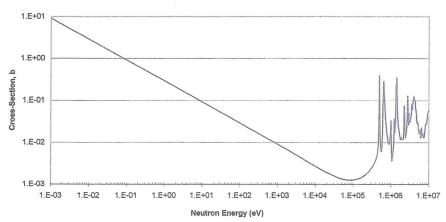
Table A-5 "Effective Cross-Sections" for the ¹⁴N(n,p)¹⁴C Reaction in the BWR

		"Effective Cross-Section", b	
Neutron Group	Group Energy	Moderator	Bypass
Thermal	≤0.625 eV	1.0560	1.0903
Intermediate (I)	>0.625 eV - <1 MeV	0.0384	0.0423
Fast (F)	≥1 MeV	0.0479	0.0478
I+F	>0.625 eV	0.0408	0.0437

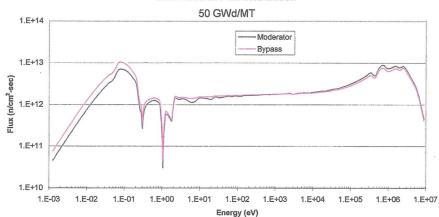
3.1 Source Term Calculation

Calculation of the cycle average ¹⁴C production rate per kilogram of water with 1.0 ppm dissolved nitrogen for each of the core regions is illustrated below. The number of ¹⁴N atoms per kilogram of water at 1 ppm nitrogen is 4.284E19. The 50 GWd/MT neutron fluxes in the three energy groups were used (see Table A-4) in this illustration.





BWR Neutron Flux Distribution



BWR C-14 Production from N-14(n,p)C-14 Reaction

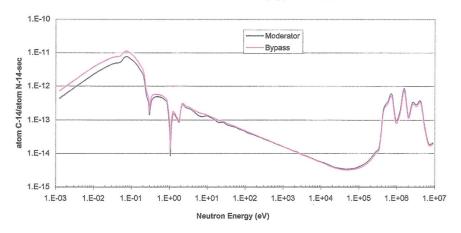


Figure A-4 ¹⁴N(n,p)¹⁴C Reaction Cross-Section, Reference BWR Neutron Flux Distribution at 50 GWd/MT and Calculated ¹⁴C Production Rate

3.1.1 Moderator Region

$$\frac{4.28 E19 \bullet (4.61 E13 \bullet 1.0560 + 1.36 E14 \bullet 0.0384 + 4.66 E13 \bullet 0.0474) \bullet 1E - 24 \bullet \ln 2}{3.7 E4 \bullet 5730 \bullet 365.25 \bullet 24 \bullet 3600}$$

$$= 2.489 E - 7 \mu Ci / \sec - kg - ppm - N$$

3.1.2 Bypass Region

$$\frac{4.28E19 \bullet (6.67E13 \bullet 1.0903 + 1.31E14 \bullet 0.0423 + 4.11E13 \bullet 0.0478) \bullet 1E - 24 \bullet \ln 2}{3.7E4 \bullet 5730 \bullet 365.25 \bullet 24 \bullet 3600}$$

$$= 3.557E - 7 \mu Ci / \sec - kg - ppm - N$$

3.1.3 Core Production Rate

The production rate at a 3579 MW_{th} GE-BWR with 748 fuel assemblies, 12,655 kg of water in the moderator region and 17,100 kg of water in the bypass or leakage region is as follows:

$$2.489E-7*12,655 + 3.557E-7*17,100 = 9.23E-3 \mu Ci/sec-ppm N or 0.29 Ci/yr-ppm N$$

Domestic BWRs are expected to have reactor coolant nitrogen concentrate <u>much less</u> than 1 ppm since all makeup water to the BWR during operation is deaerated in the condenser prior to entering the feedwater or CRD system. The concentration of nitrogen in the feedwater is expected to less than 5 ppb and much lower in the bulk reactor coolant.

4.0 Supplemental BWR Neutron Flux Data

Lin (1980) provided core average neutron flux data for GE BWRs in three energy groups (Table A-6). He stressed that when calculating the activation of fuel deposits, the neutron flux at the BWR fuel surface is higher by ~10-20%.

Table A-6 Core Average Neutron Flux in Various BWRs (Lin, 1980)

		Core Average Neutron Flux at 100% Power 10 ¹³ n/cm ² -sec		
BWR Type	Power Density w/cc	Thermal 0 – 0.625 eV	Epithermal 0.625 – 5.5E3 eV	Fast 5.5E3 - ∞ eV
BWR/2,3	41	3.51	4.81	10.9
BWR/4	51	4.11	6.10	13.8
BWR/5	50	3.89	5.59	13.5
BWR/6	54	4.22	6.47	14.7

A three group radial neutron flux distribution at the core axial mid-plane for a GE BWR is shown in Figure A-5 (Evans, et al., 1984). The approximate flux value at the core mid-plane is summarized in Table A-7.

Table A-7
Approximate BWR Neutron Flux at Core Mid-Plane from Figure A-5

	Neutron Energy	Neutron Flux, n/cm²-sec
Thermal	<0.6 eV	~3.4 E13
Epithermal	0.6 eV to 1 MeV	~4.0 E14
Fast	>1 MeV	~7.0 E13

Both sets of these supplemental data are consistent with the more detailed flux data used to derive values for the "effective" cross-sections.

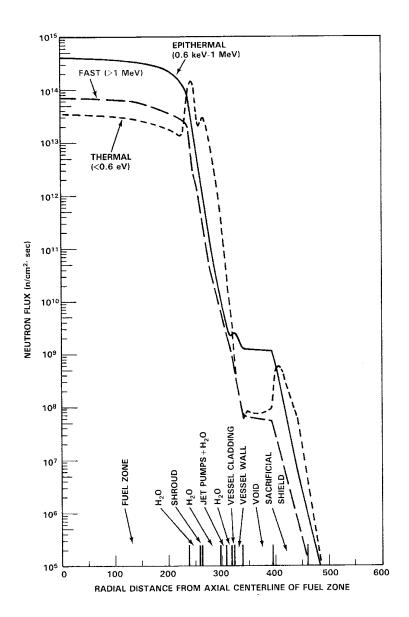


Figure A-5
Radial Three Group Neutron Flux Distribution at Core Axial Mid-Plane, ANISN Calculation for General Electric BWR (Evans, et al.,1984)

B

SITE SPECIFIC BWR SOURCE TERM CALCULATION

1.0 BWR

The three BWR units are General Electric BWR/4s originally rated at 3293 MW_{th} with Mark 1 containments. Units 2 and 3 have been up-rated to 3458 MW_{th}.

The utility staff provided the EPRI ¹⁴C project with summary data for nine operating cases (Table B-1). Three cases were generated at BOC, MOC and EOC with different core flow rates. Note that the coolant mass in the BWR is both flow rate and fuel cycle duration dependent. The active in-channel mass was calculated for each case by taking the in-channel nodal density times the nodal in-channel free volume for every node in the core. The reported bypass mass value is approximate, as the code did not edit out bypass fluid density by node. The utility used a representative density since liquid in this region remains sub-cooled. The bypass volume was that only inside the core, and the fluid flowing around the outside of the core inside the shroud was not considered.

The methodology described Appendix A was used to calculate the 14 C source terms for the nine cases. Calculations were done using EXCEL and are reported in Table B-2 for the two source term reactions. The results were averaged to estimate the cycle average 14 C source terms. The 17 O(n, α) 14 C reaction was the major production reaction yielding an annual production value of 13.7 Ci/yr. At a reactor coolant nitrogen concentration of 10 ppb (higher than expected in a domestic BWR reactor coolant), the production by the 14 N(n,p) 14 C reaction is only 0.002 Ci/yr.

Table B-1
BWR Power Station Core Flux and Reactor Coolant Mass Data

	Cycle Exposure	CAVEX	Flow	FLOW%	Active Flow Mass	Bypass Flow Mass	Total Mass of Coolant	Thermal Flux	Intermediate Flux	Fast Flux
Low	1500.0	17325.5	87.82	85.68%	26130.54952	24804.92071	50935.47023	3.46993E+13	1.38235E+14	5.1128E+13
Mid	2000.0	17825.6	91.74	89.50%	26426.04068	24804.92071	51230.96139	3.47874E+13	1.37539E+14	5.08707E+13
High	2500.0	18325.5	101.02	98.56%	26806.83219	24804.92071	51611.7529	3.48465E+13	1.36535E+14	5.04993E+13
Low	8000.0	23825.5	87.55	85.41%	27037.39123	24804.92071	51842.31194	3.63655E+13	1.4157E+14	5.22288E+13
Mid	8500.0	24325.5	91.46	89.23%	27009.11412	24804.92071	51814.03483	3.65203E+13	1.411E+14	5.20553E+13
High	9000.0	24825.6	100.75	98.29%	27231.56662	24804.92071	52036.48734	3.66613E+13	1.40139E+14	5.17008E+13
Low	13800.1	29625.7	91.39	89.16%	28080.94263	24804.92071	52885.86334	3.8481E+13	1.42504E+14	5.16433E+13
Mid	14200.0	30025.6	97.86	95.47%	2889180027	24804.92071	53696.72098	3.86556E+13	1.41169E+14	5.11593E+13
High	14450.0	30275.6	102.15	99.66%	29410.50821	24804.92071	54215.42892	3.87694E+13	1.4083E+14	5.08553E+13
	MWd/MTU	MWd/MTU	Mbm/hr	-	lbm	lbm	lbm	ф< 0.625 ev	0.625 eV < φ<1 MeV	φ>1MeV

