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# 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 ($^{14}C$) 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 $^{14}C$ discharge can be estimated by sample measurements or by use of a normalized $^{14}C$ 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 $^{14}C$ source term and release estimation.

## Objective

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

## Approach

This report reviews $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ in both types of reactors is discussed, and $^{14}C$ generation rates for each type of reactor are calculated based on plant-specific parameters. A brief summary of $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ from the $^{14}N(n,p)$ $^{14}C$ reaction also contributes to the PWR $^{14}C$ 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 $^{14}C$ produced in a BWR is released in a gaseous form by the off-gas system, primarily in the form of $^{14}CO_2$.
- Gaseous release of $^{14}C$ 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 $^{14}CO_2$.
- A method was developed to allow PWR and BWR personnel to calculate a site-specific $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ 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 |
| | | |
| µCi/min | Ci/yr | 0.5260 |
| | GBq/yr | 19.46 |
| | | |
| µCi/$MW_{th}$-sec | Ci/$MW_{th}$-yr | 31.56 |
| | GBq/$MW_{th}$-yr | 1167.6 |
| | 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/$GW_{th}$-yr | 0.34 [(a)] |
| | | |
| 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%.

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# 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 ($^{14}C$) 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 $^{14}C$ is a principal gaseous effluent, and if so, report the amount of $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ generation and releases.
- Evaluation of techniques to monitor and estimate $^{14}C$ release rates via liquid, gaseous and solid waste streams.
- Compilation of available information on $^{14}C$ 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.

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 $^{14}C$ in the environment and describes the nuclear reactions leading to its production. A summary of the $^{14}C$ measurement and reporting requirements is provided.

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

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

- **Section 5 Measurement in the Nuclear Power Industry**

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

# 2
# BACKGROUND

## 2.1 Sources of $^{14}C$ in the Environment

### *2.1.1 $^{14}C$ Production Reactions*

$^{14}C$ has a half-life of 5700±30 years, and 100 percent of its decay is by beta emission to $^{14}N$. 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 $^{17}O(n,\alpha)^{14}C$ and $^{14}N(n,p)^{14}C$ 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 |
| $^{17}O(n,\alpha)^{14}C$ | 0.038 |
| $^{13}C(n,\gamma)^{14}C$ | 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.

b. Hayes, 1977.

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 $B_4C$ 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 $^{14}C$

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.

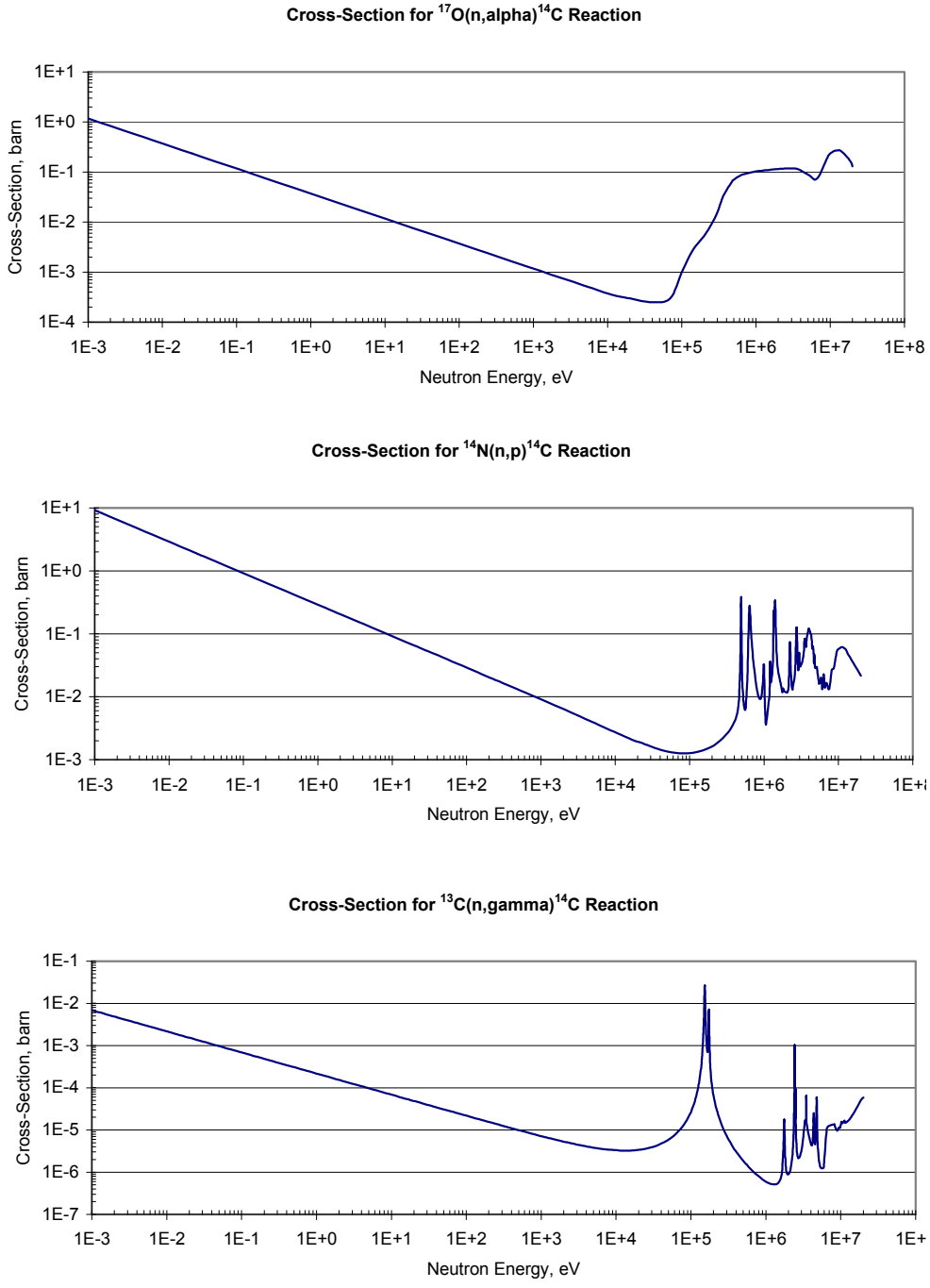



**Figure 2-1**
**Reaction Cross-Sections (ENDF)**

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)**

| $^{17}O(n,\alpha)^{14}C$ | 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 (20ºC) | 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 20ºC while LWRs operate between 285ºC and 310ºC. 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 $^{14}C$ source terms in both reactor types.

### *2.1.2 Environmental Sources of $^{14}C$*

There are numerous sources of carbon-14 dioxide ($^{14}CO_{2)}$ 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 $^{14}N(n,p)^{14}C$ reaction at a rate of 2.5 atoms/sec-cm$^2$ of earth surface (Suess; 1958). Using an earth surface area of 5.1E18 cm$^2$, 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 $^{14}CO_2$ introduced into the biosphere (Suess; 1958).

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

| Exchangeable Reservoir | Carbon Content, g/cm$^2$ 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 $^{14}C$ atoms (8297 years). The average residence time for $^{14}C$ 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 $^{14}C$ to the atmosphere. It has been estimated that each megaton of total nuclear energy (fission + fusion) produces (2±1)E26 atoms of $^{14}C$ 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 $^{14}C$ or 9.51E6 Ci. Since that time, additional atmospheric testing has further increased the $^{14}C$ inventory. To put the weapons related $^{14}C$ 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 $^{14}C$ 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 $^{14}CO_2$ 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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ in the body prior to nuclear testing was reported at 1.25 mrad per person annum. The estimated dose attributable to $^{14}C$ releases from nuclear power plants is quite small.

### *2.1.3 Chemical Forms of $^{14}C$ Produced in the LWR Primary Coolant*

The stable chemical compounds of $^{14}C$ 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 $^{14}C$ In-Core Reactions**

| Single Carbon Species | |
|---|---|
| $CO_2$ | Carbon Dioxide |
| CO | Carbon Monoxide |
| HCOOH | Formic Acid |
| $H_2C=O$ | Formaldehyde |
| $CH_3OH$ | Methanol |
| $CH_4$ | Methane |

Rosset (1994) calculated the redox properties of $^{14}C$ at 300ºC. 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 ~5.62 and at a potential between ~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, $H_2CO_3$, $HCO_2^-$, $HCO_3^-$ and $CH_3OH$ 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 $HCO_2^-$ and $CH_3OH$ are the expected stable products. Although these types of potential-pH diagrams provide some indication of the expected chemical forms of $^{14}C$ 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 $^{14}C$ in either gaseous or liquid effluents. Since $^{14}C$ 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 $^{14}C$ has a minimal impact on the world inventory of $^{14}C$. 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 (~3200 $MW_{th}$ PWR), with 2.6 Curie per year as $CO_2$ (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 $^{14}C$ as a potential principal radionuclide for effluent monitoring. The document states "radioactive effluents from commercial nuclear power plants have decreased to a point that $^{14}C$ is likely to be a principal radionuclide in gaseous effluents". It further states "because the dose contribution of $^{14}C$ from liquid radioactive waste is much less than that contributed by gaseous radioactive waste, evaluation of $^{14}C$ in liquid radioactive waste is not required". It requests that licensees evaluate whether $^{14}C$ 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 $^{14}C$ in gaseous waste discharged can be estimated by:

1. Sample measurements,
2. Use of normalized $^{14}C$ 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)

If sampling is performed, "the sampling frequency may be adjusted to that interval that allows adequate measurement and reporting of effluents." If estimating $^{14}C$ 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 $^{14}C$ or to include $^{14}C$ 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 $^{14}C$ in solid waste forms and as a separate category, $^{14}C$ 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 long-lived species.

Because $^{14}C$ is a difficult to measure nuclide, 10 CFR Part 61 permits the use of scaling factors in which the $^{14}C$ 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 $^{14}C/^{60}Co$ scaling factors to determine the $^{14}C$ content of their solid radioactive wastes. The two exceptions are India which does not include $^{14}C$ 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 $^{14}C/^{60}Co$ scaling factor because of the wide variation in the $^{14}C/^{60}Co$ ratios. Most of the solid waste $^{14}C$ is found with the primary coolant ion exchange resins, and they estimate the total accumulation of $^{14}C$ on these resins based on the $MW_e$ generated over the life of the resins, the established production rate of $^{14}C$ 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 $^{14}C$ 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 $^{14}CO_2$, $^{14}CO$, $H^{14}CO_2H$, $H_2{}^{14}CO$, 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 $^{14}C$ Source Term Estimations and Measurements

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

- 3.9E4 kg, mass of water in the reactor core
- 1.3E22 atoms of $^{17}O$/kg of natural water
- 0.80 plant capacity factor
- 1.03E-22 Ci/atom, specific activity of $^{14}C$
- 2.4E-25 $cm^2$, thermal neutron cross section for $^{17}O$, and
- 3E13 n/$cm^2$-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 $^{17}O(n,\alpha)^{14}C$ 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/$cm^2$-sec).

Fowler, et al., (1976) reported on the health considerations of $^{14}C$ discharges from the nuclear power industry. This report was later updated by Fowler and Nelson (1979). In their 1976 report, the production rates of $^{14}C$ 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 $MW_{th}$ BWR/6) was 39.5 metric ton (MT).

**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]** |
| BWR Fuel | O-17 | 4 | 8.4 | 10.9 | | 2.7 |
| | N-14 | 18 | 12.9 | 21.2 | | 10.9 |
| | Total | 22 | 21.3 | 32.1 | 20[b] | 13.6 |
| BWR Coolant | O-17 | 8.9 | 9.9 | 11.5 | | |
| | N-14 | 0.26 | 1.3 | -- | | |
| | 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.

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 in liquid-metal cooled fast breeder reactors. The calculated value for the $^{17}O(n,\alpha)^{14}C$ 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 $^{17}O(n,\alpha)^{14}C$ reaction.

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

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 $^{14}C$ 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 ±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[a] | BWR[b] |
|---|---|
| $^{17}O(n,\alpha)^{14}C$ | 14.5 Ci/GW(e)-yr |
| $^{14}N(n,p)^{14}C$ | 0.29 Ci/GW(e)-yr/ppm |
| $^{13}C(n,\gamma)^{14}C$ | 0.027 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 for the $^{13}C(n,\gamma)^{14}C$ reaction: 0.006 b.

b. Thermal flux: 4.8E13 $n/cm^2$-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

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:

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

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

where:

$N_a$ = Rate of production, atoms/sec
$N_T$ = Number of target $^{17}O$ or $^{14}N$ target species per kg of coolant
$\sigma_i$ = "effective" neutron cross-section for each of the 2 or 3 energy groups, $cm^2$
$\varphi_i$ = neutron flux for each of the 2 or 3 energy groups, neutron/$cm^2$-sec

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

$$A_a = N_a \bullet \lambda_a$$

Where $\lambda_a$ is the decay constant of the species.

The source term in μCi/sec is given by:

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

The following methodology for estimating a site specific BWR $^{14}C$ 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]**

| BOC | | 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 $^{14}C$ 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 μCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction and μCi/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**

| Neutron Group | Group Energy | "Effective Cross-Section (σ)", b | |
|---|---|---|---|
| | | 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 $^{14}N(n,p)^{14}C$ Reaction in the BWR**

| Neutron Group | Group Energy | "Effective Cross-Section (σ)", b | |
|---|---|---|---|
| | | 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 $^{17}O(n,\alpha)^{14}C$ reaction is calculated using the three energy group flux distribution as follows:

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | intermediate neutron flux, n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux, n/cm$^2$-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-μCi |

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

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

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

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | intermediate neutron flux, n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux, n/cm$^2$-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-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 $H_2O$. 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 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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ Source Term from $^{17}O(n,\alpha)^{14}C$ Reaction

$$12{,}655 \text{ kg} \bullet 1.749\text{E-}5 \text{ } \mu\text{Ci/sec-kg} + 17{,}100 \text{ kg} \bullet 2.046\text{E-}5 \text{ } \mu\text{Ci/sec-kg}$$

$$= 0.571 \text{ } \mu\text{Ci/sec}$$
$$= 18.0 \text{ Ci/yr}$$
$$= 0.574 \text{ } \mu\text{Ci/MW}_{th}\text{-h}$$
$$= 5.03 \text{ Ci/GW}_{th}\text{-yr}$$
$$= 14.8 \text{ Ci/GW}_{e}\text{-yr @ 34\% efficiency}$$
$$= 21.3 \text{ kBq/MW}_{th}\text{-h}$$

#### 3.3.4.2 $^{14}C$ Source Term from $^{14}N(n,p)^{14}C$ Reaction

$$12{,}655 \text{ kg} \bullet 2.151\text{E-}7 \text{ } \mu\text{Ci/sec-kg-ppm N} + 17{,}100 \text{ kg} \bullet 3.226\text{E-}7 \text{ } \mu\text{Ci/sec-kg-ppm N}$$
$$= 8.239\text{E-}3 \text{ } \mu\text{Ci/sec-ppm N}$$

For 10 ppb N in the reactor coolant during steady state operation, this reaction will produce only 0.0026 Ci/yr or ~ 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 $^{14}C$ source term is the sum of the production rates from $^{17}O$ and $^{14}N$. However, the nitrogen concentration in the reactor coolant is so low that the $^{14}C$ generation from nitrogen can be neglected.