Table B-2 BWR Power Station Calculated ¹⁴C Source Term

			Mod	derator Region		E	Bypass Region		
O-17(n,alpha)C-14 "effective	" cross-se	ction, b	0.1325	0.0238	0.1106	0.1386	0.0222	0.1106	
					Produc	tion Rate, uCi/sec			
	Cyde	Core Flow							Sum
	BOC	Low	5.644E-02	5.218E-02	9.353E-02	8.715E-02	4.461E-02	7.836E-02	0.4123
		Mid	5.733E-02	5.250E-02	9.408E-02	8.744E-02	4.438E-02	7.793E-02	0.4137
		High	5.838E-02	5.286E-02	9.470E-02	8.769E-02	4.405E-02	7.733E-02	0.4150
	MOC	Low	6.194E-02	5.527E-02	9.876E-02	9.115E-02	4.568E-02	7.995E-02	0.4328
		Mid	6.217E-02	5.503E-02	9.834E-02	9.148E-02	4.553E-02	7.969E-02	0.4322
		High	6.302E-02	5.510E-02	9.845E-02	9.188E-02	4.521E-02	7.912E-02	0.4328
	EOC	Low	6.883E-02	5.776E-02	1.013E-01	9.633E-02	4.597E-02	7.896E-02	0.4492
		Mid	7.139E-02	5.886E-02	1.032E-01	9.701E-02	4.552E-02	7.815E-02	0.4541
		High	7.304E-02	5.955E-02	1.043E-01	9.746E-02	4.525E-02	7.764E-02	0.4573
		Cyde Average:	0.0636	0.0555	0.0985	0.0920	0.0451	0.0786	0.4333
	%in Flu	ux/Core Region:	29.24	25.49	45.28	42.64	20.93	36.43	
	,	Sum: uCi/sec	0.4333						
		Ci/yr	13.67						
		-, ,.							
			Mod	derator Region		E	Bypass Region		
N-14(n,p)C-14 "effective" arc	ss-section	n, b	1.056	0.0384	0.0479	1.0903	0.0423	0.0478	
					Production	n Rate, uCi/sec-ppm	N		
	Cyde	Core Flow				, , , , , , , , , , , , , , , , , , , ,			Sum
	BOC	Low	1.517E-03	2.840E-04	1.366E-04	2.313E-03	2.867E-04	1.142E-04	0.0047
		Mid	1.541E-03	2.857E-04	1.374E-04	2.320E-03	2.853E-04	1.136E-04	0.0047
		High	1.570E-03	2.877E-04	1.383E-04	2.327E-03	2.832E-04	1.127E-04	0.0047
	MOC	Low	1.665E-03	3.008E-04	1.443E-04	2.419E-03	2.936E-04	1.166E-04	0.0049
		Mid	1.671E-03	2.995E-04	1.437E-04	2.428E-03	2.926E-04	1.162E-04	0.0050
		High	1.694E-03	2.999E-04	1.438E-04	2.438E-03	2.906E-04	1.154E-04	0.0050
	EOC	Low	1.851E-03	3.144E-04	1.480E-04	2.556E-03	2.954E-04	1.151E-04	0.0053
		Mid	1.919E-03	3.203E-04	1.507E-04	2.574E-03	2.926E-04	1.139E-04	0.0054
		High	1.964E-03	3.241E-04	1.524E-04	2.586E-03	2.908E-04	1.132E-04	0.0054
		Cyde Average:	0.00171	0.00030	0.00014	0.00244	0.00029	0.00011	0.00500
	%in Flu	ux/Core Region:	79.32	14.00	6.68	85.78	10.20	4.03	0.00500
Sur	n:	uCi/sec-ppmN	0.0050						
		Ci/yr-ppm N	0.16						

C PWR ¹⁴C SOURCE TERM CALCULATION

1.0 Core Flux Energy Distribution

Westinghouse (Secker (2009)) used the PARAGON fuel assembly lattice code to predict the neutron spectrum in 70 energy groups for Westinghouse 17x17 VANTAGE 5 fuel assemblies with IFM grids and ZIRLOTM cladding. The fuel assemblies contained 128 ZrB₂ IFBA burnable absorbers. This effort was done to support the calculation of fuel deposit activation products. The two enrichments which were considered (4.2 and 4.8 w/o). The plant parameters used in their calculations are given in Table C-1

Table C-1
Plant Parameters

Parameter	Value
Core Power	3565 MW _{th}
RCS Flow	396,084 gpm
Bypass Flow	5.0 %
Core Average Temperature	590.0 °F
Core Inlet Temperature	556.9 °F
Pressure	2250 psi

The outputs for one selected set of their neutron flux calculations are shown in Figure C-1. There is considerable structure in the neutron flux spectrum, and the flux in any energy group varies with burn-up.

For evaluating the PWR ¹⁴C source term, the data set shown in Figure C-1 was used to calculate the "effective cross-section" in selected neutron energy groups as a function of fuel burn-up. In these spectra, the group flux distributions which were considered are given in Table C-2.

The neutron flux data did not allow a cutoff at 1.0 MeV since the lower and upper energy boundaries for the energy group closest to the 1.0 MeV cutoff were 0.821 MeV and 1.35 MeV, respectively (average 1.087 MeV). The intermediate plus fast group was added to this evaluation since some plants only consider a two group neutron distribution.

Calculations of the effective cross sections for the two major ¹⁴C production reactions are given below.

Neutron Flux vs Energy 17x17 Westinghouse Fuel - 4.80 w/o

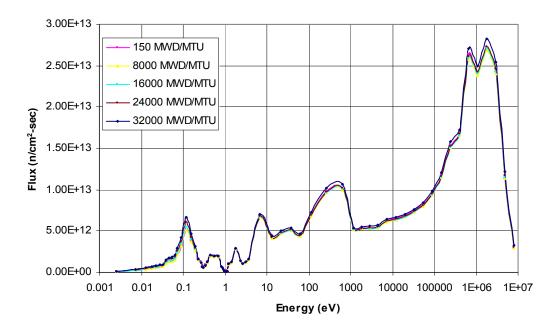


Figure C-1 Sample of the 70 Group Neutron Spectrum at Various Burnups – 4.20 w/o Fuel (Secker (2009))

Table C-2 PWR Flux Distribution

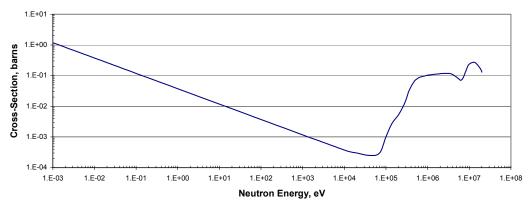
	Thermal <0.625 eV	Intermediate 0.625 eV to <1.353 MeV	Fast >1.353 MeV	Intermediate + Fast ≥0.625 eV
GWd/MTU Exposure		Neutron Flux (φ), n/cm²-sec	
0.15	3.552E13	2.286E14	6.569E13	2.943E14
8	3.506E13	2.237E14	6.519E13	2.889E14
16	3.672E13	2.244E14	6.583E13	2.902E14
24	3.968E13	2.269E14	6.676E13	2.937E14
32	4.368E13	2.355E14	6.916E13	3.047E14

2.0 Effective Cross Sections

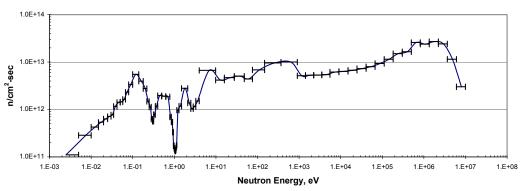
2.1 ¹⁷O(n,α)¹⁴C Reaction

The $^{17}O(n,\alpha)^{14}C$ reaction cross-section, the reference PWR neutron flux distribution at 16 GWd/MT and the ^{14}C production by this nuclear reaction are shown in Figure C-2. The x-axis error bars on the neutron flux and production rate graphs represent the width and height of each of the 70 group flux distribution values. The effective cross-sections for the four neutron energy groups are provided in Table C-3. The average of the five data sets is recommended for ^{14}C source term calculations.

O-17 (n,alpha) C-14 Production Reaction



Average Neutron Flux (16,000 MWD/MTU) - PWR



C-14 Production from the O-17(n,alpha)C-14 Reaction

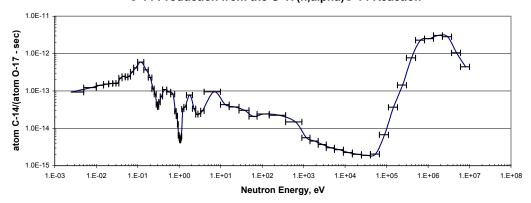


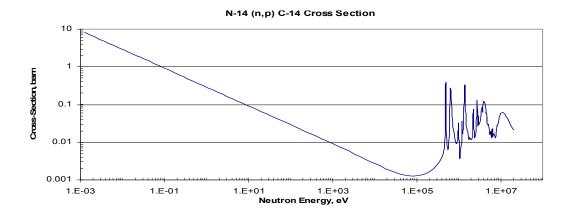
Figure C-2 $^{17}O(n,\alpha)^{14}C$ Reaction Cross-Section, Reference PWR Neutron Flux Distribution at 16 GWd/MT and Calculated ^{14}C Production

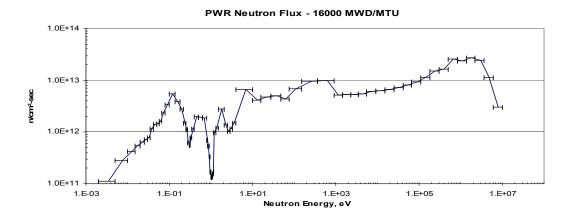
Table C-3 $^{17}\text{O}(\text{n},\alpha)^{14}\text{C}$ "Effective Cross-Section" Estimation in the PWR

	Thermal <0.625 eV	Intermediate 0.625 eV to <1.353 MeV	Fast >1.353 MeV	Intermediate + Fast, ≥0.625 eV	
GWd/MT Exposure	"Effective" Cross-Section (σ), barn				
0.15	0.1145	0.0294	0.1124	0.0476	
8	0.1194	0.0291	0.1124	0.0479	
16	0.1215	0.0291	0.1124	0.0480	
24	0.1236	0.0290	0.1123	0.0480	
32	0.1250	0.0289	0.1124	0.0478	
Average:	0.121	0.0291	0.1124	0.0479	

2.2 ¹⁴N(n,p)¹⁴C Reaction:

The $^{14}N(n,p)^{14}C$ reaction cross-section, the reference PWR neutron flux distribution at 16 GWd/MTU and the ^{14}C production by this nuclear reaction are shown in Figure C-3.





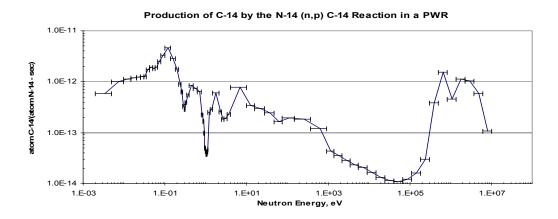


Figure C-3 ¹⁴N(n,p)¹⁴C Reaction Cross-Section, Reference PWR Neutron Flux Distribution at 16 GWd/MT and Calculated ¹⁴C Production

The effective cross-sections for the four neutron energy groups are provided in Table C-4. The average of the five data sets is recommended for ¹⁴C source term calculations.

Table C-4

14N(n,p)14C "Effective Cross-Section" Estimation in the PWR

	Thermal <0.625 eV	Intermediate 0.625 eV to <1.353 MeV	Fast >1.353 MeV	Intermediate + Fast, ≥0.625 eV
GWd/MT Exposure	"Effective" Cross-Section (σ), barn			
0.15	0.9188	0.0382	0.0435	0.0394
8	0.9354	0.0380	0.0435	0.0393
16	0.9520	0.0378	0.0436	0.0391
24	0.9682	0.0377	0.0436	0.0390
32	0.9797	0.0377	0.0436	0.0390
Average:	0.951	0.0379	0.0436	0.0392

3.0 Supplemental Neutron Flux Data

Several other sources of PWR fluxes were identified:

Table C-5
Typical Westinghouse PWR Neutron Flux Values (Table 4-2, EPRI-1009951(2004))

Core Average Neutron Flux	Flux, n/cm²-sec
Thermal (<0.65 eV)	5.0E13
Epithermal (>0.65 eV to <1MeV)	2.325E14
Fast (>1MeV)	7.75E13

Table C-6
Radial Three Group Neutron Flux Distribution at Core Axial Midplane, ANISN Calculation for a Typical Westinghouse PWR (see Figure C-4) (Evans, et al.(1984))

Core Average Neutron Flux	Flux, n/cm ² -sec
Thermal (<0.65 eV)	~5.5E13
Epithermal (>0.65 eV to <1MeV)	~2.0E14
Fast (>1MeV)	~7.0E13

These supplemental data are consistent with the more detailed flux data used to derive values for the "effective" cross-sections.

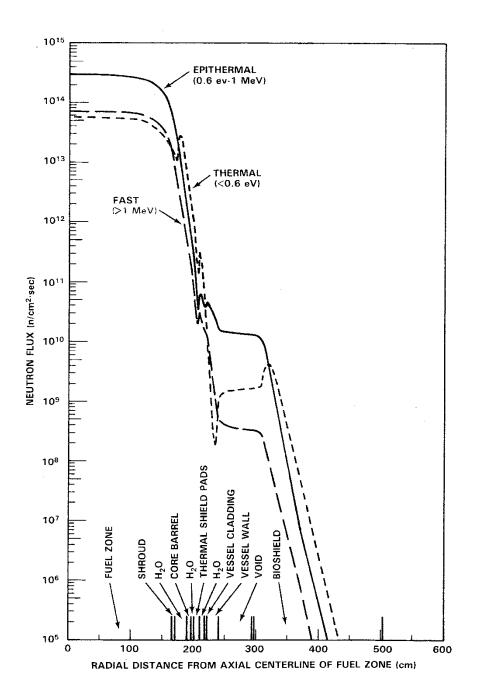


Figure C-4
Radial Three Group Neutron Flux Distribution at Core Axial Mid-Plane, ANISN Calculation for Westinghouse PWR (Evans, et al., 1984)

D

PWR SITE SPECIFIC SOURCE TERM CALCULATION

Summary of Appendix D PWR ¹⁴C Source Term Calculations

Westinghouse Reactors

			¹7O(n,α)¹4C	¹⁴ N(n,p) ¹⁴ C
	MW _{th}	Coolant Mass, kg	μCi/MW _{th} -h	μCi/MW _{th} -h-ppm N
W-A	3216	13,498	0.357	3.02E-3
W-B	3188	13,498	0.360	3.06E-3
W-C	3650	13,567	0.432	3.18E-3
W-D	1540	7,775	0.387	3.25E-3
W-E	1540	7,775	0.387	3.22E-3
W-F	3455	13,868	0.396	3.19E-3
W-G	3626	14,132	0.387	3.51E-3
		Average:	0.387±0.023	(3.20±0.16)E-3

	MW _{th}	Coolant Mass, kg	Ci/GW _e -y ^a	Ci/GW _e -y-ppm N ^a
W-A	3216	13,498	9.20	0.078
W-B	3188	13,498	9.28	0.079
W-C	3650	13,567	11.1	0.082
W-D	1540	7,775	10.0	0.084
W-E	1540	7,775	9.98	0.083
W-F	3455	13,868	10.2	0.082
W-G	3626	14,132	9.98	0.090
		Average:	9.96±0.63	0.083±0.004

a. Assumed 34% efficiency.

Combustion Engineering Reactors

			¹⁷ O(n,α) ¹⁴ C	¹⁴ N(n,p) ¹⁴ C
	MW _{th}	Coolant Mass, kg	μCi/MW _{th} -h	μCi/MW _{th} -h-ppm N
CE-A	2700	14,071	0.467	3.96E-3
			Ci/GW _e -y ^a	Ci/GW _e -y-ppm N ^e
			12.0	0.102
			μCi/MW _{th} -h	μCi/MW _{th} -h-ppm N
CE-B	3716	15,610	0.422	3.80E-3
			Ci/GW _e -y ^a	Ci/GW _e -y-ppm N ^a
			10.9	0.097

a. Assumed 34% efficiency.

1.0 Westinghouse - A (W-A)

W-A is a Westinghouse 4 loop PWR rated at 3216 MW_{th} with a net electrical rating of 1032 MW_{e} . The unit has 193 15x15 fuel assemblies, each with 204 fuel rods which have an OD of 0.422-inches and an active fuel length of 144 inches. There are 53 control rod assemblies. The coolant pressure is 2235 psi and the operating temperature 562 °F.

Table D-1 W-A Cycle 20 BOC and EOC Core Flux

	"Core Average" Neutron Flux, n/cm²-sec			
	≤0.625 eV	>0.625 eV		
BOC	3.18E13	2.78E14		
EOC	3.96E13	2.90E14		
Reaction	"Effective" Cross-Section ^a , b			
¹⁷ O(n,α) ¹⁴ C	0.121	0.0479		
¹⁴ N(n,p) ¹⁴ C	0.951	0.0392		

a. The effective cross section data were obtained from Tables C-3 and C-4.

The following equation was used to calculate the production rate in units of μ Ci/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

Production Rate (
$$\mu$$
Ci/sec - kg) =
$$\frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

$$\begin{array}{lll} N & = & 1.27E22 \ atoms \ ^{17}O/kg \ H_2O \\ \sigma_{th} & = & \text{``effective''} \ thermal \ cross-section, b \\ \phi_{th} & = & \text{thermal neutron flux } (\leq\!0.625 \ eV), \ n/cm^2\text{-sec} \\ \sigma_{i+f} & = & \text{``effective''} \ intermediate \ plus \ fast \ cross-section, b \\ \phi_{i+f} & = & \text{intermediate \ plus \ fast \ neutron \ flux } (>0.625 \ eV), \ n/cm^2\text{-sec} \\ 1.0E-24 & = & Conversion \ factor, \ 1.0E-24 \ cm^2/b \\ \lambda & = & \ ^{14}C \ decay \ constant, \ 3.833E-12/sec \\ 3.7E4 & = & Conversion \ factor, \ 3.7E4 \ d/sec-\mu Ci \\ \end{array}$$

BOC calculation

PR =
$$\frac{1.27 \text{ E } 22 \bullet [0.121 \bullet 3.18 \text{ E} 13 + 0.0479 \bullet 2.78 \text{ E} 14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E} 4}$$
= 2.258 E - 15 \(\mu \text{Ci} \) / sec - kg

EOC calculation

$$PR = \frac{1.27 \text{ E } 22 \bullet [0.121 \bullet 3.96 \text{ E} 13 + 0.0479 \bullet 2.90 \text{ E} 14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E} 4}$$
$$= 2.458 \text{ E} - 5\mu \text{Ci} / \text{sec} - \text{kg}$$

The cycle average 14 C production rates for the 17 O(n, α) 14 C reaction were as follows:

Table D-2 W-A Average Production Rates for the $^{17}{\rm O}({\rm n},\alpha)^{14}{\rm C}$ Reaction

		Production Rate, μCi/sec-kg	
вос		2.258E-5	
EOC		2.458E-5	
	Average:	2.36E-5	

If an "active core" mass of 13,498 kg is assumed, the total 14 C produced by the 17 O(n, α) 14 C reaction is:

$$2.36E-5 \mu \text{Ci/sec-kg} \cdot 13,498 \text{ kg} \cdot 3.156E7 \text{ sec/yr} = 1.01E7 \mu \text{Ci/yr} (10.1 \text{ Ci/yr})$$

or

$$[2.36\text{E}-5 \,\mu\text{Ci/sec-kg} \cdot 13,498 \,\text{kg} \cdot 3.6\text{E}3 \,\text{sec/hr}]/3216 \,\text{MW}_{\text{th}} = 0.357 \,\mu\text{Ci/MW}_{\text{th}}-\text{h}$$

or

0.357
$$\mu$$
Ci/MW_{th}-h • 8,766 h/yr • MW_{th}/0.34 MW_e • 1E3 MW_e/GW_e • Ci/1E6 μ Ci = 9.20 Ci/GW_e-yr

To calculate the production rate in units of $\mu \text{Ci/sec-kg-ppm N}$ for the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction, the following equation is employed:

Production Rate (
$$\mu$$
Ci/sec-kg) =
$$\frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

BOC calculation:

$$PR = \frac{4.28 \text{ E}19 \bullet [0.951 \bullet 3.18 \text{ E}13 + 0.0392 \bullet 2.78 \text{ E}14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E}4}$$
$$= 1.826 \text{ E} - 7 \mu \text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

PWR Site Specific Source Term Calculation

EOC calculation:

$$PR = \frac{4.28 \text{ E}19 \bullet [0.951 \bullet 3.96 \text{ E}13 + 0.0392 \bullet 2.90 \text{ E}14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E}4}$$
$$= 2.176 \text{ E} - 7 \mu \text{Ci} / \text{sec} - \text{kg}$$

The production rate are summarized below:

Table D-3
W-A Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

		Production Rate, μCi/sec-kg-ppm N	
вос		1.826E-7	
EOC		2.176E-7	
	Average:	2.00E-7	

If an "active core" mass of 13,498 kg is assumed, the total ¹⁴C produced by the ¹⁴N(n,p)¹⁴C reaction is:

2.00E-7
$$\mu$$
Ci/sec-kg-ppm N • 13,498 kg • 3.156E7 sec/yr = 8.52E4 μ Ci/yr-ppm N = 0.085 Ci/yr-ppm-N

or

[2.00E-7
$$\mu$$
Ci/sec-kg-ppm N • 13,498 kg • 3.6E3 sec/hr]/3216 MW_{th}
= 3.02E-3 μ Ci/MW_{th}-h–ppm N

or

3.02E-3
$$\mu$$
Ci/MW_{th}-h-ppm N • 8,766 h/yr • MW_{th}/0.34 MW_e • 1E3 MW_e/GW_e • Ci/1E6 μ Ci
$$= 0.078 \text{ Ci/GW}_{e}\text{-yr-ppm N}$$

2.0 Westinghouse-B (W-B)

W-B is a Westinghouse 4 loop PWR rated at 3188 MW_{th} with a net electrical rating of 1051 MW_e. The unit has 193 15x15 fuel assemblies, each with 204 fuel rods which have an OD of 0.42 inches and an active fuel length of 144 inches. There are 53 control rod assemblies. The coolant pressure is 2235 psi and the operating temperature 567 $^{\circ}$ F.

Table D-4
W-B BOC and EOC Core Flux

	"Core Average" Neutron Flux, n/cm²-sec		
	≤0.625 eV	>0.625 eV	
BOC	3.21E13	2.77E14	
EOC	3.99E13	2.89E14	
Reaction	"Effective" Cross-Section ^a , b		
¹⁷ O(n,α) ¹⁴ C	0.121	0.0479	
¹⁴ N(n,p) ¹⁴ C	0.951	0.0392	

a. The effective cross section data were obtained from Tables C-3 and C-4.