Calculation of the $^{14}C$ production rate from $^{17}O$ 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 $^{14}C$ is released from the core in a volatile form such as $^{14}CO$ or $^{14}CO_2$. The $^{14}CO$ will be drawn off at the SJAE and will be converted to $^{14}CO_2$ in the recombiner and/or in the environment. The $^{14}CO_2$ will partition at the steam/water interface in the reactor vessel and in the condenser.

If $CO_2$ is the principal species produced in the BWR coolant, the fraction of the $^{14}CO_2$ carried over in the steam can be calculated based on high temperature partitioning and hydrolysis data for $CO_2$ 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).

$$CO_{2(l)} \leftrightarrow CO_{2(g)} \qquad K = 7.9 \text{ M Pa kg/mole}$$

$$H_2CO_{3(aq)} \leftrightarrow H^+_{(aq)} + HCO_3^-{}_{(aq)} \qquad K_1 = 7.5E\text{-}9 \text{ mol}$$

$$HCO_3^-{}_{(aq)} \leftrightarrow H^+_{(aq)} + CO_3^{=}{}_{(aq)} \qquad K_2 = 3.03E\text{-}12 \text{ mole}$$

$$H_2O \leftrightarrow H^+_{(aq)} + OH^-_{(aq)} \qquad K_w = 5.62E\text{-}12 \text{ mol}^2$$

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:

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

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

Assuming an ideal gas and saturated steam at a temperature of 285.9 ºC (559.0 K), the steam $CO_2$ concentration will be;

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

and the mass partition coefficient will be:

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

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 · 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 µCi/h (18.0 Ci/y) for the example 3578 $MW_{th}$ BWR, the concentration of $^{14}C$ in the returning liquid from the steam/water interface should be ~5.9E-9 µCi/g. The observed concentrations of $^{14}C$ in the BWR coolant is considerably higher (8E-8 to 4E-6 µCi/g) which indicates that other less volatile species of $^{14}C$ 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 $^{11}C$ atoms in aqueous solutions. (Note: The chemical fate of $^{14}C$ is expected to be identical to that observed by Stenström (1970) for $^{11}C$.) 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 $^{11}C$ Species in Triple Distilled, Degassed Water Irradiated with 185 MeV Protons, in Per Cent of the Total Yield of $^{11}C$**

| Dose (rad) | $^{11}CO_2$ | $^{11}CO$ | $H^{11}COOH$ | $H^{11}CHO$ | $^{11}CH_3OH$ | $^{11}CH_4$ | 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), $CO_2$ (23±1%), CO (32±1%), $HCO_2H$ (17±2%), $H_2CO$ (21±2%), $CH_3OH$ (6±2%) and $CH_4$ (0.2±0.03%) are the observed reaction products. At higher doses (>20,000 rad), the dominant product is $CO_2$. 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 $^{14}CO_2$ should be the dominant reaction product released from the BWR core. However, it is worthwhile to note that the chemical form of $^{14}C$ 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, $CO_2$, CO, $CH_4$, $C_2H_6$, $C_3H_8$ and $C_4H_{10}$). 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 $^{14}C$ in the sample was separately determined by passing another aliquot with added carriers over CuO at 800ºC prior to counting. The results of these measurement were 95% $CO_2$, 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 $^{14}C$ release rate of 0.182 μCi/sec from the air ejector and a release rate of <3E-4 μCi/sec at the turbine gland seal condenser exhaust. The estimated amounts of $^{14}C$ 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 $^{14}C$ as $CO_2$ as compared to other chemical species. The average reactor coolant $^{14}C$ concentration after the steam separation was reported to be 4.0E-6 μCi/ml (Table 3-10). The $^{14}C$ 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 $^{14}C$ Released from the Oyster Creek Main Condenser Steam Jet Air Ejectors[a]**

| | Concentration, µCi/cc | | |
|---|---|---|---|
| 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 $^{14}C$ 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 $MW_e$ 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. $^{14}C$ releases from each reactor building during power operation were nearly the same. The release of $^{14}C$ in the reactor building exhaust air was not influenced much by operation of either plant and most of the $^{14}C$ released from both reactor buildings was in the oxidized form. $^{14}C$ 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 $^{14}C$ to the main stack releases during power operation.

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

| | Unit 1 Turbine Building | | Unit 2 Turbine Building | |
|---|---|---|---|---|
| **Date in 1982** | **µCi/sec** | **% Oxidized** | **µCi/sec** | **% 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 Reactor Building | | Unit 2 Reactor 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**
**$^{14}C$ Measurements at Brunswick Units 1 and 2 (NUREG/CR-4245, 1985) (continued)**

| | Radwaste | | Main Stack | |
|---|---|---|---|---|
| **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 $^{14}C$ during power operation and 60 to 70% during shutdowns. When both units were running, $^{14}C$ 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 µCi/$MW_{th}$-h values were reasonably consistent. This average value of 0.68 µCi/$MW_{th}$-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/$MW_{th}$-h**

| Measurement Date in 1982 | Measured Main Stack Release, µCi/sec | % Oxidized | Estimated % Power | | Estimated Total $MW_{th}$ | Estimated µCi/ $MW_{th}$-h |
|---|---|---|---|---|---|---|
| | | | Unit 1 | Unit 2 | | |
| 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 µCi/$cm^3$. 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% $^{14}CO_2$ 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 $^{14}C$ was detected for the deep-bed condensate demineralizers. This is not to say that there is no $^{14}C$, as carbonate, retained on the bed resins, rather, it can be interpreted to mean that the beds were saturated with respect to $CO_2$.

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 µCi/ml whereas the outlet was 2.9E-7 µCi/ml, a removal efficiency of ~86%. A waste water composite contained 1.2E-7 µCi/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% |
| $^{14}CH_4$, $^{14}C_2H_6$, 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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ in the Nordic BWR were normally equal or slightly lower than the calculated production rate. The two Olkiluoto plants showed an increased release of $^{14}C$ 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 $^{14}CO_2$ with out-of-reactor surfaces may be responsible for the small percentage of methane (~5%) seen in the gaseous effluent.

**Table 3-15**
**$^{14}C$ 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 $^{14}C$ 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**
**$^{14}C$ 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 $^{14}CO_2$ Adsorption on Charcoal*

Magnusson and Stenström (2005) measured the concentration of $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ through the charcoal beds is compared to the calculate source term in Table 3-18.

**Table 3-18**
**Transport of $^{14}C$ 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** | **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 $^{14}C$ 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 25ºC 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 ~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 ~2.6E3 µCi/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 $^{14}CO_2$ is not believed to be tightly held on the activated carbon. The delay time can be estimated using the relationship:

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

where:

$t_D$ = Delay time, minutes
M = Mass of charcoal, g
F = Flowrate, cc/min
$K_d$ = Dynamic adsorption coefficient, cc/gram

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

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 $^{14}C$ 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 $^{16}N$ 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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ 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 $^{60}Co$ 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 μCi 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 μCi/g. A steady state release of 10% of the production rate in a single defective fuel rod would be ~2E-6 μCi/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 $^{14}C$ 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 (1995-2010 values):** | | | **5.1±0.6** | | |

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

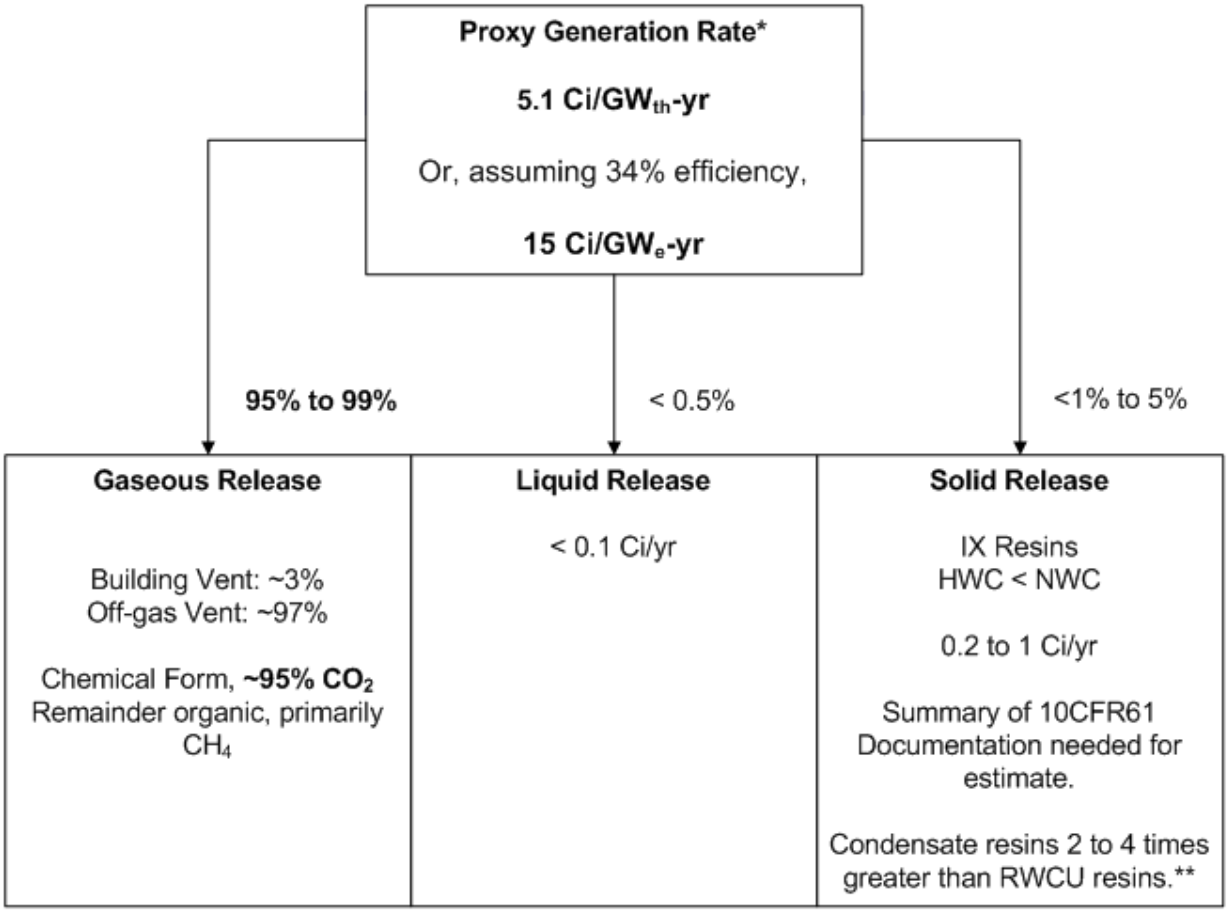
b. Lundgren's value: 23-24 $kBq/MW_{th}$-h.

**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, $^{14}C$ 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 ±15%.

**The RWCU ion exchange resins are expected to contain $^{14}C$ 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.

**Figure 3-1**
**$^{14}C$ Transport in the BWR**

# 4
# CARBON-14 GENERATION AND RELEASE IN PWR SYSTEMS

## 4.1 Overview of $^{14}C$ 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 $^{14}C$ species initially produced are organic and contain only a single carbon atom. Possible species include methane ($^{14}CH_4$), methanol ($^{14}CH_3OH$), formaldehyde ($H_2{}^{14}C{=}O$ or the *gem*-diol $H_2{}^{14}C(OH)_2$ (Dong and Dasgupta, 1988)), and formic acid ($H^{14}COOH$). 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 $^{14}CH_4$ which partitions itself between the reactor coolant liquid and gas phases in the VCT and pressurizer. A calculation performed by the authors indicates that ~43% of the methane is dissolved in the reactor coolant, ~50% is present in the VCT vapor space and ~7% in the pressurizer vapor space.

Analyses of pressurized PWR reactor coolant samples shows that the $^{14}C$ species are essentially 100% organic, and ~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 ~20 μCi/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 μCi/g (coolant inventory of ~185 mCi). 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 μCi/g of which ~50% was in the gas phase. In general, lower concentrations (1-5 E-4 μCi/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 μCi/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 μCi/min leaving another ~18 μCi/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.

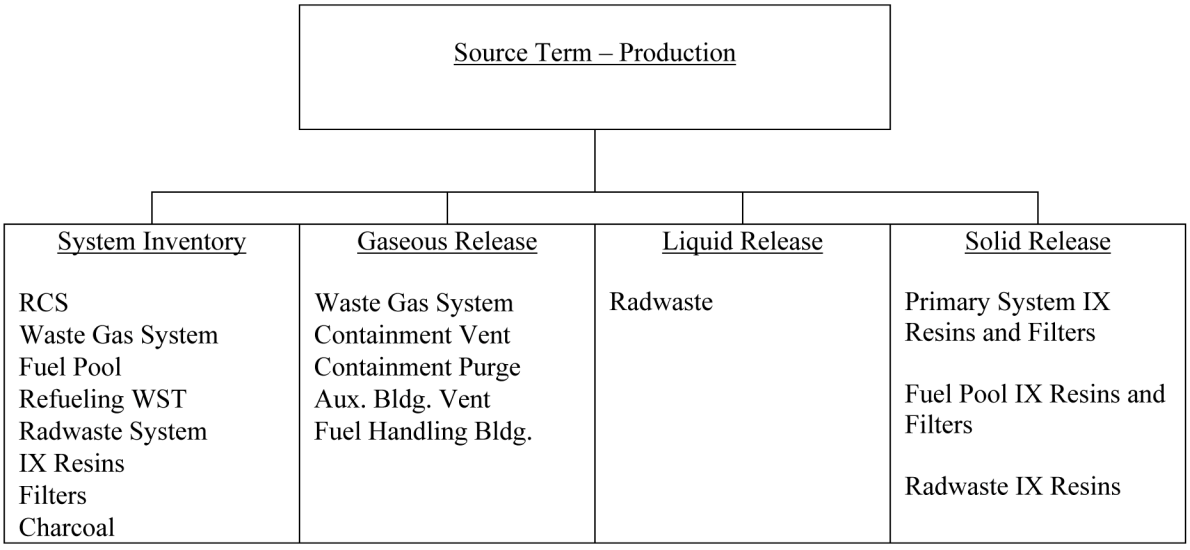



**Figure 4-1**
**Carbon-14 Mass Balance for PWR**

The $^{14}C$ 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 $^{14}C$ in some chemical form. Measurements of $^{14}C$ concentrations in the various liquid systems have been performed, and some of the reported data are included in this report. As a general rule, $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ source term due to the low neutron flux in containment.