The following equation was used to calculate the production rate in units of μ Ci/sec-kg for the 17 O(n, α) 14 C reaction:

ProductionRate(
$$\mu$$
Ci/sec-kg) =
$$\frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

 $\begin{array}{lll} N & = & 1.27E22 \ atoms \ ^{17}O/kg \ H_2O \\ \sigma_{th} & = & \text{``effective''} \ thermal \ cross-section, b \\ \phi_{th} & = & \text{thermal neutron flux } (\leq 0.625 \ eV), \ n/cm^2-sec \\ \sigma_{i+f} & = & \text{``effective''} \ intermediate \ plus \ fast \ cross-section, b \\ \phi_{i+f} & = & \text{Intermediate plus fast neutron flux } (>0.625 \ eV), \ n/cm^2-sec \\ 1.0E-24 & = & \text{Conversion factor, } 1.0E-24 \ cm^2/b \\ \lambda & = & \ ^{14}C \ decay \ constant, \ 3.833E-12/sec \\ 3.7E4 & = & \text{Conversion factor, } 3.7E4 \ d/sec-\mu Ci \\ \end{array}$

BOC calculation:

$$PR = \frac{1.27 E 22 \bullet [0.121 \bullet 3.21E13 + 0.0479 \bullet 2.77E14] \bullet 1.0E - 24 \bullet 3.833 E - 12}{3.7E4}$$
$$= 2.257 E - 5\mu Ci / sec - kg$$

EOC calculation:

$$PR = \frac{1.27 \text{ E} 22 \bullet [0.121 \bullet 3.99 \text{ E}13 + 0.0479 \bullet 2.89 \text{ E}14] \bullet 1.0 \text{E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E}4}$$
$$= 2.456 \text{ E} - 5\mu\text{Ci} / \text{sec} - \text{kg}$$

The calculated cycle average ¹⁴C production rate using the above data is:

Table D-5 W-B Average Production Rates for the $^{17}\text{O}(\text{n},\alpha)^{14}\text{C}$ Reaction

		Production Rate, μCi/sec-kg	
вос		2.257E-5	
EOC		2.456E-5	
	Average:	2.36E-5	

If an "active core" mass of 13,498 kg is assumed, the total 14 C produced by the 17 O(n, α) 14 C reaction is:

$$2.36E-5 \mu \text{Ci/sec-kg} \cdot 13,498 \text{ kg} \cdot 3.156E7 \text{ sec/yr} = 1.01E6 \mu \text{Ci/yr} (10.1 \text{ Ci/yr})$$

or

$$[2.36E-5 \mu Ci/sec-kg \cdot 13,498 kg \cdot 3.6E3 sec/hr]/3188 MWth = 0.360 \mu Ci/MWth-h$$

or

0.360
$$\mu$$
Ci/MW_{th}-h • 8,766 h/yr • MW_{th}/0.34 MW_e • 1E3 MW_e/GW_e • Ci/1E6 μ Ci
$$= 9.28 \text{ Ci/GW}_{e}\text{-yr}$$

The following equation was used to calculate the production rate in units of $\mu \text{Ci/sec-kg-ppm N}$ for the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction.

$$ProductionRate(\mu Ci/sec-kg-ppm\cdot N\,) = \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 3.21E13 + 0.0392 \bullet 2.77E14] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 1.837E - 7\mu Ci / sec - kg - ppm \cdot N$$

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 3.99E13 + 0.0393 \bullet 2.89E14] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$

$$= 2.188E - 7\mu Ci / sec - kg$$

The calculated production rate using the above data is:

Table D-6
W-B Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

		Production Rate, μCi/sec-kg-ppm N	
вос		1.837E-7	
EOC		2.188E-7	
	Average:	2.01E-7	

The total ¹⁴C produced by the ¹⁴N(n,p)¹⁴C reaction is:

$$2.01E-7~\mu Ci/sec-kg-ppm~N~ \cdot 13,498~kg~ \cdot 3.156E7~sec/yr = 8.56E4~\mu Ci/yr-ppm~N$$
 or
$$= 0.086~Ci/yr-ppm-N$$
 or
$$[2.01E-7~\mu Ci/sec-kg-ppm~N~ \cdot 13,498~kg~ \cdot 3.6E3~sec/hr]/3188~MW_{th} =$$

$$= 3.06E-3~\mu Ci/MW_{th}-h-ppm~N$$

3.0 Combustion Engineering-A (CE-A)

CE-A is a Combustion Engineering (CE) PWR operating at a core nominal power of 2700 MW_{th}. The unit has 217 14x14 fuel assemblies with 176 fuel pins per assembly and 5 large guide tubes/instrument tubes. The volume of the coolant in the core is 5098.3 gallons (19299.2 liters). Other operating parameters for the unit are as follows:

Table D-7 CE-A System Parameters

	Cycle 20
Power Rating	2700 MW _{th}
System Pressure	2250 psia
Core Average Moderator Temperature, HZP	532 ºF
Core Inlet Moderator Temperature, HFP	545 ºF
Core Average Moderator Temperature, HFP	570.0 °F
Core Average Outlet Moderator Temperature, HFP	595.4 ºF
Vessel Average Temperature, HFP	569.5 °F
(T-Hot-HFP + T-Cold-HFP)/2	
Vessel Average Outlet Temperature, HFP	593.9 °F
Nominal Core Bypass Flow (%)	3.03

Site personnel provided the following neutron flux information for Cycle 20:

Table D-8 CE-A Cycle 20 Core Flux

		Thermal	Intermediate	Fast
		≤0.625 eV	0.625 eV to 1 MeV	>1 MeV
	Exposure, MWD/MTU	Neutron Flux, n/barn-sec		
вос	150	0.34194E-10	0.23830e-09	0.64388E-10
МОС	8000	0.36723E-10	0.24257E-09	0.65711E-10
EOC	15500	0.41123E-10	0.24465E-09	0.66194E-10
			Neutron Flux, n/cm²	-sec
вос	150	3.419E13	2.383E14	6.439E13
МОС	8000	3.672E13	2.426E14	6.571E13
EOC	15500	4.112E13	2.447E14	6.619E13

At a coolant volume of 19299.2 liters, reactor pressure of 2250 psia and core average moderator temperature of 570.0 °F, the coolant mass is 14,071 kg (density = 0.7291 kg/liter).

From Table C-3 of Appendix C, the cross section data are as follows:

Table D-9 "Effective" Cross-Section for the ¹⁷O(n,α) ¹⁴C Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.121
Intermediate	>0.625 eV - <1 MeV	0.0291
Fast	≥1 MeV	0.1124

The following equation was used to calculate the production rate in units of μ Ci/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

$$ProductionRate(\mu Ci/see-kg) = \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

N =
$$1.27E22$$
 atoms $^{17}O/kg$ H₂O
 σ_{th} = "effective" thermal cross-section, b
 ϕ_{th} = thermal neutron flux (≤ 0.625 eV), n/cm²-sec
 σ_{i} = "effective" intermediate cross-section, b
 ϕ_{i} = intermediate neutron flux (0.625 eV to 1 MeV), n/cm²-sec
 σ_{f} = "effective" fast cross-section, b
 ϕ_{f} = fast neutron flux (> 0.625 eV), n/cm²-sec
1.0E-24 = conversion factor, 1.0E-24 cm²/b
 λ = ^{14}C decay constant, 3.833E-12/sec
3.7E4 = conversion factor, 3.7E4 d/sec-μCi

BOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.419E13 + 0.0291 \bullet 2.383E14 + 0.1124 \bullet 6.439E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$

$$= 2.409E - 5\mu Ci/sec - kg$$

MOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.672E13 + 0.0291 \bullet 2.426E14 + 0.1124 \bullet 6.571E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 2.485E - 5\mu \text{Ci/sec-kg}$$

EOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 4.112E13 + 0.0291 \bullet 2.447E14 + 0.1124 \bullet 6.619E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 2.570E - 5\mu Ci/sec - kg$$

The calculated cycle average ¹⁴C production rate using the above data is:

Table D-10 CE-A Cycle 20 Average Production Rates for the 17 O(n, α) 14 C Reaction

	Production Rate, μCi/sec-kg
BOC	2.409E-5
мос	2.485E-5
EOC	2.570E-5
Average:	2.49E-5

For an "active core mass" of 14,071 kg, the total 14C produced by the 17O(n, α)14C reaction is:

$$2.49E-5 \mu Ci/sec-kg \cdot 14,071 kg \cdot 3.156E7 sec/yr = 1.11E7 \mu Ci/yr (11.1 Ci/yr)$$

The following "effective" cross-sections and equation were used to calculate the production rate in units of μ Ci/sec-kg-ppm N for the 14 N(n,p) 14 C reaction.

Table D-11 "Effective" Cross-Section for the ¹⁴N(n,p) ¹⁴C Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.951
Intermediate	>0.625 eV - <1 MeV	0.0379
Fast	≥1 MeV	0.0436

$$ProductiorRate(\mu Ci/see \cdot kg - ppm \cdot N) = \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7F4}$$

where:

 $N = 4.284E19 \text{ atoms} ^{14}\text{N/kg-ppm N}$

 σ_{th} = "effective" thermal cross-section, b

 φ_{th} = thermal neutron flux, n/cm²-sec

 σ_i = "effective" intermediate cross-section, b

 φ_i = intermediate neutron flux (0.625 eV to 1 MeV), n/cm²-sec

 σ_f = "effective" fast cross-section, b

 φ_f = fast neutron flux, n/cm²-sec

 $1.0E-24 = conversion factor, 1.0E-24 cm^2/b$

 $\lambda = {}^{14}\text{C decay constant}, 3.833\text{E}-12/\text{sec}$

 $3.7E4 = \text{conversion factor}, 3.7E4 \text{ d/sec-}\mu\text{Ci}$

BOC calculation:

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 3.419E13 + 0.0379 \bullet 2.383E14 + 0.0436 \bullet 6.439E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 1.968E - 7\mu\text{Ci} / \text{sec-kg-ppm} \cdot \text{N}$$

MOC calculation:

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 3.672E13 + 0.0379 \bullet 2.426E14 + 0.0436 \bullet 6.571E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 2.085E - 7\mu Ci / sec - kg - ppm \cdot N$$

EOC calculation:

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 4.112E13 + 0.0379 \bullet 2.447E14 + 0.0436 \bullet 6.619E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 2.275E - 7\mu Ci / sec - kg - ppm \cdot N$$

The calculated production rate using the above data is:

Table D-12 CE-A Cycle 20 Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

	Production Rate, μCi/sec-kg-ppm N	
BOC	1.968E-7	
мос	2.085E-7	
EOC	2.275E-7	
Average:	2.11E-7	

The total ¹⁴C produced by the ¹⁴N(n,p)¹⁴C reaction is:

2.11E-7
$$\mu$$
Ci/sec-kg-ppm N • 14,071 kg • 3.156E7 sec/yr = 9.37E4 μ Ci/yr-ppm N = 0.094 Ci/yr-ppm-N

or

[2.11E-7
$$\mu$$
Ci/sec-kg-ppm N • 14,071 kg • 3.6E3 sec/hr]/2700 MW_{th} =
= 3.96E-3 μ Ci/MW_{th}-h–ppm N

4.0 Westinghouse-C (W-C)

W-C is a Westinghouse (W) four-loop PWR operating at a core nominal power of 3650 MW_{th}. The unit has 193 17x17 fuel assemblies with 264 fuel pins per assembly, 24 guide tubes and 1 instrument tube. The volume of the coolant in the core is 5083.5 gallons (19243.1 liters). Other operating parameters for the unit are:

Table D-13 W-C System Parameters

	Cycle 14
Power Rating	3650 MW _{th}
System Pressure	2250 psia
Core Average Moderator Temperature, HZP	557 ºF
Core Inlet Moderator Temperature, HFP	556.6 ºF
Core Average Moderator Temperature, HFP	590.2 ºF
Core Average Outlet Moderator Temperature, HFP	620.4 ºF
Vessel Average Temperature, HFP (T-Hot-HFP + T-Cold-HFP)/2	587.0 ºF
Vessel Average Outlet Temperature, HFP	617.5 °F

Site personnel provided the following Cycle 14 neutron fluxes data:

Table D-14 W-C Cycle 14 Core Flux

		Thermal ≤0.625 eV	Intermediate 0.625 eV to 1 MeV	Fast >1 MeV
	Exposure, MWD/MTU	ı	Neutron Flux, n/barn-se	c
вос	150	0.36833E-10	0.32629E-09	0.88555E-10
MOC	10000	0.37923E-10	0.33434E-09	0.90916E-10
EOC	20000	0.42468E-10	0.33763E-09	0.91889E-10
			Neutron Flux, n/cm²-sec	•
вос	150	3.683E13	3.263E14	8.856E13
MOC	10000	3.792E13	3.343E14	9.092E13
EOC	20000	4.247E13	3.376E14	9.189E13

At a coolant volume of 19243.1 liters, a reactor pressure of 2250 psia and a core average coolant temperature of 590.2 °F, the coolant mass is 13,567 kg (density = 0.7051 kg/liter).

Table D-15 "Effective" Cross-Section for the ¹⁷O(n,α) ¹⁴C Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.121
Intermediate	>0.625 eV - <1 MeV	0.0291
Fast	≥1 MeV	0.1124

The following equation was used to calculate the production rate in units of μ Ci/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

$$Productio\Re ate(\mu Ci/see\,kg) = \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

1.27E22 atoms $^{17}O/kg$ H₂O N "effective" thermal cross-section, b σ_{th} thermal neutron flux ($\leq 0.625 \text{ eV}$), n/cm²-sec ϕ_{th} "effective" intermediate cross-section, b σ_{i} intermediate neutron flux (0.625 eV to 1 MeV), n/cm²-sec ϕ_i "effective" fast cross-section, b $\sigma_{\rm f}$ fast neutron flux (>0.625 eV), n/cm²-sec 1.0E-24 Conversion factor, 1.0E-24 cm²/b ¹⁴C decay constant, 3.833E-12/sec 3.7E4 Conversion factor, 3.7E4 d/sec-µCi

BOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.683E13 + 0.0291 \bullet 3.263E14 + 0.1124 \bullet 8.856E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 3.145E - 5\mu Ci / sec - kg$$

MOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.792E13 + 0.0291 \bullet 3.343E14 + 0.1124 \bullet 9.092E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 3.228E - 5\mu Ci / sec - kg$$

EOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 4.247E13 + 0.0291 \bullet 3.376E14 + 0.1124 \bullet 9.189E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 3.327E - 5\mu\text{Ci}/\text{sec-kg}$$

The calculated cycle average ¹⁴C production rate using the above data is:

Table D-16 W-C Cycle 14 Average Production Rates for the $^{17}\text{O}(\text{n},\alpha)^{14}\text{C}$ Reaction

		Production Rate, μCi/sec-kg
вос		3.145E-5
MOC		3.228E-5
EOC		3.327E-5
	Average:	3.23E-5

For an "active core mass" of 13,567 kg, the total 14 C produced by the 17 O(n, α) 14 C reaction is:

$$3.23E-5 \mu \text{Ci/sec-kg} \cdot 13,567 \text{ kg} \cdot 3.156E7 \text{ sec/yr} = 1.38E7 \mu \text{Ci/yr} (13.8 \text{ Ci/yr})$$

or

$$[3.23E-5 \mu Ci/sec-kg \cdot 13,567 kg \cdot 3.6E3 sec/hr]/3650 MWth = 0.432 \mu Ci/MWth-h$$

The following "effective" cross-sections and equation were used to calculate the production rate in units of μ Ci/sec-kg-ppm N for the 14 N(n,p) 14 C reaction.

Table D-17 "Effective" Cross-Section for the ¹⁴N(n,p)¹⁴C Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.951
Intermediate	>0.625 eV - <1 MeV	0.0379
Fast	≥1 MeV	0.0436

$$ProductiorRate(\mu Ci/see-kg-ppm\cdot N) = \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

```
\begin{array}{lll} N & = & 4.284E19 \ atoms \ ^{14}N/kg\text{-ppm N} \\ \sigma_{th} & = & \text{``effective''' thermal cross-section, b} \\ \phi_{th} & = & \text{thermal neutron flux, n/cm}^2\text{-sec} \\ \sigma_i & = & \text{``effective''' intermediate cross-section, b} \\ \phi_i & = & \text{intermediate neutron flux (0.625 eV to 1 MeV), n/cm}^2\text{-sec} \\ \sigma_f & = & \text{``effective''' fast cross-section, b} \\ \phi_f & = & \text{fast neutron flux, n/cm}^2\text{-sec} \\ 1.0E-24 & = & \text{conversion factor, 1.0E-24 cm}^2/b \\ \lambda & = & \text{$^{14}$C decay constant, 3.833E-12/sec} \\ 3.7E4 & = & \text{conversion factor, 3.7E4 d/sec-$\mu$Ci} \\ \end{array}
```

BOC calculation:

$$PR = \frac{4.284 \, E19 \bullet [0.951 \bullet 3.683 \, E13 + 0.0379 \bullet 3.263 \, E14 + 0.0436 \bullet 8.856 \, E13] \bullet 1.0E - 24 \bullet 3.833 \, E - 12}{3.7E4} \\ = 2.275 \, E - 7 \mu Ci \, / \, sec - kg - ppm \cdot N$$

MOC calculation:

$$PR = \frac{4.284 \, E19 \bullet [0.951 \bullet 3.792 \, E13 + 0.0379 \bullet 3.343 \, E14 + 0.0436 \bullet 9.092 \, E13] \bullet 1.0E - 24 \bullet 3.833 \, E - 12}{3.7E4} \\ = 2.339 \, E - 7 \mu Ci \, / \, sec - \, kg - ppm \cdot N$$

EOC calculation:

$$PR = \frac{4.284 \, \text{E}19 \bullet [0.951 \bullet 4.247 \, \text{E}13 + 0.0379 \bullet 3.376 \, \text{E}14 + 0.0436 \bullet 9.189 \, \text{E}13] \bullet 1.0E - 24 \bullet 3.833 \, \text{E} - 12}{3.7E4} \\ = 2.538 \, \text{E} - 7 \mu \text{Ci} \, / \, \text{sec} - \, \text{kg} - \text{ppm} \cdot \text{N}$$

The calculated production rate using the above data is:

Table D-18 W-C Cycle 14 Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

		Production Rate, μCi/sec-kg-ppm N
вос		2.275E-7
MOC		2.339E-7
EOC		2.538E-7
	Average:	2.38E-7

The total ¹⁴C produced by the ¹⁴N(n,p)¹⁴C reaction is:

2.38E-7
$$\mu$$
Ci/sec-kg-ppm N • 13,567 kg • 3.156E7 sec/yr = 1.02E5 μ Ci/yr-ppm N = 0.102 Ci/yr-ppm-N

or

[2.38E-7
$$\mu$$
Ci/sec-kg-ppm N • 13,567 kg • 3.6E3 sec/hr]/3650 MW_{th}
= 3.18E-3 μ Ci/MW_{th}-h–ppm N

5.0 Westinghouse-D (W-D) and Westinghouse-E (W-E)

W-D and W-E are two loop Westinghouse PWRs each licensed to operate at 1540 MW_{th}. The plants operate at an average temperature of 570 °F at 2250 psia. The following cycle core average neutron flux data were provided by the site at BOC, MOC and EOC. The n/barn-sec data were converted to n/cm^2 -sec by dividing by 1.0E-24 cm²/barn.

Table D-19
W-D Core Flux Distribution

		n/barn-sec		
	MWD/MTU	≤0.625 eV	>0.625 eV	
вос	150	2.98E-11	2.55E-10	
MOC	8000	3.15E-11	2.59E-10	
EOC	14000	3.41E-11	2.61E-10	
		n/cm²-sec		
	MWD/MTU	≤0.625 eV	>0.625 eV	
вос	150	2.98E13	2.55E14	
MOC	8000	3.15E13	2.59E14	
EOC	14000	3.41E13	2.61E14	

Table D-20 W-E Core Flux Distribution

		n/bar	n-sec
	MWD/MTU	≤0.625 eV	>0.625 eV
BOC	150	2.95E-11	2.55E-10
MOC	8000	3.09E-11	2.59E-10
EOC	14000	3.34E-11	2.61E-10
		n/cm²-sec	
	MWD/MTU	≤0.625 eV	>0.625 eV
BOC	150	2.95E13	2.55E14
MOC	8000	3.09E13	2.59E14
EOC	14000	3.34E13	2.61E14

Each reactor has $121 \ 14x14$ fuel assemblies with a rod OD of 0.422 inches and an active fuel length of 144 inches. An estimate of $362 \ \text{ft}^3$ of water in the active fuel region was provided.

Using a specific volume of 0.02112 ft³/lb, the active coolant volume was estimated to be 17,140 lbs or 7775 kg. Cross sections used in the calculations of ¹⁴C production rates were as follows:

Table D-21 Effective Cross Sections

	"Effective" Cross-Section, b		
Reaction	≤0.625 eV	>0.625 eV	
¹⁷ O(n,α) ¹⁴ C	0.121	0.0479	
¹⁴ N(n,p) ¹⁴ C	0.951	0.0392	

5.1 Westinghouse-D (W-D)

The following equation was used to calculate the production rate for W-D in units of μ Ci/sec-kg for the 17 O(n, α) 14 C reaction.

$$Production \ Rate \ (\mu Ci/sec - kg) \ = \ \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

 $\begin{array}{lll} N & = & 1.27E22 \ atoms \ ^{17}O/kg \ H_2O \\ \sigma_{th} & = & \text{``effective'' thermal cross-section, b} \\ \phi_{th} & = & \text{thermal neutron flux } (\leq 0.625 \ eV), \ n/cm^2-sec \\ \sigma_{i+f} & = & \text{``effective'' intermediate plus fast cross-section, b} \\ \phi_{i+f} & = & \text{Intermediate plus fast neutron flux } (>0.625 \ eV), \ n/cm^2-sec \\ 1.0E-24 & = & \text{Conversion factor, } 1.0E-24 \ cm^2/b \\ \lambda & = & \ ^{14}C \ decay \ constant, \ 3.833E-12/sec \\ 3.7E4 & = & \text{Conversion factor, } 3.7E4 \ d/sec-\mu Ci \\ \end{array}$

BOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 2.98E13 + 0.0479 \bullet 2.55E14] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4} = 2.081E - 5\mu Ci / sec - kg$$

MOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.15E13 + 0.0479 \bullet 2.59E14] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4} = 2.134E - 5\mu Ci / sec - kg$$

EOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.41E13 + 0.0479 \bullet 2.61E14] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4} = 2.188E - 5\mu Ci / sec - kg$$

The calculated cycle average ¹⁴C production rate using the above data is:

Table D-22 W-D Average Production Rates for the ¹⁷O(n,α)¹⁴C Reaction

	Production Rate, μCi/sec-kg
BOC	2.081E-5
Mid-Cycle	2.134E-5
EOC	2.188E-5
Average:	2.13E-5

The total 14 C produced by the 17 O(n, α) 14 C reaction is:

$$2.13E-5 \mu Ci/sec-kg \cdot 7.775E3 kg \cdot 3.156E7 sec/yr = 5.23E6 \mu Ci/yr (5.23 Ci/yr)$$

or

$$[2.13E-5 \mu Ci/sec-kg \cdot 7.775E3 kg \cdot 3.6E3 sec/hr]/1540 MWth = 0.387 \mu Ci/MWth-h$$