## 4.2 $^{14}$C Source Term Estimation and Measurement

In the PWR-GALE Code, (NUREG-0017 (1985)),"The annual quantity of $^{14}$C released from a pressurized water reactor is 7.3 Ci/yr. It is assumed that most of the $^{14}$C 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 $^{14}$C 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)**

| Plant* | Year | | | | Ci/Yr-Unit |
|---|---|---|---|---|---|
| | 1975 | 1976 | 1977 | 1978 | |
| | | | | | |
| Conn. Yankee | 44 | 40 | 30 | 70 | 46 |
| | | | | | |
| Yankee Rowe | 1.6 | 0.13 | 0.24 | 0.33 | 0.58 |
| **Plant**** | **Area** | | | | **Ci/Yr-Unit** |
| Turkey Point 3/4 | Auxiliary Building | | | | 2.4 |
| (NUREG/CR-1629) | Containment | | | | 0.075 |
| | Waste Gas Processing System | | | | 0.82 |
| | Spent Fuel Area | | | | 0.38 |
| | | | | Total: | 3.7 |
| Fort Calhoun | Fuel Pool and Auxiliary Building | | | | 0.30 |
| (NUREG/CR-0140) | Waste Gas Processing System | | | | 0.81 |
| | Containment Building*** | | | | 0.78 |
| | | | | Total: | 1.9 |
| Zion Units 1 & 2 | Containment Building | | | | 1.8 |
| (NUREG/CR-0715) | Fuel Handling and Auxiliary Building | | | | 1.4 |
| | Waste Gas Processing System | | | | 0.062 |
| | | | | Total: | 3.3 |
| Prairie Island 1/2 | Containment Building | | | | 0.016 |
| (NUREG/CR-4397) | Fuel Handling and Auxiliary Building | | | | 3.3 |
| Waste Gas Processing System | | | | | 0.25 |
| | | | | Total: | 3.6 |
| Rancho Seco | Containment Building | | | | 0.9 |
| (NUREG/CR-2348) | Fuel Handling and Auxiliary Building | | | | 1.85 |
| | Waste Gas Processing System | | | | 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 "$^{14}C$ reacts to form volatile compounds (principally $CH_4$. $C_2H_6$ and $CO_2$) 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 "$^{14}C$ 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 $^{14}C$ 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] |
| PWR Fuel | O-17 | 4 | 7.1 | 4.0 | | 2.7 |
| | N-14 | 18 | 12.2 | 7.6 | | 10.9 |
| | Total | 22 | 19.3 | 11.6 | 17[b] | 13.6 |
| PWR Coolant | O-17 | 3.2 | 9.8 | 3.3 | | |
| | N-14 | 0.09 | 1.3 | 0.1 | | |
| | 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 $^{17}O(n,\alpha)^{14}C$ 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[a] | Production Rate[b] |
|---|---|
| $^{17}O(n,\alpha)^{14}C$ | 6.0 Ci/GW(e)-yr |
| $^{14}N(n,p)^{14}C$ | 0.12 Ci/GW(e)-yr/ppm |
| $^{13}C(n,\gamma)^{14}C$ | 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/$cm^2$-sec, PWR coolant mass exposed to flux: 13,400 kg.

Magnusson (2008) performed an extensive program to characterize $^{14}C$ in Swedish light water reactors. The work involved the development of $^{14}C$ analysis techniques, mapping of $^{14}C$ in waste streams and measurements of $^{14}C$ 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 $^{14}C$ 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 ±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:

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

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

where:

| | | |
|---|---|---|
| $N_a$ | = | Rate of production, atoms/sec |
| $N_T$ | = | Number of target $^{17}O$ or $^{14}N$ target species per kg of coolant |
| $\sigma_i$ | = | "effective" neutron cross-section for each of the 2 or 3 energy groups, $cm^2$ |
| $\varphi_i$ | = | neutron flux for each of the 2 or 3 energy groups, neutron/$cm^2$-sec |

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

$$A_a = N_a \bullet \lambda_a$$

Where $\lambda_a$ is the decay constant of the species.

The source term in μCi/sec is given by:

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

The following methodology for estimating a site specific PWR $^{14}C$ 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/cm$^2$-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/cm$^2$-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; ≤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 μCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction (Table 4-7) and μCi/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 $^{14}$C from $^{17}O(n,\alpha)^{14}C$ 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 $^{17}O(n,\alpha)^{14}C$ reaction is calculated for the three group flux distribution as follows:

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | Intermediate neutron flux, n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux, n/cm$^2$-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-μCi |

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

**Table 4-8**
**PWR $^{14}C$ Production Rate from the $^{17}O(n,\alpha)^{14}C$ 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 $^{14}C$ from $^{14}N(n,p)^{14}C$ Reaction:

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

**Table 4-9**
**"Effective Cross-Sections" for the $^{14}N(n,p)^{14}C$ 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:

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | Intermediate neutron flux, n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux, n/cm$^2$-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-μCi |

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

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

| | Production Rate, μCi/sec-kg-ppm N |
|---|---|
| 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 $^{14}C$ 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 $MW_e$ (~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 $^{14}C$ Source Term from $^{17}O(n,\alpha)^{14}C$ Reaction

The production rate for this reaction would be:

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

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 $^{14}C$ Source Term from $^{14}N(n,p)^{14}C$ Reaction

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

$$1.0\ ppm\ N \bullet 2.100E\text{-}7\ \mu Ci/sec\text{-}kg\text{-}ppm\ N \bullet 14{,}100\ kg = 2.961E\text{-}3\ \mu Ci/sec$$

Guidance for calculating nitrogen concentrations in reactor coolant is provided in Appendix E. If it is assumed that this reactor operated with ~12 % $N_2$ in the VCT and 0.53 ppm $NH_3$, 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 $^{14}C$ 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 $^{14}C$ release was in a non-$CO_2$ form.

Kunz, et al., 1974 measured the chemical form of $^{14}C$ 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 $^{14}C$ 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 |
| $C_2H_6$ | 22.4 | 7.5 | 3.5 | 37.5 | 36.0 |
| $C_3H_8$ | 2.4 | 7.2 | 2.7 | 3.9 | 2.3 |
| $C_4H_{10}$ | 0.9 | 10.0 | 2.7 | n.d. | 0.5 |
| $CO_2$ | 4.6 | 0.3 | 3.4 | 1.8 | 2.6 |
| CO | 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 ($CH_4$ and $C_2H_6$). The $CO_2$ 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 $CO_2$. Since less than 5% of the releases were as $CO_2$ 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 $MW_e$ 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 $^{14}C$ release and chemical form of $^{14}C$ 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**
**$^{14}C$ 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% |
| $^{14}CH_4$, $^{14}C_2H_6$, 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 $^{14}C$ 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 $^{14}CO_2$ at the plant vent location.

The average $^{14}C$ concentration in the nine decay tank samples collected between 1973 and 1981 was 1E-3 μCi/cm$^3$. 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% $^{14}CH_4$, 16% $^{14}C_2H_6$, 6% $^{14}C_3H_8$ and $^{14}C_4H_{10}$ and 4% $^{14}CO_2$.

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 μCi/ml with an average of 1.1E-4 μCi/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 μCi/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 $^{14}CO_2$ 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 $^{14}CO_2$ was estimated at 2.5 Ci/GW(e)-yr, or 26% of the total gaseous $^{14}C$ release. The $^{14}CO_2$ 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 $^{14}C$ Activity Detected in Release Pathways at Indian Point Unit-3**

| | % of $^{14}C$ Chemical Compounds | | |
|---|---|---|---|
| **$^{14}C$ Compound** | **Decay Tank[a]** | **Containment Air[b]** | **Plant Vent[c]** |
| $CH_4$ | 62 | 60 | 46 |
| $C_2H_6$, $C_3H_8$ and $C_4H_{10}$ | 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 μCi/ml with an average of 6.6E-5 μCi/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 $^{14}C$ in the waste streams. The largest amounts of $^{14}C$ were found in the resins and filters used for RWCU. From these data the annual amounts of $^{14}C$ 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 $^{14}C$ concentrations (Table 4-15). The data indicate that a significant fraction of the $^{14}C$ species in the reactor coolant is organic. However, it is not clear whether pressurized samples were taken to retain the gaseous $^{14}C$ species.

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

| Plant | Sample Date | Total 14C μCi/cc | Inorganic 14C μCi/cc | % Organic 14C |
|---|---|---|---|---|
| B | 1/29/92 | 1.13E-4 | 1.65E-5 | 85.4 |
| B | 2/05/92 | 1.41E-4 | 7.51E-6 | 94.7 |
| C | 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 $^{14}C$ 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 $^{14}C$ Analysis, 2005**

| | Gas Phase | | Liquid Phase | | | |
|---|---|---|---|---|---|---|
| | **Inorganic** | **Organic** | **Inorganic** | **Organic** | **Total** | **%** |
| **System** | **Activity Concentration, 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 Tank | | | 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 Calculated Production** | |
| **Gaseous Waste** | | |
| Stack Release | 86[c] | 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.
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)

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

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

The Ringhals stack release data (Figures 4-2 and 4-3) show that the $^{14}C$ release rate normally increases during a fuel cycle and that a significant fraction of the total $^{14}C$ 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.