The following equation was used to calculate the production rate in units of μ Ci/sec-kg-ppm N for the 14 N(n,p) 14 C reaction.

$$Production \ Rate \ (\mu Ci/sec - kg - ppm \cdot N) \ = \ \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

BOC calculation:

$$PR = \frac{4.284 \text{ E19} \cdot [0.951 \cdot 2.98 \text{ E13} + 0.0392 \cdot 2.55 \text{ E14}] \cdot 1.0 \text{ E} - 24 \cdot 3.833 \text{ E} - 12}{3.7 \text{ E4}}$$

$$= 1.701 \text{ E} - 7 \mu \text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

MOC calculation:

$$PR = \frac{4.284 \text{ E}19 \bullet [0.951 \bullet 3.15 \text{ E}13 + 0.0392 \bullet 2.59 \text{ E}14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E}4}$$
$$= 1.780 \text{ E} - 7 \mu \text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

EOC calculation:

$$PR = \frac{4.284 \text{ E19} \bullet [0.951 \bullet 3.41 \text{ E13} + 0.0392 \bullet 2.61 \text{ E14}] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E} 4}$$

$$= 1.893 \text{ E} - 7 \mu \text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

The calculated production rate using the above data is:

Table D-23
W-D Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

	Production Rate, μCi/sec-kg-ppm N
BOC	1.701E-7
Mid-Cycle	1.780E-7
EOC	1.893E-7
Average:	1.79E-7

The total ¹⁴C produced by the ¹⁴N(n,p)¹⁴C reaction is:

1.79E-7
$$\mu$$
Ci/sec-kg-ppm N • 7.775E3 kg • 3.156E7 sec/yr = 4.39E4 μ Ci/yr-ppm N = 0.044 Ci/yr-ppm-N

or

[1.79E-7
$$\mu$$
Ci/sec-kg-ppm N • 7.775E3 kg • 3.6E3 sec/hr]/1540 MW_{th}

$$= 3.25\text{E-3} \ \mu\text{Ci/MW}_{th}\text{-h-ppm N}$$

5.2 Westinghouse-E (W-E)

The following equation was used to calculate the production rate in units of μ Ci/sec-kg for the 17 O(n, α) 14 C reaction.

Production Rate (
$$\mu$$
Ci/sec - kg) =
$$\frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

N	=	$1.27E22 \text{ atoms} ^{17}O/\text{kg} \text{H}_2\text{O}$
σ_{th}	=	"effective" thermal cross-section, b
ϕ_{th}	=	Thermal neutron flux ($\leq 0.625 \text{ eV}$), n/cm ² -sec
σ_{i+f}	=	"effective" intermediate plus fast cross-section, b
ϕ_{i+f}	=	Intermediate plus fast neutron flux (>0.625 eV), n/cm ² -sec
1.0E-24	=	Conversion factor, 1.0E-24 cm ² /b
λ	=	¹⁴ C decay constant, 3.833E-12/sec
3.7E4	=	Conversion factor, 3.7E4 d/sec-μCi

PWR Site Specific Source Term Calculation

BOC calculation:

$$PR = \frac{1.27 \text{ E } 22 \bullet [0.121 \bullet 2.95 \text{ E} 13 + 0.0479 \bullet 2.55 \text{ E} 14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E} 4}$$

$$= 2.077 \text{ E} - 5\mu \text{Ci} / \text{sec} - \text{kg}$$

MOC calculation:

$$PR = \frac{1.27 \text{ E } 22 \bullet [0.121 \bullet 3.09 \text{ E } 13 + 0.0479 \bullet 2.59 \text{ E } 14] \bullet 1.0 \text{ E } -24 \bullet 3.833 \text{ E } -12}{3.7 \text{ E } 4}$$
$$= 2.124 \text{ E } -5 \mu \text{Ci / sec } -\text{kg}$$

EOC calculation:

$$PR = \frac{1.27 \text{ E } 22 \bullet [0.121 \bullet 3.34 \text{ E} 13 + 0.0479 \bullet 2.61 \text{ E} 14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E} 4}$$
$$= 2.177 \text{ E} - 5 \mu \text{Ci / sec} - \text{kg}$$

The calculated cycle average ¹⁴C production rate using the above data is:

Table D-24 W-E Average Production Rates for the $^{17}O(n,\alpha)^{14}C$ Reaction

	Production Rate, μCi/sec-kg
BOC	2.077E-5
Mid-Cycle	2.124E-5
EOC	2.177E-5
Average:	2.13E-5

The total 14 C produced by the 17 O(n, α) 14 C reaction is:

$$2.13E-5 \mu Ci/sec-kg \cdot 7.775E3 kg \cdot 3.156E7 sec/yr = 5.23E6 \mu Ci/yr (5.23 Ci/yr)$$

or

$$[2.13E-5 \ \mu Ci/sec-kg \bullet 7.775E3 \ kg \bullet 3.6E3 \ sec/hr]/1540 \ MW_{th} = 0.387 \ \mu Ci/MW_{th}-h$$

The following equation was used to calculate the production rate in units of μ Ci/sec-kg-ppm N for the 14 N(n,p) 14 C reaction.

$$Production \ Rate \ (\mu Ci/sec - kg - ppm \cdot N) \ = \ \frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

BOC calculation:

$$PR = \frac{4.284 \text{ E}19 \bullet [0.951 \bullet 2.95 \text{ E}13 + 0.0392 \bullet 2.55 \text{ E}14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E}4}$$
$$= 1.689 \text{ E} - 7\mu\text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

MOC calculation:

$$PR = \frac{4.284 \text{ E19} \cdot [0.951 \cdot 3.09 \text{ E13} + 0.0392 \cdot 2.59 \text{ E14}] \cdot 1.0 \text{ E} - 24 \cdot 3.833 \text{ E} - 12}{3.7 \text{ E4}}$$

$$= 1.755 \text{ E} - 7\mu \text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

EOC calculation:

$$PR = \frac{4.284 \text{ E}19 \bullet [0.951 \bullet 3.34 \text{ E}13 + 0.0392 \bullet 2.61 \text{ E}14] \bullet 1.0 \text{ E} - 24 \bullet 3.833 \text{ E} - 12}{3.7 \text{ E}4}$$

$$= 1.864 \text{ E} - 7 \mu \text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

The calculated production rate using the above data is:

Table D-25
W-E Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

	Production Rate, μCi/sec-kg-ppm N	
BOC	1.689E-7	
Mid-Cycle	1.755E-7	
EOC	1.864E-7	
Average:	1.77E-7	

The total ¹⁴C produced by the ¹⁴N(n,p)¹⁴C reaction is:

1.77E-7
$$\mu$$
Ci/sec-kg-ppm N • 7.775E3 kg • 3.156E7 sec/yr = 4.34E4 μ Ci/yr-ppm N = 0.043 Ci/yr-ppm-N

or

[1.77E-7
$$\mu$$
Ci/sec-kg-ppm N • 7.775E3 kg • 3.6E3 sec/hr]/1540 MW_{th} =
= 3.22E-3 μ Ci/MW_{th}-h-ppm N

6.0 Westinghouse-F (W-F)

W-F is a Westinghouse 4 loop PWR rated at 3455 MW $_{th}$ with a net electrical rating of 1126 MW $_{e}$. The unit has 193 17x17 fuel assemblies, each with 264 fuel rods which have an OD of 0.360-inches and an active fuel length of 144-inches. There are 53 control rod assemblies. The coolant pressure is 2235 psi and the operating temperature 567 °F.

The following core average neutron fluxes were calculated at the BOC, MOC and EOC for three neutron energy groups.

Table D-26
W-F Core Flux Distribution

	Thermal	Intermediate	Fast
	E≤0.625 eV	>0.625 eV <e<1 mev<="" th=""><th>E≥1 MeV</th></e<1>	E≥1 MeV
	(Core Average Flux, n/cm²-sec	
вос	3.6728E+13	2.2268E+14	8.1527E+13
MOC	3.8275E+13	2.2927E+14	8.3939E+13
EOC	4.2959E+13	2.3365E+14	8.5545E+13

Table D-27 "Effective" Cross-Section for the $^{17}{\rm O}({\rm n},\alpha)^{14}{\rm C}$ Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.121
Intermediate	>0.625 eV - <1 MeV	0.0291
Fast	≥1 MeV	0.1124

The following equation was used to calculate the production rate in units of $\mu \text{Ci/sec-kg}$ for the $^{17}\text{O}(n,\alpha)^{14}\text{C}$ reaction.

Production Rate (
$$\mu$$
Ci/sec - kg) =
$$\frac{N \bullet [\sigma_{th} \bullet \varphi_{th} + \sigma_{i} \bullet \varphi_{i} + \sigma_{f} \bullet \varphi_{f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

1.27E22 atoms $^{17}O/kg$ H₂O Ν "effective" thermal cross-section, b σ_{th} thermal neutron flux ($\leq 0.625 \text{ eV}$), n/cm²-sec ϕ_{th} "effective" intermediate cross-section, b σ_{i} intermediate neutron flux (0.625 eV to 1 MeV), n/cm²-sec "effective" fast cross-section, b $\sigma_{\rm f}$ fast neutron flux (>0.625 eV), n/cm²-sec Conversion factor, 1.0E-24 cm²/b 1.0E-24 ¹⁴C decay constant, 3.833E-12/sec Conversion factor, 3.7E4 d/sec-µCi 3.7E4

BOC calculation:

$$PR = \frac{1.27 \, \text{E} \, 22 \, \bullet \, [0.121 \, \bullet \, 3.672 \, \text{E} \, 13 \, + \, 0.0291 \, \bullet \, 2.2267 \, \text{E} \, 14 \, + \, 0.1124 \, \bullet \, 8.1527 \, \text{E} \, 13] \, \bullet \, 1.0E \, - \, 24 \, \bullet \, 3.833 \, \text{E} \, - \, 12}{3.7E4} \\ = 2.643 \, \text{E} \, - \, 5 \mu \text{Ci} \, / \, \text{sec} \, - \, \text{kg}$$

MOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.8275E13 + 0.0291 \bullet 2.2927E14 + 0.1124 \bullet 8.8939E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 2.728E - 5\mu Ci / sec - kg$$

EOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 4.2959E13 + 0.0291 \bullet 2.3365E14 + 0.1124 \bullet 8.85545E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 2.843E - 5\mu Ci / sec - kg$$

The calculated cycle average ¹⁴C production rate using the above data is:

Table D-28 W-F Average Production Rates for the $^{17}\text{O}(\text{n},\alpha)^{14}\text{C}$ Reaction

	Production Rate, μCi/sec-kg
BOC	2.643E-5
мос	2.728E-5
EOC	2.843E-5
Average:	2.74E-5

Assuming an "active core mass" of 30,574 # (13,868 kg), the total 14 C produced by the 17 O(n, α) 14 C reaction is:

 $2.74E-5 \mu \text{Ci/sec-kg} \cdot 13,868 \text{ kg} \cdot 3.156E7 \text{ sec/yr} = 1.20E7 \mu \text{Ci/yr} (12.0 \text{ Ci/yr})$

or

$$[2.74E-5 \mu Ci/sec-kg \cdot 13,868 kg \cdot 3.6E3 sec/hr]/3455 MW_{th} = 0.396 \mu Ci/MW_{th}-h$$

The following "effective" cross-sections and equation were used to calculate the 14 C production rate in units of μ Ci/sec-kg-ppm N for the 14 N(n,p) 14 C reaction.

Table D-29 "Effective" Cross-Section for the ¹⁴N(n,p)¹⁴C Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.951
Intermediate	>0.625 eV - <1 MeV	0.0379
Fast	≥1 MeV	0.0436

Production Rate (
$$\mu$$
Ci/sec - kg - ppm · N) =
$$\frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i} \bullet \phi_{i} + \sigma_{f} \bullet \phi_{f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

BOC calculation:

$$PR = \frac{4.284 \,\text{E19} \bullet [0.951 \bullet 3.673 \,\text{E13} + 0.0379 \bullet 3.227 \,\text{E14} + 0.0436 \bullet 8.8153 \,\text{E13}] \bullet 1.0E - 24 \bullet 3.833 \,\text{E} - 12}{3.7E4}$$
$$= 2.083 \,\text{E} - 7\mu \,\text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

MOC calculation:

$$PR = \frac{4.284 \,\text{E19} \,\bullet [0.951 \,\bullet 3.828 \,\text{E13} + 0.0379 \,\bullet 2.293 \,\text{E14} + 0.0436 \,\bullet 8.394 \,\text{E13}] \,\bullet 1.0E - 24 \,\bullet 3.833 \,\text{E} - 12}{3.7E4} \\ = 2.164 \,\text{E} - 7 \mu \text{Ci} \,/ \,\text{sec} - \,\text{kg} - \,\text{ppm} \,\cdot \,\text{N}$$

EOC calculation:

$$PR = \frac{4.284 \,\text{E19} \cdot [0.951 \cdot 4.296 \,\text{E13} + 0.0379 \cdot 2.337 \,\text{E14} + 0.0436 \cdot 8.8555 \,\text{E13}] \cdot 1.0E - 24 \cdot 3.833 \,\text{E} - 12}{3.7E4}$$
$$= 2.372 \,\text{E} - 7\mu \,\text{Ci} \,/\, \text{sec} - \,\text{kg} - \,\text{ppm} \cdot \text{N}$$

The calculated production rate using the above data is:

Table D-30 W-F Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

		Production Rate, μCi/sec-kg-ppm N
вос		2.083E-7
MOC		2.164E-7
EOC		2.372E-7
	Average:	2.21E-7

The total ¹⁴C produced by the ¹⁴N(n,p)¹⁴C reaction is:

2.21E-7
$$\mu$$
Ci/sec-kg-ppm N • 13,868 kg • 3.156E7 sec/yr = 9.67E4 μ Ci/yr-ppm N = 0.097 Ci/yr-ppm-N

or

[2.21E-7
$$\mu$$
Ci/sec-kg-ppm N • 13,868 kg • 3.6E3 sec/hr]/3455 MW_{th}
= 3.19E-3 μ Ci/MW_{th}-h-ppm N

7.0 Combustion Engineering-B (CE-B) Calculation

CE-B is a Combustion Engineering (CE) two-loop PWR operating at a core nominal power of 3716 MW_{th} . The unit has 217 16x16 fuel assemblies with 236 fuel pins per assembly. The active core water mass has been calculated to be 15,610 kg at a core average coolant temperature of 574.06 °F.

Site personnel provided the following neutron flux data:

Table D-31 CE-B Core Average Neutron Flux

	Thermal	Intermediate	Fast
	≤0.625 eV	0.625 eV to 1 MeV	>1 MeV
		Neutron Flux, n/cm²-sec	•
вос	4.33E13	2.10E14	7.71E13
MOC	4.61E13	2.18E14	8.06E13
EOC	5.10E13	2.33E14	8.59E13

 $^{^{17}\}mathrm{O}(n,\alpha)^{14}\mathrm{C}$ Reaction Source Term Calculation:

Table D-32 "Effective" Cross-Section for the ¹⁷O(n,α)¹⁴C Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.121
Intermediate	>0.625 eV - <1 MeV	0.0291
Fast	≥1 MeV	0.1124

The following equation was used to calculate the production rate in units of μ Ci/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

Production Rate (
$$\mu$$
Ci/sec - kg) =
$$\frac{N \bullet [\sigma_{th} \bullet \varphi_{th} + \sigma_i \bullet \varphi_i + \sigma_f \bullet \varphi_f] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

$$\begin{array}{lll} N & = & 1.27E22 \ atoms \ ^{17}O/kg \ H_2O \\ \sigma_{th} & = & \text{``effective''' thermal cross-section, b} \\ \phi_{th} & = & \text{thermal neutron flux } (\leq 0.625 \ eV), \ n/cm^2-sec \\ \sigma_i & = & \text{``effective''' intermediate cross-section, b} \\ \phi_i & = & \text{Intermediate neutron flux } (0.625 \ eV \ to \ 1 \ MeV), \ n/cm^2-sec \\ \sigma_f & = & \text{``effective''' fast cross-section, b} \\ \phi_f & = & \text{fast neutron flux } (>0.625 \ eV), \ n/cm^2-sec \\ 1.0E-24 & = & \text{Conversion factor, } 1.0E-24 \ cm^2/b \\ \lambda & = & \ ^{14}C \ decay \ constant, \ 3.833E-12/sec \\ 3.7E4 & = & \text{Conversion factor, } 3.7E4 \ d/sec-\mu Ci \\ \end{array}$$

BOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 4.33E13 + 0.0291 \bullet 2.10E14 + 0.1124 \bullet 7.71E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 2.633E - 5\mu\text{Ci/sec-kg}$$

MOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 4.61E13 + 0.0291 \bullet 2.18E14 + 0.1124 \bullet 8.06E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$
$$= 2.760E - 5\mu\text{Ci/sec-kg}$$

EOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 5.10E13 + 0.0291 \bullet 2.33E14 + 0.1124 \bullet 8.59E13] \bullet 1.0E - 24 \bullet 3.833E - 12}{3.7E4}$$

$$= 2.974E - 5\mu\text{Ci/sec-kg}$$

The calculated cycle average ¹⁴C production rate using the above data is:

Table D-33 CE-B Average Production Rates for the $^{17}{\rm O}({\rm n},a)^{14}{\rm C}$ Reaction

	Production Rate, μCi/sec-kg
BOC	2.633E-5
мос	2.760E-5
EOC	2.974E-5
Average:	2.79E-5

For an "active core mass" of 15,610 kg, the total 14 C produced by the 17 O(n, α) 14 C reaction is:

$$2.79E-5 \mu Ci/sec-kg \cdot 15,610 kg \cdot 3.156E7 sec/yr = 1.37E7 \mu Ci/yr (13.7 Ci/yr)$$

or

$$[2.79E-5 \mu Ci/sec-kg \cdot 15,610 kg \cdot 3.6E3 sec/hr]/3716 MW_{th} = 0.422 \mu Ci/MW_{th}-h$$

or at an assuming 34% efficiency

$$0.422 \mu \text{Ci/MW}_{\text{th}}$$
-h • MW_{th}/0.34 MW_e • 8,766 h/yr = 1.09E4 $\mu \text{Ci/MW}_{\text{e}}$ -yr (10.9 Ci/GW_e-yr)

The following "effective" cross-sections and equation were used to calculate the production rate in units of $\mu Ci/sec-kg-ppm\ N$ for the $^{14}N(n,p)^{14}C$ reaction.

Table D-34 "Effective" Cross-Section for the ¹⁴N(n,p)¹⁴C Reaction in the PWR

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.951
Intermediate	>0.625 eV - <1 MeV	0.0379
Fast	≥1 MeV	0.0436

Production Rate (
$$\mu$$
Ci/sec - kg - ppm · N) =
$$\frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i} \bullet \phi_{i} + \sigma_{f} \bullet \phi_{f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

¹⁴N(n,p)¹⁴C Reaction Source Term Calculation:

where:

```
4.284E19 atoms <sup>14</sup>N/kg-ppm N
N
                        "effective" thermal cross-section, b
\sigma_{th}
                        thermal neutron flux, n/cm<sup>2</sup>-sec
\phi_{th}
                        "effective" intermediate cross-section, b
                        Intermediate neutron flux (0.625 eV to 1 MeV), n/cm<sup>2</sup>-sec
                        "effective" fast cross-section, b
\sigma_{\rm f}
                        fast neutron flux, n/cm<sup>2</sup>-sec
1.0E-24
                        conversion factor, 1.0E-24 cm<sup>2</sup>/b
                        <sup>14</sup>C decay constant, 3.833E-12/sec
                        conversion factor, 3.7E4 d/sec-μCi
3.7E4
```

BOC calculation:

$$PR = \frac{4.284 \text{ E19} \cdot [0.951 \cdot 4.33 \text{ E13} + 0.0379 \cdot 2.10 \text{ E14} + 0.0436 \cdot 7.71 \text{ E13}] \cdot 1.0 \text{E} - 24 \cdot 3.833 \text{ E} - 12}{3.7 \text{ E4}}$$

$$= 2.330 \text{ E} - 7\mu\text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

MOC calculation:

PR =
$$\frac{4.284 \text{ E19} \cdot [0.951 \cdot 4.61 \text{ E13} + 0.0379 \cdot 2.18 \text{ E14} + 0.0436 \cdot 8.06 \text{ E13}] \cdot 1.0 \text{ E} - 24 \cdot 3.833 \text{ E} - 12}{3.7 \text{ E} 4}$$
= 2.468 E - 7 μ Ci / sec - kg - ppm · N

EOC calculation:

$$PR = \frac{4.284 \text{ E19} \cdot [0.951 \cdot 5.10 \text{ E13} + 0.0379 \cdot 2.33 \text{ E14} + 0.0436 \cdot 8.591 \text{ E13}] \cdot 1.0 \text{ E} - 24 \cdot 3.833 \text{ E} - 12}{3.7 \text{ E} 4}$$

$$= 2.711 \text{ E} - 7 \mu \text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

The calculated production rate using the above data is:

Table D-35
CE-B Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

		Production Rate, μCi/sec-kg-ppm N	
вос		2.330E-7	
MOC		2.468E-7	
EOC		2.711E-7	
	Average:	2.50E-7	

The total ¹⁴C produced by the ¹⁴N(n,p) ¹⁴C reaction is:

2.50E-7
$$\mu$$
Ci/sec-kg-ppm N • 15,610 kg • 3.156E7 sec/yr = 1.23E5 μ Ci/yr-ppm N = 0.123 Ci/yr-ppm N

or

[2.50E-7
$$\mu$$
Ci/sec-kg-ppm N • 15,610 kg • 3.6E3 sec/hr]/3716 MW_{th}
= 3.78E-3 μ Ci/MW_{th}-h-ppm N

8.0 Westinghouse-G (W-G) Calculation

W-G is a Westinghouse 4 loop PWR rated at 3625.6 MW_{th} with a net electrical rating of 1233 MW_e (at an assumed efficiency of 34%). The unit has 193 17x17 OFA fuel assemblies, each with 264 fuel rods which have an OD of 0.36 inches and an active fuel length Of 144 inches. The coolant pressure is 2250 psi and the core average operating temperature is 589.5 °F.