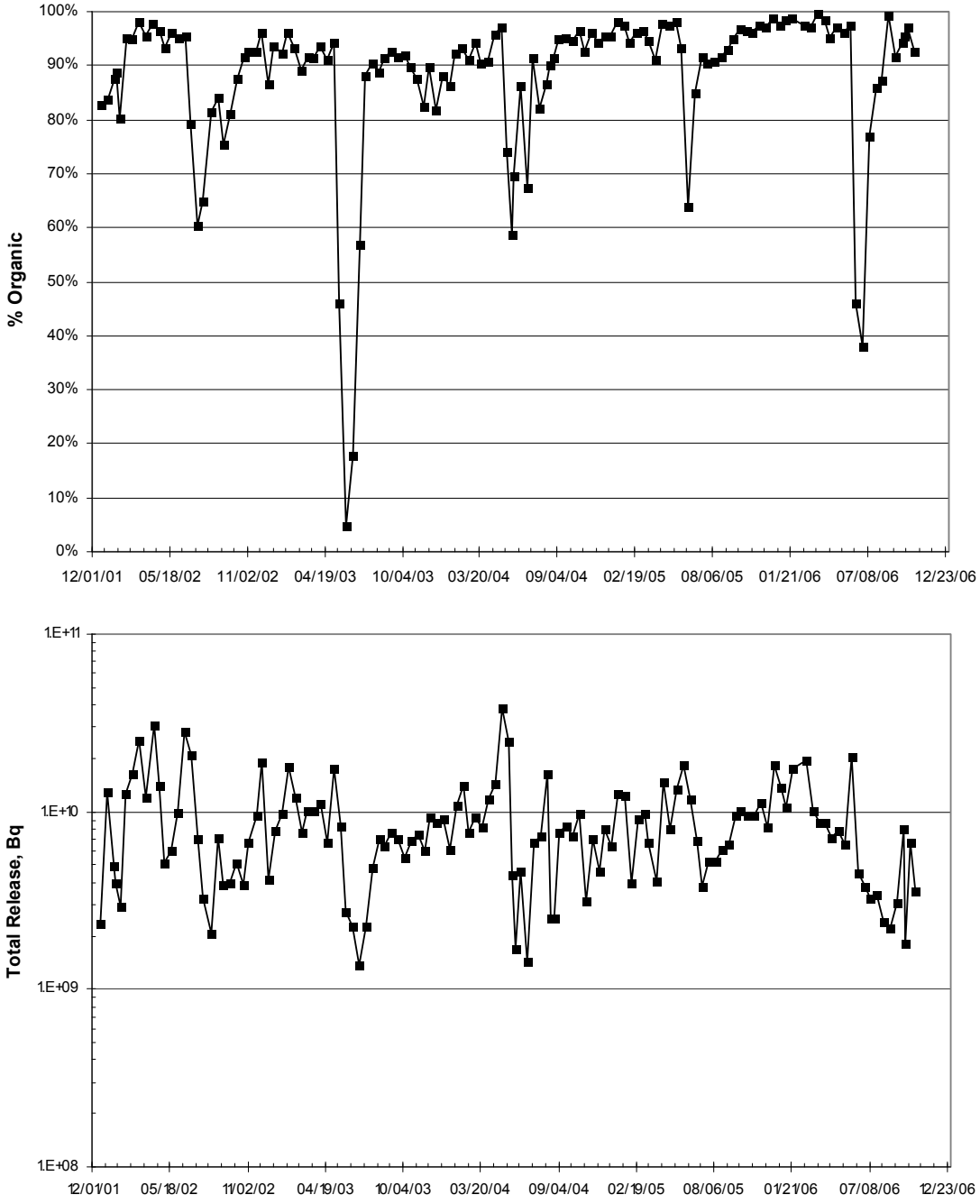



**Figure 4-2**
**Ringhals Unit-3 Main Stack $^{14}C$ Release During the Years 2002-2006 (Bengtsson, 2010)**

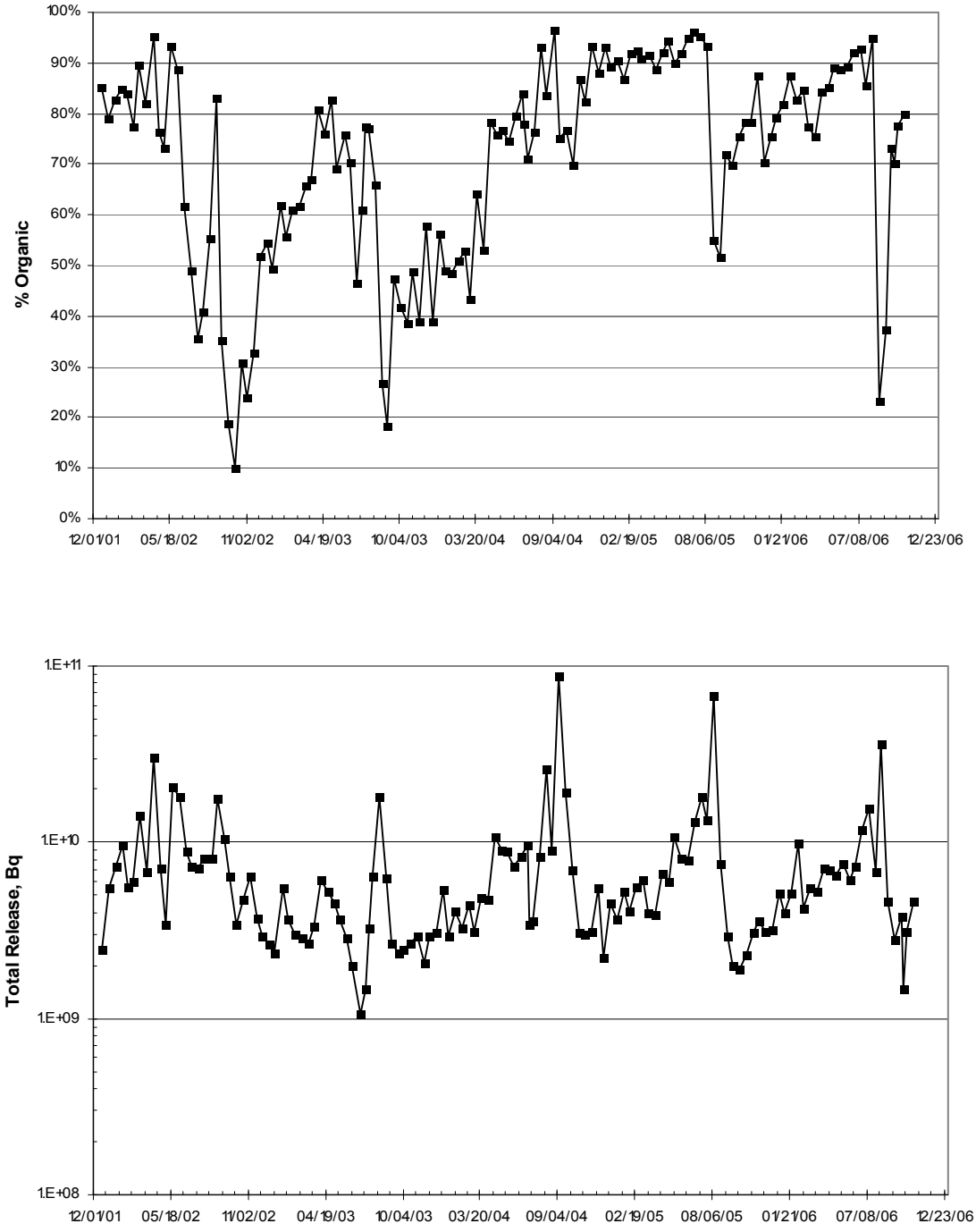



**Figure 4-3**
**Ringhals Unit-4 Main Stack $^{14}C$ Release During the Years 2002-2006 (Bengtsson, 2008)**

### *4.4.6 Diablo Canyon Units 1 and 2*

The concentrations of $^{14}C$ 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 $^{14}C$ Gaseous Sampling, April 2010**

| | $^{14}CO_2$ | $^{14}CH_4$ | Particulate | Ratio $CH_4/CO_2$ |
|---|---|---|---|---|
| | Activity Concentration, pCi/L | | | |
| **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, $^{14}CH_4$ 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 $^{14}C$ Annual Discharge Per Operating Unit During Operation**

| Release Point | $^{14}C$, Ci |
|---|---|
| Plant Vent | ~5.4 |
| Containment | ~0.2 |
| Gas Decay Tanks | ~1.2 |
| Total: | ~6.8 |
| | ~1.9 as $CO_2$ |

Sampling for $^{14}C$ 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 $^{14}C$ Liquid Sampling April 2010[a]**

| System | Unit-1 | Unit-2 |
|---|---|---|
| | µCi/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 $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ Sampling Results**

| Sampled Location | Total $^{14}C$ | $^{14}C$ as $CO_2$ | % as $^{14}CO_2$ |
|---|---|---|---|
| | 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 $^{14}C$ at five Korean PWRs. Gaseous sampling was done with NaOH bubblers, using a catalytic converter for the conversion of non-$CO_2$ into $CO_2$. Typical sampling periods were 4 weeks. LSC counting of a $BaCO_3$ 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 $MW_e$ | 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 Average % Organic[a]: | | **38.1** |
| Kori Unit 3 | 9/15/06 – 12/28/06 | Fuel Bldg. | 1.06 | 0.2 |
| W – 950 $MW_e$ | | 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 % Organic[a]: | | **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 Average % Organic[a] | | **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 $AgNO_3$. 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 $^{14}C$ 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 $HCO_2^-$.

Since hydrazine is added to the primary system to reduce oxygen concentrations below 100 ppb at temperatures above ~100°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 ~5 to ~30ppb. This concentration of zinc does not impact on the $pH_T$ of the primary coolant. Formation of $^{14}C$ 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 μCi 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 μCi/g. A steady state release of 10% of the production rate in a single defective fuel rod would be ~1E-6 μCi/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 $^{14}C$ 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 $\mu Ci/MW_{th}$-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 $\mu Ci/MW_{th}$-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, $^{14}C$ 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 $^{14}C$ Generation Rates in Coolant at PWRs[a]**

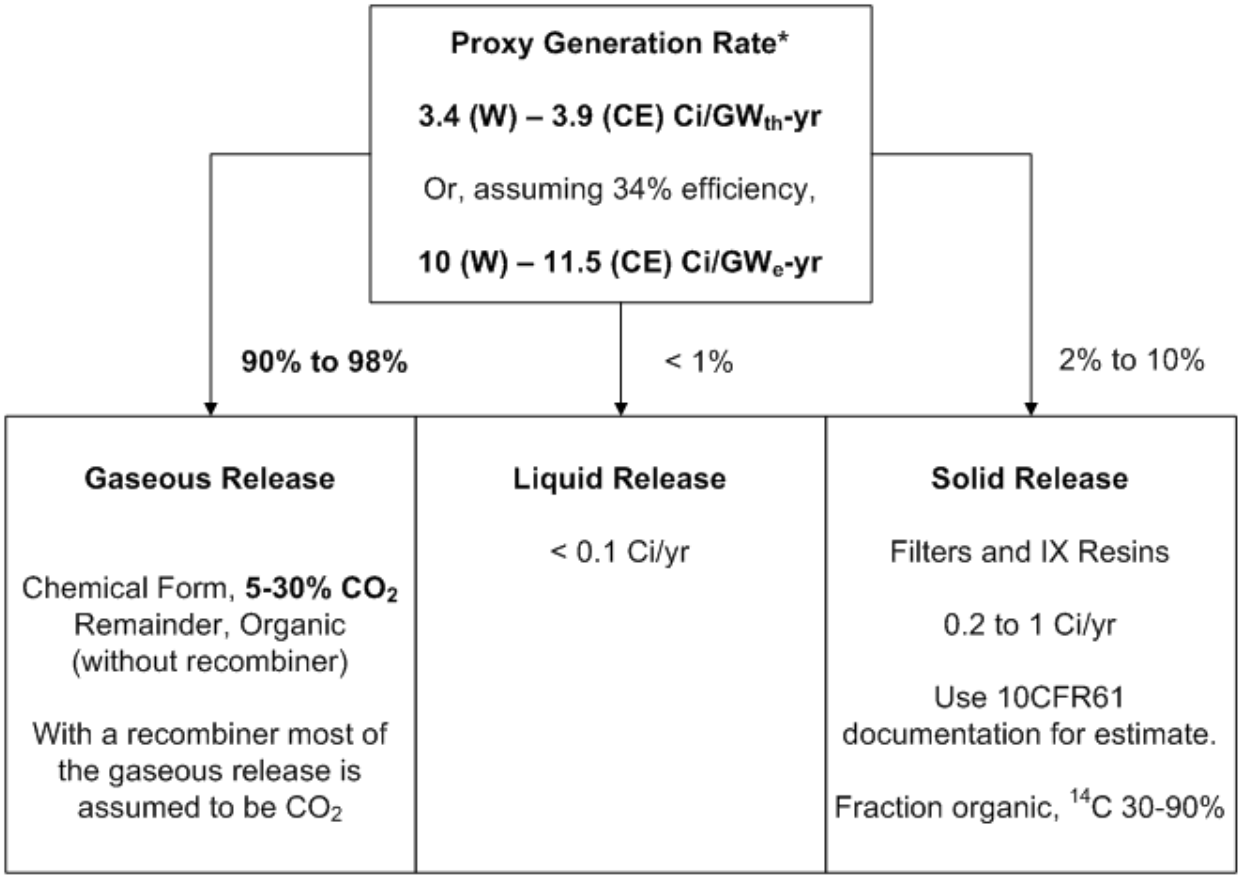
| Reference | Unit | μCi/$MW_t$-h | Ci/yr | Ci/$GW_e$-yr | GBq/$GW_e$-yr |
|---|---|---|---|---|---|
| Fowler et al (1976) | | | | **3.3** | |
| Bonka (1974) | - | | | **11.1** | |
| Kelly (1975) | 1000 $MW_e$ | | | **6** | |
| ERDA-1535 (1975) | - | | | **6** | |
| Hayes (1977) | 1000 $MW_e$ | | | **3.4** | |
| 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 | |
|---|---|---|---|
| **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 ±15%.

**Figure 4-4**
**$^{14}C$ Transport in the PWR**

# 5
# MEASUREMENT IN THE NUCLEAR POWER INDUSTRY

## 5.1 General

Historically, most of the early reactor site $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ to $^{12}C$ ratio by at least a factor of ten.

Reactor site $^{14}C$ 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 $^{14}C$ activity in the form of carbon dioxide or as a carbonate or carbamate salt. The usual gaseous effluent and atmospheric $^{14}C$ 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 $^{14}C$ in the form of carbon dioxide ($CO_2$) 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 $^{14}C$ 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 radionuclides 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 $^{14}C$ at the natural background level of approximately 1.2 pCi/m$^3$ of air, or 0.044 Bq/m$^3$ of air (Sejkora, 2010). Air is 0.034% by volume $CO_2$. This corresponds to 0.67 g of $CO_2$ per cubic meter of air. Olivier, et. al (2005) compared the liquid scintillation counting sensitivity of direct counting of a $Na_2CO_3$ solution mixed with the cocktail and precipitating the $CO_2$ with $CaCl_2$ and suspending the precipitate in the cocktail. The detection limit for the $NaCO_3$ solution was 0.3 Bq/m$^3$ and for suspended $CaCO_3$ 0.04 Bq/m$^3$. $CaCO_3$. Large samples of 100 cubic meters or more of air can result in quantities of inert $CO_2$ that exceed the capacity of the scintillation cocktail, and an analytical procedure has been developed to convert the $CO_2$ 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 $^{14}C$ 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 $^{14}C$. 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, $H_2$, $CO_2$, CO, $CH_4$, $C_2H_6$, $C_3H_8$, and $C_4H_{10}$ 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 $^{14}C$ activity of each chemical form. Pulse height analysis permitted simultaneous analysis of the tritium and $^{14}C$ fractions. The results for both the gas decay tank and containment atmosphere samples showed that in all cases over 80% of the $^{14}C$ activity was in the methane and ethane fractions with the majority as methane. The $CO_2$ fraction ranged from 4.6% to 1.8% except for the Indian Point-1 decay tank with 0.3% $CO_2$. In all samples, the CO fractions ranged from 0.4% to non-detectable.

In 1976, Kunz, et al. (Kunz, 1976) reported on $^{14}C$ measurements at the Nine Mile Point 1 BWR. Offgas samples ranging in volume from 100 to 500 cm$^3$ were taken from the main condenser SJAE discharge. These samples were mixed with measured amounts of carrier gases, Kr, Xe, CO, $CO_2$, $CH_4$, $C_2H_6$, $C_3H_8$, and $C_4H_{10}$. 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 $^{14}C$ activity in each fraction. The total $^{14}C$ 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 $CO_2$ prior to counting. The activity distributions showed that 95% of the off gas activity was in the form of $CO_2$, 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 $^{14}C$ 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 $CO_2$ absorber, (Mallcosorb™). Following sampling, the absorbed $CO_2$ was released in a closed gas circulating system by adding HCl to a water solution of the absorbent. The $CO_2$ was then reabsorbed in a NaOH solution, which was heated, and $BaCl_2$ was then added to precipitate $BaCO_3$. The precipitate was filtered, dried, weighed and checked for residual gamma activity. If gamma activity was detected, the $BaCO_3$ was dissolved in acid, and the resulting $CO_2$ was absorbed in a NaOH solution and re-precipitated as $BaCO_3$. The volume of air sampled was calculated based on the weight of the $BaCO_3$ recovered assuming a nominal 315 ppm $CO_2$ in air. The volume obtained agreed within 5% of the value obtained from the sample flow meter. The $BaCO_3$ was dissolved slowly with HCl, and the released $CO_2$ was scrubbed from the air stream using a spinning band column and a countercurrent-flow of liquid scintillation solution. The $^{14}C$ 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 $^{14}C$ 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 $^{14}C$ and $^{14}CO_2$ in the effluent gases. The continuous samplers used a 100 $cm^3$/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 $CO_2$. The gas then flowed through a solid drying agent such as Drierite™ (anhydrous $CaSO_4$) 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 $CO_2$. After sampling, the cartridge was removed, the contents acidified and helium carrier gas used to transport the released $CO_2$ to a liquid nitrogen cold trap which liquefied the $CO_2$. The volume of liquid $CO_2$ was measured, usually 660 $cm^3$ for a 2-week sampling period, and a 50 $cm^3$ aliquot of the liquid $CO_2$ 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 $^{14}C$ In addition to reviewing Kunz's work, they also published the primary coolant $^{14}C$ concentration they measured at three BWRs and three PWRs. The analysis procedure used 300 ml samples which were acidified with $H_2SO_4$ and $CO_2$ free air used to sweep the dissolved $CO_2$ into a $Ca(OH)_2$ trap precipitating $CaCO_2$. The $CaCO_2$ was further purified and the $^{14}C$ activity measured by liquid scintillation counting. The total inorganic and organic $^{14}C$ activity was determined by refluxing the coolant sample at 90°C with $K_2S_20_8$ and $AgNO_3$ catalyst to oxidize all carbon forms to $CO_2$ 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 to16 $m^3$ 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 $H_2SO_4$ to remove tritium in the form of HTO. The second bubbler was filled with 2N NaOH to trap $^{14}C$ as $^{14}CO_2$. The effluent from this bubbler went through a 600°C tube furnace filled with a platinum and palladium catalyst to oxidize organic carbon to $CO_2$. The effluent was then passed through a bubbler filled with 2N NaOH to absorb the $CO_2$. Testing with an air standard containing 400 ppm $CO_2$ and 100 ppm $CH_4$ showed that the furnace conversion efficiency exceeded 99.2%. (Note that any tritium present as HT will be oxidized to HTO in the tube furnace. Inclusion of an $H_2SO_4$ 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 2M NaOH to measure environmental, atmospheric $^{14}CO_2$ 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 $CO_2$ was more effective than $BaCO_3$ 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 $NH_4Cl$ and $BaCl_2$ was added to the NaOH bubbler solution to precipitate $BaCO_3$. Samples resulting in significant quantities of $BaCO_3$ 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 $CO_2$ absorbed in 10 ml of "Carbsorb E" plus 10 ml of "Pemafluor V".