Table D-36 W-G BOL, MOL and EOL Core Flux

	"Core Average" Neutron Flux, n/cm²-sec	
	≤0.625 eV	>0.625 eV
вос	4.202E13	3.148E14
MOC	4.518E13	3.237E14
EOC	5.069E13	3.260E14
Reaction	"Effective" Cross-Section ^a , b	
¹⁷ O(n,α) ¹⁴ C	0.121	0.0479
¹⁴ N(n,p) ¹⁴ C	0.951	0.0392

a. The effective cross section data were obtained from Tables C-3 and C-4.

The following equation was used to calculate the production rate in units of μ Ci/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction:

Production Rate (
$$\mu$$
Ci/sec - kg) =
$$\frac{N \bullet [\sigma_{th} \bullet \phi_{th} + \sigma_{i+f} \bullet \phi_{i+f}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

N 1.27E22 atoms $^{17}O/kg$ H₂O "effective" thermal cross-section, b σ_{th} thermal neutron flux ($\leq 0.625 \text{ eV}$), n/cm²-sec ϕ_{th} "effective" intermediate plus fast cross-section, b σ_{i+f} Intermediate plus fast neutron flux (>0.625 eV), n/cm²-sec ϕ_{i+f} Conversion factor, 1.0E-24 cm²/b 1.0E-24 ¹⁴C decay constant, 3.833E-12/sec Λ 3.7E4 Conversion factor, 3.7E4 d/sec-µCi

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

PWR Site Specific Source Term Calculation

BOC calculation:

$$PR = \frac{1.27 \text{ E } 22 \bullet [0.121 \bullet 4.202 \text{ E } 13 + 0.0479 \bullet 3.148 \text{ E } 14] \bullet 1.0 \text{ E } -24 \bullet 3.833 \text{ E } -12}{3.7 \text{ E } 4}$$

$$= 2.653 \text{ E } -5 \mu \text{Ci} / \text{sec} - \text{kg}$$

MOC calculation:

$$PR = \frac{1.27 \text{ E } 22 \bullet [0.121 \bullet 4.518 \text{ E } 13 + 0.0479 \bullet 3.237 \text{ E } 14] \bullet 1.0 \text{ E } -24 \bullet 3.833 \text{ E } -12}{3.7 \text{ E } 4}$$

$$= 2.759 \text{ E } -5 \mu \text{Ci} / \text{sec} - \text{kg}$$

EOC calculation:

$$PR = \frac{1.27 \text{ E } 22 \bullet [0.121 \bullet 5.069 \text{ E } 13 + 0.0479 \bullet 3.260 \text{ E } 14] \bullet 1.0 \text{ E } - 24 \bullet 3.833 \text{ E } - 12}{3.7 \text{ E } 4}$$

$$= 2.861 \text{ E } - 5 \mu \text{Ci / sec } - \text{kg}$$

The calculated cycle average ¹⁴C production rate using the above data is:

Table D-37 W-G Average Production Rates for the $^{17}\text{O}(\text{n},\alpha)^{14}\text{C}$ Reaction

	Production Rate, μCi/sec-kg		
BOC	2.653E-5		
мос	2.759E-5		
EOC	2.861E-5		
Average:	2.76E-5		

The active core volume was calculated to be 710.9 ft³. At the specified core average temperature of 589.5°F and operating 2250 psi, the "active core" mass is \sim 14,132 kg (ρ = 0.702 kg/L assumed).

The total 14 C produced by the 17 O(n, α) 14 C reaction is:

$$2.76E-5 \mu Ci/sec-kg \cdot 14,132 kg \cdot 3.156E7 sec/yr = 12.3E6 \mu Ci/yr (12.3 Ci/yr)$$

or

$$[2.76E-5 \mu Ci/sec-kg \cdot 14,132 kg \cdot 3.6E3 sec/hr]/3625.6 MW_{th} = 0.387 \mu Ci/MW_{th}-h$$

or

0.387
$$\mu$$
Ci/MW_{th}-h • 8.766E3 h/yr • MW_{th}/0.34 MW_e • 1E3 MW_e/GW_e • Ci/1E6 μ Ci
$$= 9.98 \text{ Ci/GW}_{e}\text{-yr}$$

¹⁴N(n,p)¹⁴C Reaction:

The following equation was used to calculate the production rate in units of $\mu \text{Ci/sec-kg-ppm N}$ for the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction.

Production Rate (
$$\mu \text{Ci/sec} - \text{kg} - \text{ppm} \cdot \text{N}$$
) =
$$\frac{N \bullet [\sigma_{\text{th}} \bullet \phi_{\text{th}} + \sigma_{\text{i+f}} \bullet \phi_{\text{i+f}}] \bullet 1.0E - 24 \bullet \lambda}{3.7E4}$$

where:

BOC calculation:

$$PR = \frac{4.284 \text{ E}19 \bullet [0.951 \bullet 4.202 \text{ E}13 + 0.0392 \bullet 3.148 \text{ E}14] \bullet 1.0\text{E} - 24 \bullet 3.833 \text{ E} - 12}{3.7\text{E}4}$$
$$= 2.321 \text{ E} - 7\mu\text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

MOC calculation:

$$PR = \frac{4.284 \text{ E}19 \bullet [0.951 \bullet 4.518 \text{ E}13 + 0.0392 \bullet 3.237 \text{ E}14] \bullet 1.0E - 24 \bullet 3.833 \text{ E} - 12}{3.7E4}$$
$$= 2.470 \text{ E} - 7\mu\text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

EOC calculation:

$$PR = \frac{4.284 \text{ E}19 \bullet [0.951 \bullet 5.069 \text{ E}13 + 0.0392 \bullet 3.260 \text{ E}14] \bullet 1.0E - 24 \bullet 3.833 \text{ E} - 12}{3.7E4}$$
$$= 2.707 \text{ E} - 7\mu\text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

The calculated production rate using the above data is:

Table D-38
W-G Average Production Rates for the ¹⁴N(n,p)¹⁴C Reaction

	Production Rate, μCi/sec-kg-ppm N	ı
BOC	2.321E-7	
MOC	2.470E-7	
EOC	2.707E-7	
Avei	age: 2.50E-7	

The total 14 C produced by the $^{14}N(n,p)^{14}$ C reaction is:

2.50E-7
$$\mu$$
Ci/sec-kg-ppm N • 14,132 kg • 3.156E7 sec/yr = 1.11E5 μ Ci/yr-ppm N = 0.111 Ci/yr-ppm-N

or

[2.50E-7
$$\mu$$
Ci/sec-kg-ppm N • 14,132 kg • 3.6E3 sec/hr]/3625.6 MW_{th} =
$$= 3.51\text{E-3} \ \mu\text{Ci/MW}_{th}\text{-h-ppm N}$$

$$= 0.090 \ \text{Ci/GW}_{e}\text{-yr-ppm N}$$

E

PWR PRIMARY WATER NITROGEN CONCENTRATION

1.0 General Considerations

A methodology for calculating the dissolved nitrogen gas concentration in the reactor coolant from measurements of percent nitrogen in the volume control tank (VCT) gas phase, VCT pressure and VCT temperature is given below. The VCT contains liquid water and vapor spaces. Gases, such as hydrogen, nitrogen, argon, helium and water vapor contribute to the total gas phase pressure according to their individual volume fractions.

As described by Henry's Law, the concentration of a dissolved gas in a liquid (expressed as mole fraction) is a function of the partial pressure of the gas above the liquid:

$$P_i = H \cdot X_i$$
 Eqn. E-1

where

H = Henry's Law Constant, atm/mole fraction

P_i = Partial Pressure of Species <u>i</u>, atm

X_i = Mole Fraction of Species i in the Liquid Phase

For an ideal gas, the volume fraction of each constituent equals its partial pressure fraction.

The Henry's Law constants are functions of temperature [E-1]. The Henry's Law Constant for N₂ can be calculated from the following relations based on the values developed from Reference E-1:

$$H = -11.672 \, (^{\circ}C)^2 + 1897.3 \, (^{\circ}C) + 46710 \, at \, 20^{\circ}C \, to \, 50^{\circ}C$$
 Eqn. E-2

or

$$H = -3.6024 (^{\circ}F)2 + 1284.6 (^{\circ}F) + 9290.5 \text{ at } 68^{\circ}F \text{ to } 122^{\circ}F$$
 Eqn. E-3

2.0 Sample Calculation

Assuming a VCT pressure of 23 psig, a temperature of 35°C (93°F) and a VCT nitrogen volume percent of ~12%, the estimated partial pressure of nitrogen is:

$$P(N_2) = 0.12 \bullet \frac{23 \text{ psig} + 14.7 \text{ psig}}{14.7 \text{ psig/atm}} = 0.308 \text{ atm}$$

From Equation E-2, Henry's Law Constant at 35°C is calculated to be:

$$H = -11.672(35)^2 + 1897.3(35) + 46710 = 9.882E4$$
 atm/mole-fraction

The mole fraction of nitrogen in the VCT liquid phase is:

$$X(N_2) = P(N_2)/H = 0.308$$
 atm/9.882E4 atm/mole-fraction = 3.12E-6

The mole fraction of 3.12E-6 is equivalent to \sim 4.85 ppm N₂, e.g.:

$$\frac{3.12E - 6 \text{ mole N}_2}{\text{mole H}_2 O} \bullet \frac{\text{mole H}_2 O}{18.02 \text{ g H}_2 O} \bullet \frac{28.01 \text{ g N}_2}{\text{mole N}_2} = 4.85E - 6 \text{ g N/g H}_2 O = 4.85 \text{ ppm}$$

Note that the total nitrogen concentration in the VCT liquid is that due to the dissolved nitrogen gas and that due to ammonia in solution, and that the total nitrogen concentration must be used to evaluate the ¹⁴C source term. For example, if the coolant contained 1 ppm NH₃, the total nitrogen concentration would be

Total Nitrogen =
$$4.85 + 1 \left(\frac{14}{17} \right) = 5.67 \text{ ppm}$$

3.0 Effects of VCT Operating Practices

The approach to VCT venting, i.e., continuous or intermittent, can have a significant impact on the gas phase nitrogen concentration and thus the dissolved nitrogen concentration in the primary coolant. Continuous venting has the following effects:

- The concentration of ¹⁴C (primarily methane) in the RCS and the VCT will be reduced.
- The formation of ¹⁴C from the activation of nitrogen will be reduced. The concentration of nitrogen in the VCT, the activation of dissolved nitrogen in the coolant and the formation of ammonia will be reduced.
- The steady state concentration of hydrogen in the VCT will be increased.
- Major ¹⁴C gaseous releases will be from the gas treatment systems.
- For PWRs without recombiners in the gas treatment system, a greater fraction of the released ¹⁴C will be organic which will reduce local environmental radiation exposures.
- The gas phase concentration of ¹⁴C in containment will be reduced, and less release will occur via the containment venting pathway.

Reference

E-1 IAWPS Guidelines on the Equilibrium Constant for the Distribution of Gaseous Solutes between Steam and Water, London, United Kingdom, September 1996.

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