The authors also describe the benzene synthesis method that converted the $CO_2$ from the $BaCO_3$ to benzene that then was added directly to the scintillation cocktail. This method was used infrequently.

## 5.3 Current Methods

Essentially all $^{14}C$ measurements involve trapping of $CO_2$. In the case of BWRs, at least 95% of the gaseous $^{14}C$ is released as $CO_2$. At most PWRs, a large fraction of the $^{14}C$ release is in an organic form, and to determine the total $^{14}C$ activity these compounds can be oxidized to $CO_2$ 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 $CO_2$. Vegetation and animal tissue as well as resins can be oxidized in combustion units to convert any carbon compounds to $CO_2$.

The released $CO_2$ can be trapped in a bubbler containing NaOH or KOH and later precipitated as $BaCO_3$. If it is an atmospheric sample, it can be filtered, washed, dried and weighed to determine the chemical yield based on the known atmospheric $CO_2$ concentration, and the sample volume. If further radiochemical purification is needed, the precipitate can be dissolved in a closed system using HCl or $H_2SO_4$, and nitrogen carrier gas used to transfer the resulting $CO_2$ to another caustic containing bubbler. At this point, there are several choices. There are organic $CO_2$ absorbers such as ethanolamine that form carbamates that are miscible with LSC scintillation phosphors. Alternately, the $BaCO_3$ can be suspended in the scintillation cocktail. The choice of methodology depends primarily on the amount of inert $CO_2$ 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 $^{14}C$ production rate of 15 Ci/yr and 25 scfm air inleakage, the offgas system discharge concentration is 80 d/m per $cm^3$. 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 $^{14}C$ 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 $CO_2$, the atmospheric concentration of $CO_2$ is 0.0168 millimoles per liter, and a 100 liter sample would contain 1.68 millimoles of $CO_2$ 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 $CO_2$; whereas, the Hague 7000 uses four 250 ml vials intended to be filled with sodium hydroxide. The cascaded bubblers provide assurance that all the $^{14}C$ 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 $CO_2$ and is more suitable for environmental monitoring.

# 6
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# A
# BWR $^{14}C$ SOURCE TERM CALCULATION

## 1.0 Cross-Section Estimation

The "effective cross-sections" for the two dominant nuclear reactions producing $^{14}C$ 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.

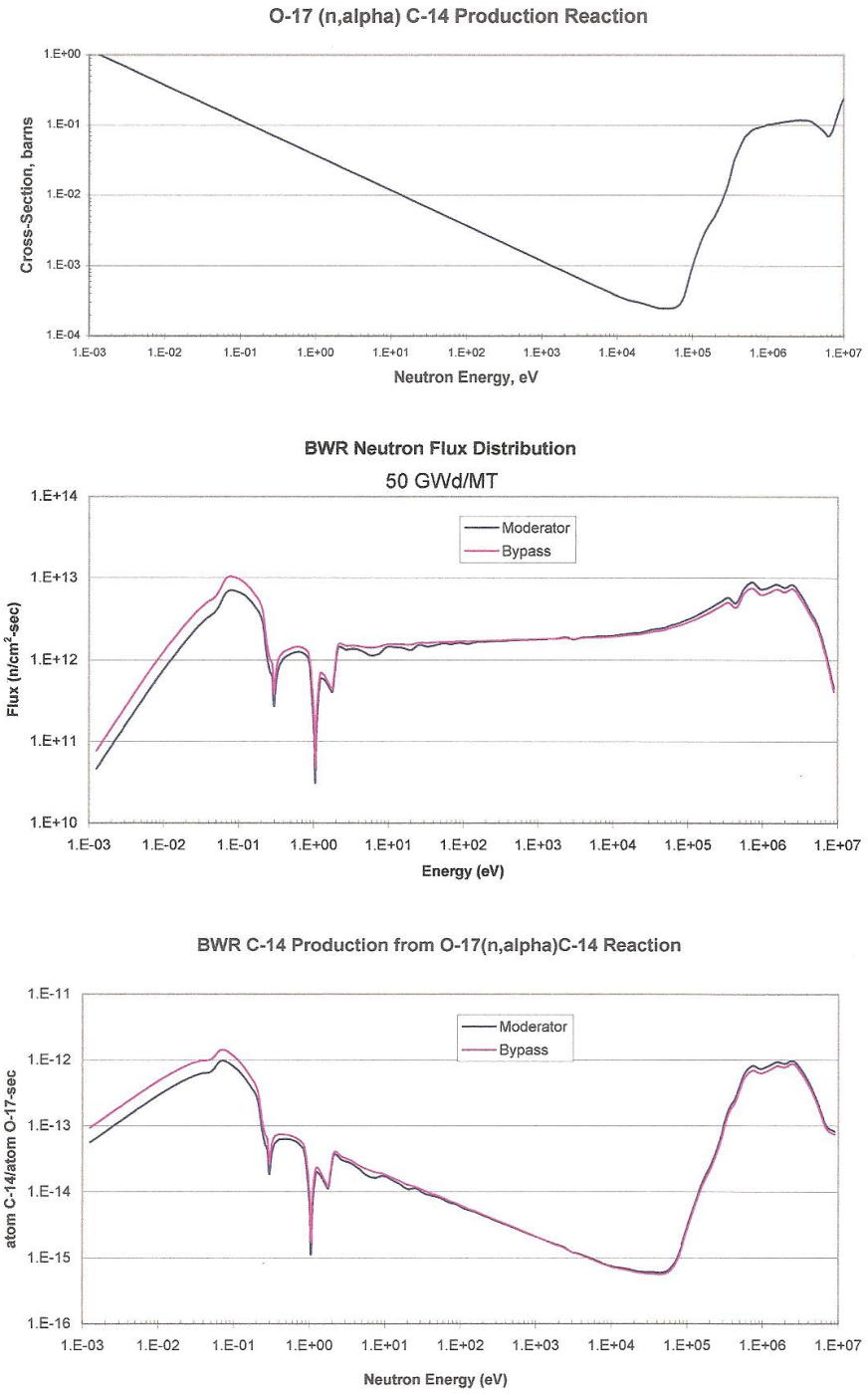


**Figure A-1**
**$^{17}O(n,\alpha)^{14}C$ Reaction Cross-Section, Reference BWR Neutron Flux Distribution at 50 GWd/MT and $^{14}C$ Production**

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

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

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

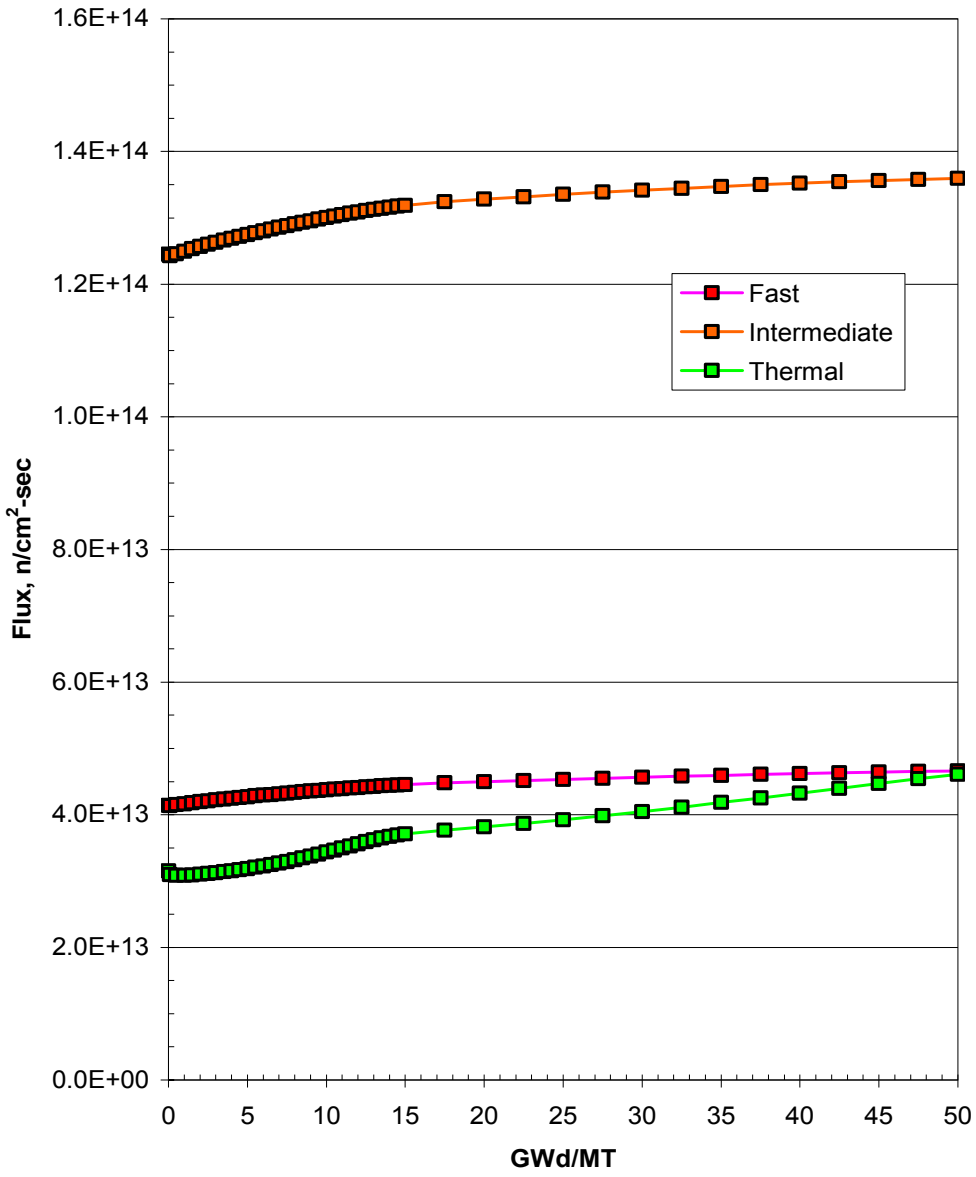


**Figure A-2**
**BWR Moderator Neutron Flux vs. Fuel Exposure**

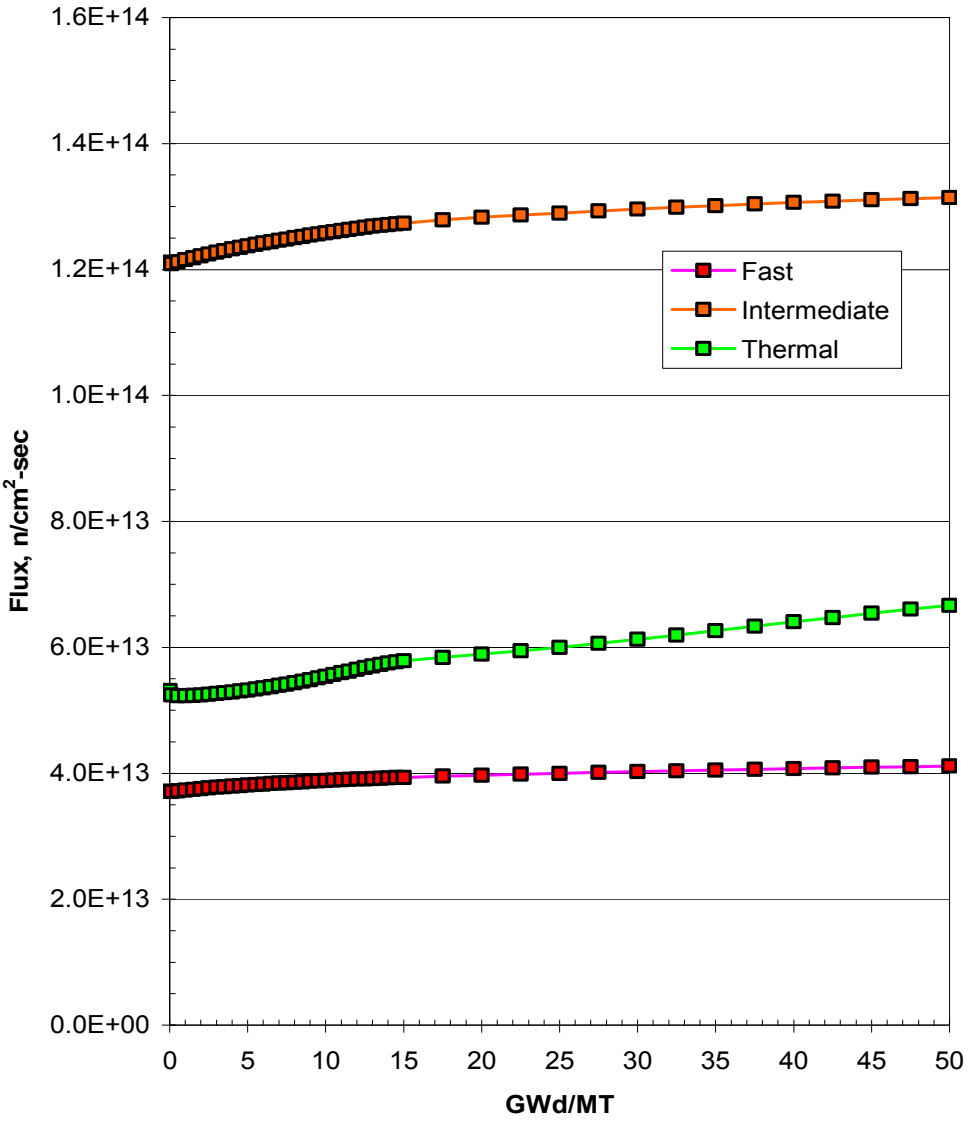


**Figure A-3**
**BWR Bypass Neutron Flux vs. Fuel Exposure**

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

| | Neutron Flux, n/cm$^2$-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 Rate, µCi/sec-kg | | | | | |
| | Moderator Region | | | 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 $^{14}C$ production per kilogram of water at an exposure of 30 GWd/MT is illustrated below. The number of $^{17}O$ atoms per kilogram of water is 1.27E22.

### 2.1.1 Moderator Region

$$\frac{1.27E22 \bullet (4.05E13 \bullet 0.1334 + 1.34E14 \bullet 0.0238 + 4.57E13 \bullet 0.1106) \bullet 1E-24 \bullet \ln 2}{3.7E4 \bullet 5730 \bullet 365.25 \bullet 24 \bullet 3600} = 1.80E-5\ \mu Ci/\sec - kg$$

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

### 2.1.2 Bypass Region

$$\frac{1.27E22 \bullet (6.13E13 \bullet 0.1389 + 1.30E14 \bullet 0.0222 + 4.03E13 \bullet 0.1106) \bullet 1E-24 \bullet \ln 2}{3.7E4 \bullet 5730 \bullet 365.25 \bullet 24 \bullet 3600} = 2.08E-5\ \mu Ci/\sec - kg$$

Production in the bypass region is ~54% from thermal neutrons, ~18% from intermediate neutrons and ~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:

1.80E-5*12,655 + 2.08E-5*17,100 = 0.573 μCi/sec
= 18.1 Ci/yr
= 14.9 Ci/GW(e)-yr (@34% efficiency)
= 21.3 kBq/$MW_{th}$-h
= 0.576 μCi/$MW_{th}$-h
= 5.05 Ci/$GW_{th}$-yr

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

The $^{14}N(n,p)^{14}C$ reaction cross-section, the reference BWR neutron flux distribution (GE9 Fuel, 8x8 with large center water rod) at 50 GWd/MT and the $^{14}C$ 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 $^{14}N(n,p)^{14}C$ 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/cm$^2$-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 $^{14}N(n,p)^{14}C$ 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 $^{14}C$ production rate per kilogram of water with 1.0 ppm dissolved nitrogen for each of the core regions is illustrated below. The number of $^{14}N$ 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.

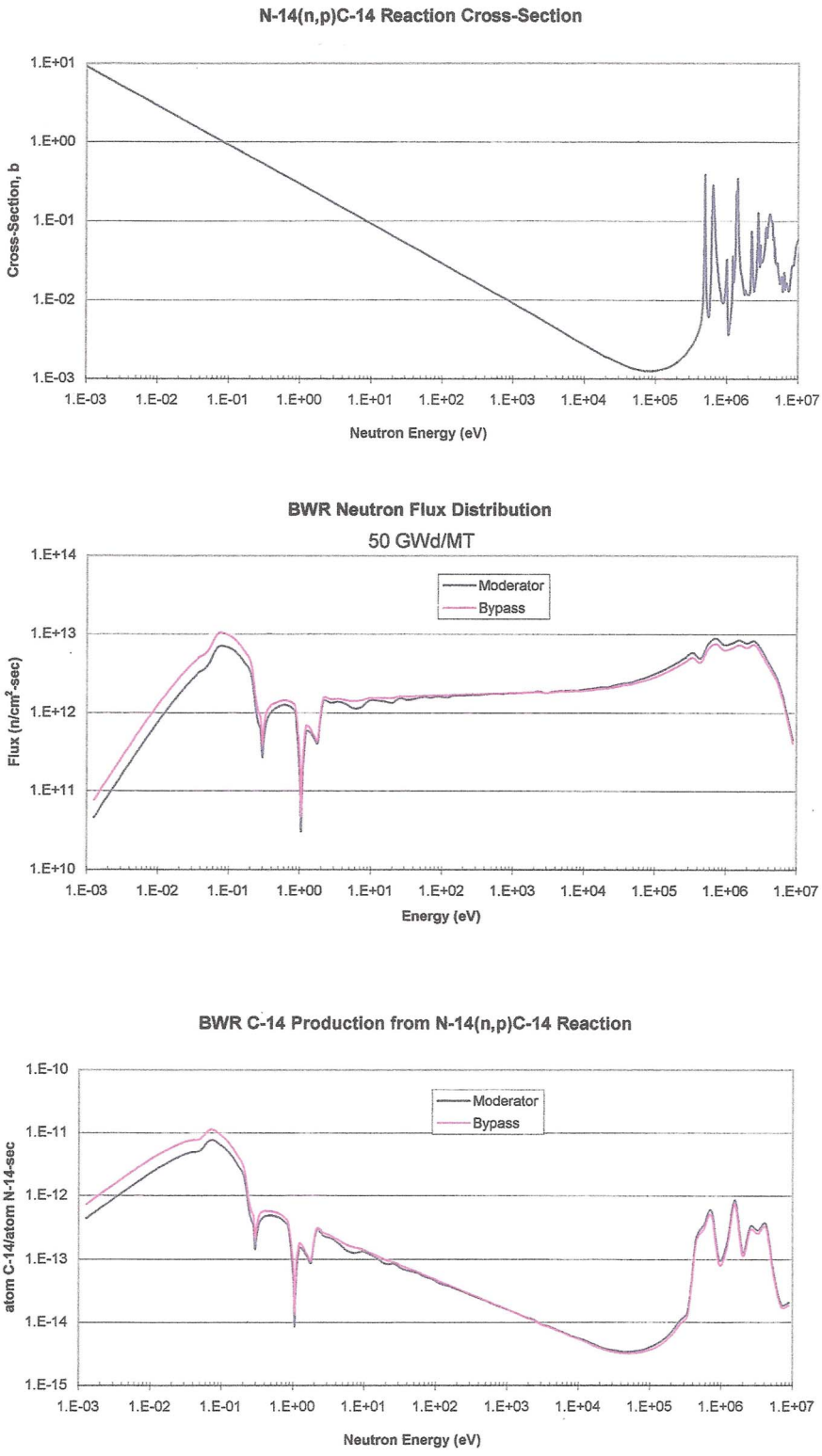


**Figure A-4**
**$^{14}N(n,p)^{14}C$ Reaction Cross-Section, Reference BWR Neutron Flux Distribution at 50 GWd/MT and Calculated $^{14}C$ Production Rate**

### 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

$$\frac{4.28\,E19 \bullet (6.67\,E13 \bullet 1.0903 + 1.31\,E14 \bullet 0.0423 + 4.11\,E13 \bullet 0.0478) \bullet 1E-24 \bullet \ln 2}{3.7\,E4 \bullet 5730 \bullet 365.25 \bullet 24 \bullet 3600} = 3.557\,E-7\ \mu Ci/\sec - kg - ppm - N$$

### 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 μCi/sec-ppm N or 0.29 Ci/yr-ppm N

Domestic BWRs are expected to have reactor coolant nitrogen concentrate much less 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/cm$^2$-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.

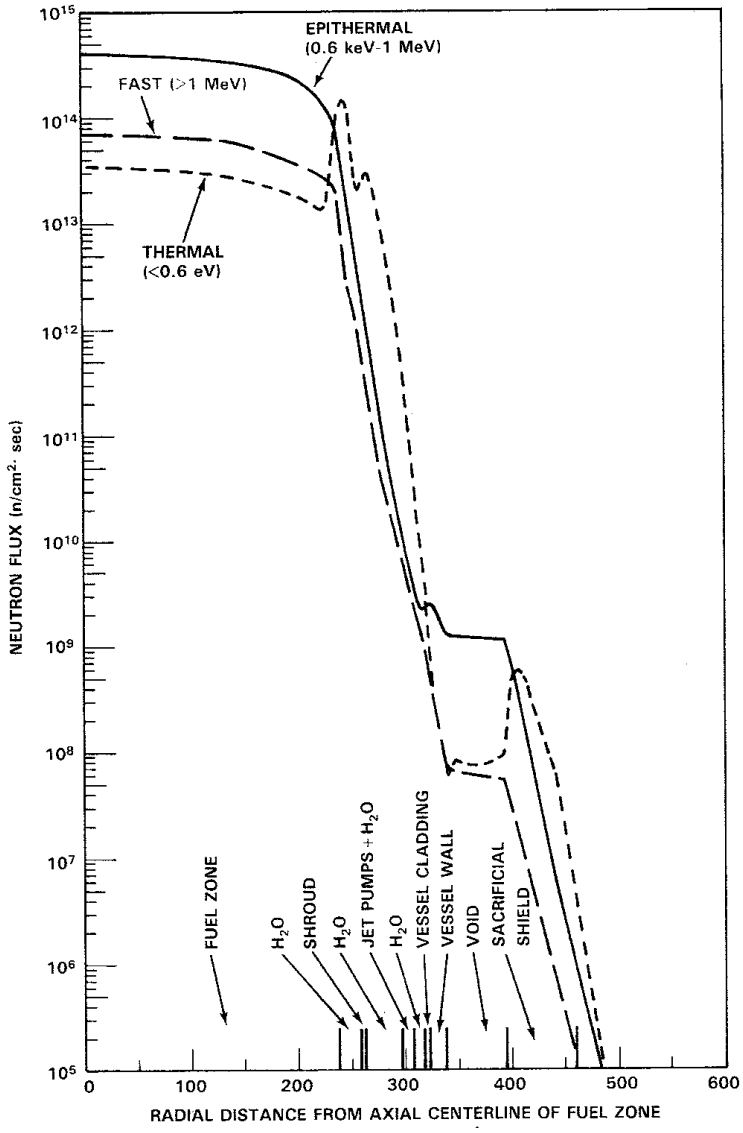


**Figure A-5**
**Radial Three Group Neutron Flux Distribution at Core Axial Mid-Plane, ANISN Calculation for General Electric BWR (Evans, et al.,1984)**

# 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 $^{14}C$ 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 $^{17}O(n,\alpha)^{14}C$ 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% | 28891.80027 | 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 | 1.4033E+14 | 5.08553E+13 |
| | MWd/MTU | MWd/MTU | Mlbm/hr | – | lbm | lbm | lbm | $\phi < 0.625$ ev | 0.625 eV $< \phi <$ 1 MeV | $\phi > 1$ MeV |

## Table B-2
## BWR Power Station Calculated $^{14}C$ Source Term

| | | Moderator Region | | | Bypass Region | | | |
|---|---|---|---|---|---|---|---|---|
| O-17(n,alpha)C-14 "effective" cross-section, b | | 0.1325 | 0.0238 | 0.1106 | 0.1386 | 0.0222 | 0.1106 | |
| | | | | **Production Rate, uCi/sec** | | | | |
| Cycle | 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 |
| | Cycle Average: | 0.0636 | 0.0555 | 0.0985 | 0.0920 | 0.0451 | 0.0786 | 0.4333 |
| | % in Flux/Core Region: | 29.24 | 25.49 | 45.28 | 42.64 | 20.93 | 36.43 | |

Sum: uCi/sec 0.4333
Ci/yr 13.67

| | | Moderator Region | | | Bypass Region | | | |
|---|---|---|---|---|---|---|---|---|
| N-14(n,p)C-14 "effective" cross-section, b | | 1.056 | 0.0384 | 0.0479 | 1.0903 | 0.0423 | 0.0478 | |
| | | | | **Production Rate, uCi/sec-ppm N** | | | | |
| Cycle | 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 |
| | Cycle Average: | 0.00171 | 0.00030 | 0.00014 | 0.00244 | 0.00029 | 0.00011 | 0.00500 |
| | % in Flux/Core Region: | 79.32 | 14.00 | 6.68 | 85.78 | 10.20 | 4.03 | |

Sum: uCi/sec-ppm N 0.0050
Ci/yr-ppm N 0.16

# C
# PWR $^{14}$C 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 ZIRLO™ cladding. The fuel assemblies contained 128 $ZrB_2$ 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 ºF |
| Core Inlet Temperature | 556.9 ºF |
| 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 $^{14}$C 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 $^{14}$C production reactions are given below.

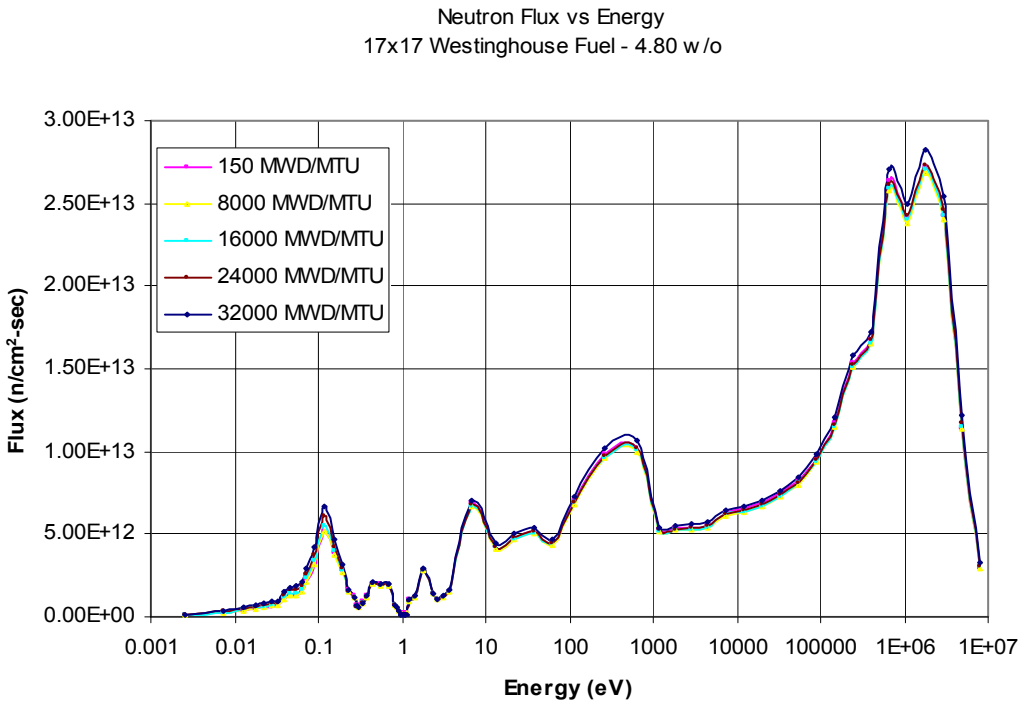


**Figure C-1**
**Sample of the 70 Group Neutron Spectrum at Various Burnups – 4.20 w/o Fuel (Secker (2009))**

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

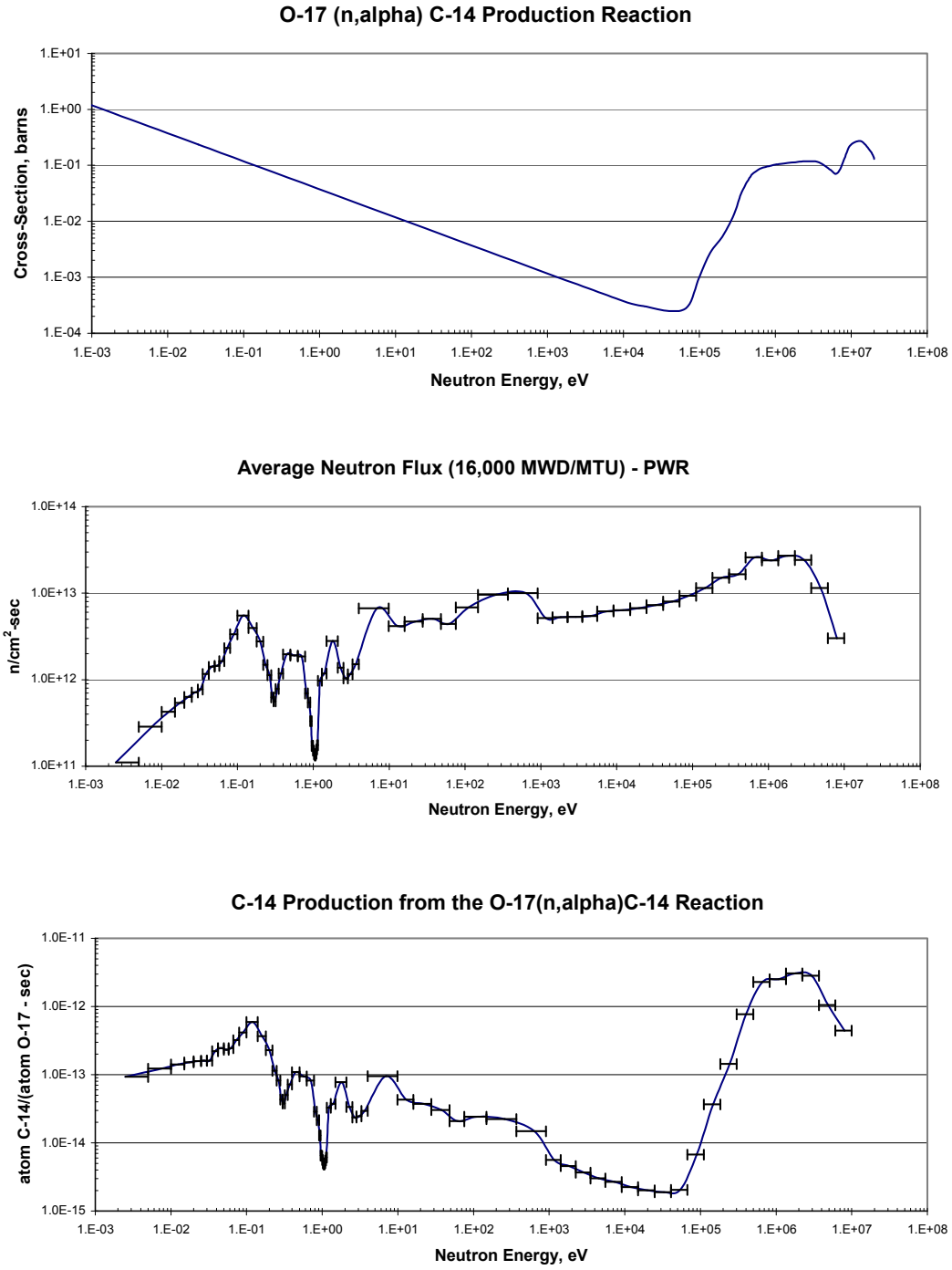


**Figure C-2**
**$^{17}O(n,\alpha)^{14}C$ Reaction Cross-Section, Reference PWR Neutron Flux Distribution at 16 GWd/MT and Calculated $^{14}C$ Production**

**Table C-3**
**$^{17}O(n,\alpha)^{14}C$ "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 $^{14}N(n,p)^{14}C$ 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.

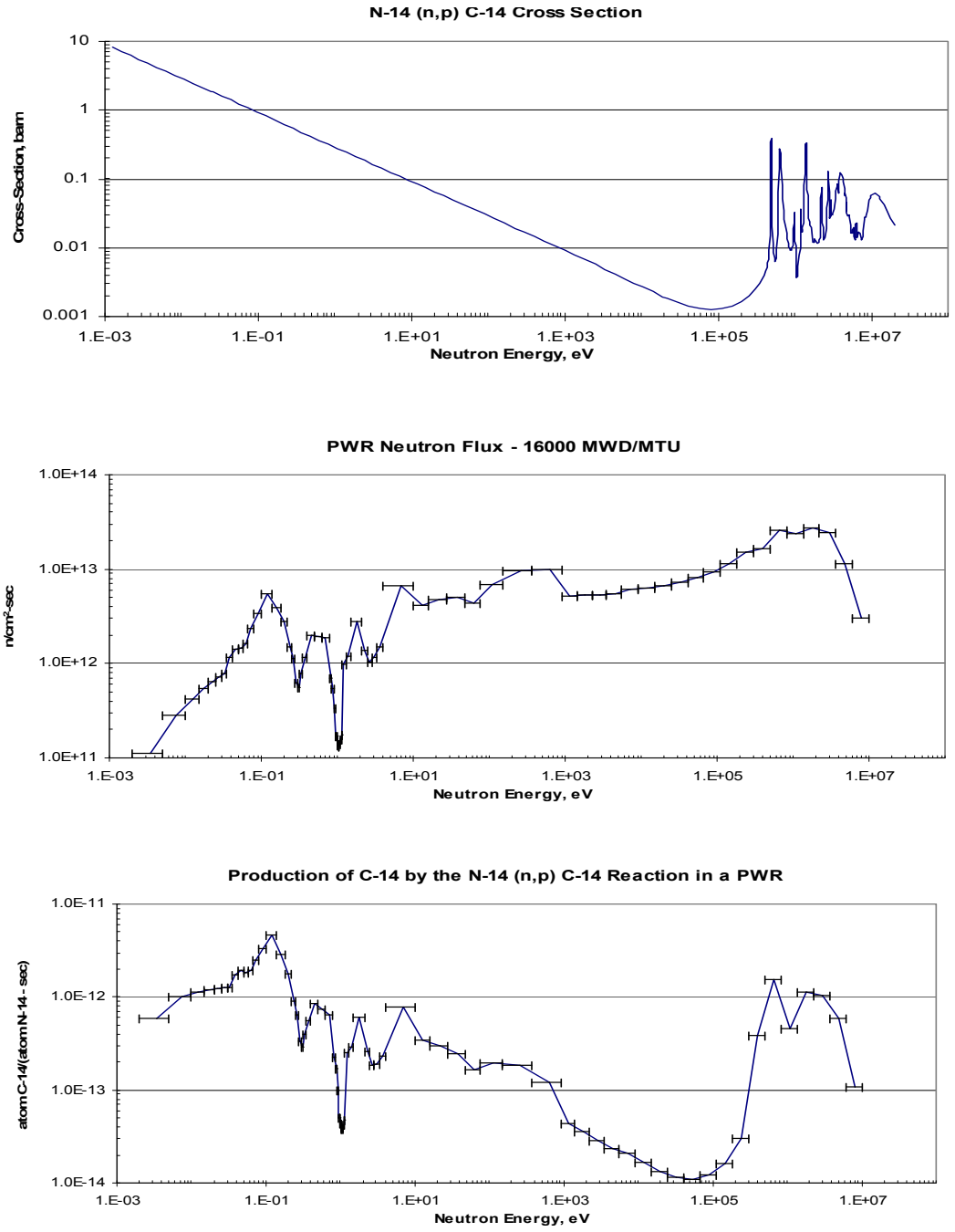


**Figure C-3**
**$^{14}N(n,p)^{14}C$ Reaction Cross-Section, Reference PWR Neutron Flux Distribution at 16 GWd/MT and Calculated $^{14}C$ Production**

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 $^{14}C$ source term calculations.

**Table C-4**
**$^{14}N(n,p)^{14}C$ "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 | 0.9188 | 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, $n/cm^2$-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.

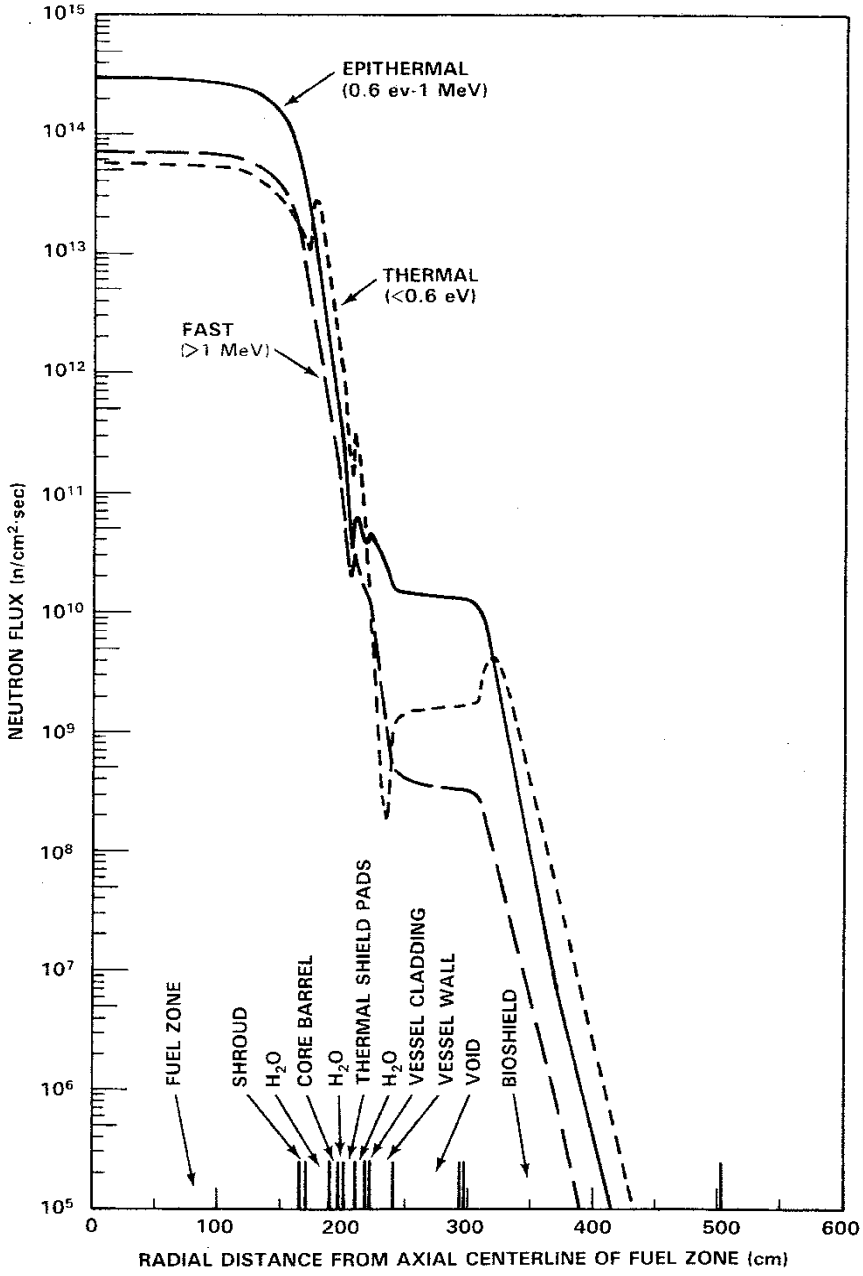


**Figure C-4**
**Radial Three Group Neutron Flux Distribution at Core Axial Mid-Plane, ANISN Calculation for Westinghouse PWR (Evans, et al., 1984)**

# D
# PWR SITE SPECIFIC SOURCE TERM CALCULATION

Summary of Appendix D PWR $^{14}C$ Source Term Calculations

Westinghouse Reactors

| | | | $^{17}O(n,\alpha)^{14}C$ | $^{14}N(n,p)^{14}C$ |
|---|---|---|---|---|
| | **$MW_{th}$** | **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

| | | | $^{17}O(n,\alpha)^{14}C$ | $^{14}N(n,p)^{14}C$ |
|---|---|---|---|---|
| | **$MW_{th}$** | **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[a]** |
| | | | 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 ºF.

**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** | |
| $^{17}O(n,\alpha)^{14}C$ | 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 µCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux (≤0.625 eV), n/cm²-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | intermediate plus fast neutron flux (>0.625 eV), n/cm²-sec |
| 1.0E-24 | = | Conversion factor, 1.0E-24 cm²/b |
| $\lambda$ | = | $^{14}C$ decay constant, 3.833E-12/sec |
| 3.7E4 | = | Conversion factor, 3.7E4 d/sec-µCi |

BOC calculation

$$PR = \frac{1.27\,E22 \bullet [0.121 \bullet 3.18\,E13 + 0.0479 \bullet 2.78\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$

$$= 2.258\,E-15\,\mu Ci/sec-kg$$

EOC calculation

$$PR = \frac{1.27\,E22 \bullet [0.121 \bullet 3.96\,E13 + 0.0479 \bullet 2.90\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$

$$= 2.458\,E-5\,\mu Ci/sec-kg$$

The cycle average $^{14}C$ production rates for the $^{17}O(n,\alpha)^{14}C$ reaction were as follows:

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

| | Production Rate, µCi/sec-kg |
|---|---|
| BOC | 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 $^{17}O(n,\alpha)^{14}C$ reaction is:

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

or

$$[2.36E\text{-}5\ \mu Ci/sec\text{-}kg \bullet 13{,}498\ kg \bullet 3.6E3\ sec/hr]/3216\ MW_{th} = 0.357\ \mu Ci/MW_{th}\text{-}h$$

or

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

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

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | Intermediate plus fast neutron flux, n/cm$^2$-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-µCi |

BOC calculation:

$$PR = \frac{4.28\,E19 \bullet [0.951 \bullet 3.18\,E13 + 0.0392 \bullet 2.78\,E14] \bullet 1.0\,E-24 \bullet 3.833\,E-12}{3.7\,E4}$$
$$= 1.826\,E-7\,\mu Ci/sec-kg-ppm \cdot N$$

EOC calculation:

$$PR = \frac{4.28\,E19 \bullet [0.951 \bullet 3.96\,E13 + 0.0392 \bullet 2.90\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$

$$= 2.176\,E-7\,\mu Ci/sec-kg$$

The production rate are summarized below:

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

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

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

2.00E-7 µCi/sec-kg-ppm N • 13,498 kg • 3.156E7 sec/yr = 8.52E4 µCi/yr-ppm N

= 0.085 Ci/yr-ppm-N

or

[2.00E-7 µCi/sec-kg-ppm N • 13,498 kg • 3.6E3 sec/hr]/3216 $MW_{th}$

= 3.02E-3 µCi/$MW_{th}$-h–ppm N

or

3.02E-3 µCi/$MW_{th}$-h-ppm N • 8,766 h/yr • $MW_{th}$/0.34 $MW_e$ • 1E3 $MW_e$/$GW_e$ • Ci/1E6 µCi

= 0.078 Ci/$GW_e$-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 ºF.

**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** | |
| $^{17}O(n,\alpha)^{14}C$ | 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 μCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction:

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux (≤0.625 eV), n/cm²-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | Intermediate plus 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:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.21E13 + 0.0479 \bullet 2.77E14] \bullet 1.0E-24 \bullet 3.833E-12}{3.7E4}$$

$$= 2.257E-5 \mu Ci / sec-kg$$

EOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.99E13 + 0.0479 \bullet 2.89E14] \bullet 1.0E-24 \bullet 3.833E-12}{3.7E4}$$

$$= 2.456E-5 \mu Ci / sec-kg$$

The calculated cycle average $^{14}C$ production rate using the above data is:

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

| | Production Rate, µCi/sec-kg |
|---|---|
| BOC | 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 $^{17}O(n,\alpha)^{14}C$ reaction is:

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

or

$$[2.36E\text{-}5\ \mu Ci/sec\text{-}kg \bullet 13{,}498\ kg \bullet 3.6E3\ sec/hr]/3188\ MW_{th} = 0.360\ \mu Ci/MW_{th}\text{-}h$$

or

$$0.360\ \mu Ci/MW_{th}\text{-}h \bullet 8{,}766\ h/yr \bullet MW_{th}/0.34\ MW_e \bullet 1E3\ MW_e/GW_e \bullet Ci/1E6\ \mu Ci$$

$$= 9.28\ Ci/GW_e\text{-}yr$$

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

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | intermediate plus fast neutron flux, n/cm$^2$-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-µCi |

BOC calculation:

$$PR = \frac{4.284\text{E}19 \bullet [0.951 \bullet 3.21\text{E}13 + 0.0392 \bullet 2.77\text{E}14] \bullet 1.0\text{E} - 24 \bullet 3.833\text{E} - 12}{3.7\text{E}4}$$

$$= 1.837\text{E} - 7\mu\text{Ci} / \text{sec} - \text{kg} - \text{ppm} \cdot \text{N}$$

EOC calculation

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

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

The calculated production rate using the above data is:

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

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

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

2.01E-7 µCi/sec-kg-ppm N • 13,498 kg • 3.156E7 sec/yr = 8.56E4 µCi/yr-ppm N

= 0.086 Ci/yr-ppm-N

or [2.01E-7 µCi/sec-kg-ppm N • 13,498 kg • 3.6E3 sec/hr]/3188 $MW_{th}$ =

= 3.06E-3 µCi/$MW_{th}$-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 ºF |
| Core Average Outlet Moderator Temperature, HFP | 595.4 ºF |
| Vessel Average Temperature, HFP<br>(T-Hot-HFP + T-Cold-HFP)/2 | 569.5 ºF |
| Vessel Average Outlet Temperature, HFP | 593.9 ºF |
| 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**<br>**≤0.625 eV** | **Intermediate**<br>**0.625 eV to 1 MeV** | **Fast**<br>**>1 MeV** |
|---|---|---|---|---|
| | **Exposure, MWD/MTU** | **Neutron Flux, n/barn-sec** | | |
| BOC | 150 | 0.34194E-10 | 0.23830e-09 | 0.64388E-10 |
| MOC | 8000 | 0.36723E-10 | 0.24257E-09 | 0.65711E-10 |
| EOC | 15500 | 0.41123E-10 | 0.24465E-09 | 0.66194E-10 |
| | | | | |
| | | **Neutron Flux, n/cm$^2$-sec** | | |
| BOC | 150 | 3.419E13 | 2.383E14 | 6.439E13 |
| MOC | 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 ºF, 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 $^{17}O(n,\alpha)^{14}C$ 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 μCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux (≤0.625 eV), n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | intermediate neutron flux (0.625 eV to 1 MeV), n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | 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 |

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:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.672E13 + 0.0291 \bullet 2.426E14 + 0.1124 \bullet 6.571E13] \bullet 1.0E-24 \bullet 3.833E-12}{3.7E4}$$

$$= 2.485E-5 \mu Ci/sec-kg$$

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 $^{14}C$ production rate using the above data is:

**Table D-10**
**CE-A Cycle 20 Average Production Rates for the $^{17}O(n,\alpha)^{14}C$ Reaction**

| | Production Rate, µCi/sec-kg |
|---|---|
| BOC | 2.409E-5 |
| MOC | 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 µCi/sec-kg • 14,071 kg • 3.156E7 sec/yr = 1.11E7 µCi/yr (11.1 Ci/yr)

or [2.49E-5 µCi/sec-kg • 14,071 kg • 3.6E3 sec/hr]/2700 $MW_{th}$ = 0.467 µCi/$MW_{th}$-h

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

**Table D-11**
**"Effective" Cross-Section for the $^{14}N(n,p)^{14}C$ 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 |

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/$cm^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | intermediate neutron flux (0.625 eV to 1 MeV), n/$cm^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux, n/$cm^2$-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-µCi |

BOC calculation:

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 3.419E13 + 0.0379 \bullet 2.383E14 + 0.0436 \bullet 6.439E13] \bullet 1.0E-24 \bullet 3.833E-12}{3.7E4}$$

$$= 1.968E-7 \mu Ci / sec - kg - ppm \cdot N$$

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

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

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

2.11E-7 µCi/sec-kg-ppm N • 14,071 kg • 3.156E7 sec/yr = 9.37E4 µCi/yr-ppm N

= 0.094 Ci/yr-ppm-N

or

[2.11E-7 µCi/sec-kg-ppm N • 14,071 kg • 3.6E3 sec/hr]/2700 $MW_{th}$ =

= 3.96E-3 µCi/$MW_{th}$-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<br>(T-Hot-HFP + T-Cold-HFP)/2 | 587.0 ºF |
| Vessel Average Outlet Temperature, HFP | 617.5 ºF |

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** | **Neutron Flux, n/barn-sec** | | |
| BOC | 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/$cm^2$-sec** | | |
| BOC | 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 ºF, the coolant mass is 13,567 kg (density = 0.7051 kg/liter).

**Table D-15**
**"Effective" Cross-Section for the $^{17}O(n,\alpha)^{14}C$ 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 μCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux (≤0.625 eV), n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | intermediate neutron flux (0.625 eV to 1 MeV), n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | 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 |

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:

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

The calculated cycle average $^{14}C$ production rate using the above data is:

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

| | Production Rate, µCi/sec-kg |
|---|---|
| BOC | 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 $^{17}O(n,\alpha)^{14}C$ reaction is:

3.23E-5 µCi/sec-kg • 13,567 kg • 3.156E7 sec/yr = 1.38E7 µCi/yr (13.8 Ci/yr)

or

[3.23E-5 µCi/sec-kg • 13,567 kg • 3.6E3 sec/hr]/3650 $MW_{th}$ = 0.432 µCi/$MW_{th}$-h

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

**Table D-17**
**"Effective" Cross-Section for the $^{14}N(n,p)^{14}C$ 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 |

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/$cm^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | intermediate neutron flux (0.625 eV to 1 MeV), n/$cm^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux, n/$cm^2$-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-µCi |

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:

$$PR = \frac{4.284\,E19 \bullet [0.951 \bullet 4.247\,E13 + 0.0379 \bullet 3.376\,E14 + 0.0436 \bullet 9.189\,E13] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$
$$= 2.538\,E-7 \mu Ci / sec - kg - ppm \cdot N$$

The calculated production rate using the above data is:

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

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

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

2.38E-7 µCi/sec-kg-ppm N • 13,567 kg • 3.156E7 sec/yr = 1.02E5 µCi/yr-ppm N

= 0.102 Ci/yr-ppm-N

or

[2.38E-7 µCi/sec-kg-ppm N • 13,567 kg • 3.6E3 sec/hr]/3650 $MW_{th}$

= 3.18E-3 µCi/$MW_{th}$-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 ºF 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 $cm^2$/barn.

**Table D-19**
**W-D Core Flux Distribution**

| | | **n/barn-sec** | |
|---|---|---|---|
| | **MWD/MTU** | **≤0.625 eV** | **>0.625 eV** |
| BOC | 150 | 2.98E-11 | 2.55E-10 |
| MOC | 8000 | 3.15E-11 | 2.59E-10 |
| EOC | 14000 | 3.41E-11 | 2.61E-10 |
| | | **n/$cm^2$-sec** | |
| | **MWD/MTU** | **≤0.625 eV** | **>0.625 eV** |
| BOC | 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/barn-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/$cm^2$-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 $ft^3$ of water in the active fuel region was provided.

Using a specific volume of 0.02112 $ft^3$/lb, the active coolant volume was estimated to be 17,140 lbs or 7775 kg. Cross sections used in the calculations of $^{14}C$ production rates were as follows:

**Table D-21**
**Effective Cross Sections**

| | "Effective" Cross-Section, b | |
|---|---|---|
| Reaction | ≤0.625 eV | >0.625 eV |
| $^{17}O(n,\alpha)^{14}C$ | 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 μCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux (≤0.625 eV), n/$cm^2$-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | 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 |

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 $^{14}C$ production rate using the above data is:

**Table D-22**
**W-D Average Production Rates for the $^{17}O(n,\alpha)^{14}C$ 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 $^{17}O(n,\alpha)^{14}C$ reaction is:

2.13E-5 µCi/sec-kg • 7.775E3 kg • 3.156E7 sec/yr = 5.23E6 µCi/yr (5.23 Ci/yr)

or

[2.13E-5 µCi/sec-kg • 7.775E3 kg • 3.6E3 sec/hr]/1540 $MW_{th}$ = 0.387 µCi/$MW_{th}$-h

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

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/$cm^2$-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | intermediate plus fast neutron flux, n/$cm^2$-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-µCi |

BOC calculation:

$$PR = \frac{4.284\ E19 \bullet [0.951 \bullet 2.98\ E13 + 0.0392 \bullet 2.55\ E14] \bullet 1.0E-24 \bullet 3.833\ E-12}{3.7E4}$$
$$= 1.701\ E-7\,\mu Ci/sec-kg-ppm \cdot N$$

MOC calculation:

$$PR = \frac{4.284\ E19 \bullet [0.951 \bullet 3.15\ E13 + 0.0392 \bullet 2.59\ E14] \bullet 1.0E-24 \bullet 3.833\ E-12}{3.7E4}$$
$$= 1.780\ E-7\,\mu Ci/sec-kg-ppm \cdot N$$

EOC calculation:

$$PR = \frac{4.284\ E19 \bullet [0.951 \bullet 3.41\,E13 + 0.0392 \bullet 2.61\,E14] \bullet 1.0E-24 \bullet 3.833\ E-12}{3.7E4}$$
$$= 1.893\ E-7\,\mu Ci/sec-kg-ppm \cdot N$$

The calculated production rate using the above data is:

**Table D-23**
**W-D Average Production Rates for the $^{14}N(n,p)^{14}C$ 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 $^{14}C$ produced by the $^{14}N(n,p)^{14}C$ reaction is:

1.79E-7 µCi/sec-kg-ppm N • 7.775E3 kg • 3.156E7 sec/yr = 4.39E4 µCi/yr-ppm N

= 0.044 Ci/yr-ppm-N

or

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

= 3.25E-3 µCi/$MW_{th}$-h–ppm N

## *5.2 Westinghouse-E (W-E)*

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

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | Thermal neutron flux (≤0.625 eV), n/$cm^2$-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | Intermediate plus fast neutron flux (>0.625 eV), n/$cm^2$-sec |
| 1.0E-24 | = | Conversion factor, 1.0E-24 $cm^2$/b |
| $\lambda$ | = | $^{14}C$ decay constant, 3.833E-12/sec |
| 3.7E4 | = | Conversion factor, 3.7E4 d/sec-µCi |

BOC calculation:

$$PR = \frac{1.27\,E22 \bullet [0.121 \bullet 2.95\,E13 + 0.0479 \bullet 2.55\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$
$$= 2.077\,E-5\,\mu Ci/sec-kg$$

MOC calculation:

$$PR = \frac{1.27\,E22 \bullet [0.121 \bullet 3.09\,E13 + 0.0479 \bullet 2.59\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$
$$= 2.124\,E-5\,\mu Ci/sec-kg$$

EOC calculation:

$$PR = \frac{1.27\,E22 \bullet [0.121 \bullet 3.34\,E13 + 0.0479 \bullet 2.61\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$
$$= 2.177\,E-5\,\mu Ci/sec-kg$$

The calculated cycle average $^{14}C$ 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 $^{17}O(n,\alpha)^{14}C$ reaction is:

2.13E-5 µCi/sec-kg • 7.775E3 kg • 3.156E7 sec/yr = 5.23E6 µCi/yr (5.23 Ci/yr)

or

[2.13E-5 µCi/sec-kg • 7.775E3 kg • 3.6E3 sec/hr]/1540 $MW_{th}$ = 0.387 µCi/$MW_{th}$-h

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

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | intermediate plus fast neutron flux, n/cm$^2$-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-μCi |

BOC calculation:

$$PR = \frac{4.284\ E19 \bullet [0.951 \bullet 2.95\,E13 + 0.0392 \bullet 2.55\,E14] \bullet 1.0E-24 \bullet 3.833\ E-12}{3.7E4}$$

$$= 1.689\ E-7 \mu Ci / sec - kg - ppm \cdot N$$

MOC calculation:

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

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

EOC calculation:

$$PR = \frac{4.284\ E19 \bullet [0.951 \bullet 3.34\,E13 + 0.0392 \bullet 2.61\,E14] \bullet 1.0E-24 \bullet 3.833\ E-12}{3.7E4}$$

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

The calculated production rate using the above data is:

**Table D-25**
**W-E Average Production Rates for the $^{14}N(n,p)^{14}C$ 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 $^{14}C$ produced by the $^{14}N(n,p)^{14}C$ reaction is:

1.77E-7 μCi/sec-kg-ppm N • 7.775E3 kg • 3.156E7 sec/yr = 4.34E4 μCi/yr-ppm N

= 0.043 Ci/yr-ppm-N

or

[1.77E-7 μCi/sec-kg-ppm N • 7.775E3 kg • 3.6E3 sec/hr]/1540 $MW_{th}$ =

= 3.22E-3 μCi/$MW_{th}$-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 ºF.

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 | E≥1 MeV |
| | Core Average Flux, n/cm²-sec | | |
| BOC | 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 $^{17}O(n,\alpha)^{14}C$ 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 μCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux (≤0.625 eV), n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | intermediate neutron flux (0.625 eV to 1 MeV), n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux (>0.625 eV), n/cm$^2$-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-μCi |

BOC calculation:

$$PR = \frac{1.27E22 \bullet [0.121 \bullet 3.672E13 + 0.0291 \bullet 2.2267E14 + 0.1124 \bullet 8.1527E13] \bullet 1.0E-24 \bullet 3.833E-12}{3.7E4}$$

$$= 2.643E-5 \mu Ci/sec-kg$$

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 $^{14}C$ production rate using the above data is:

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

| | Production Rate, μCi/sec-kg |
|---|---|
| BOC | 2.643E-5 |
| MOC | 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 $^{17}O(n,\alpha)^{14}C$ reaction is:

2.74E-5 μCi/sec-kg • 13,868 kg • 3.156E7 sec/yr = 1.20E7 μCi/yr (12.0 Ci/yr)

or

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

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

**Table D-29**
**"Effective" Cross-Section for the $^{14}N(n,p)^{14}C$ 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 |

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | intermediate neutron flux (0.625 eV to 1 MeV), n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux, 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 |

BOC calculation:

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 3.673E13 + 0.0379 \bullet 3.227E14 + 0.0436 \bullet 8.8153E13] \bullet 1.0E-24 \bullet 3.833E-12}{3.7E4}$$

$$= 2.083E-7 \mu Ci/sec-kg-ppm \cdot N$$

MOC calculation:

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 3.828E13 + 0.0379 \bullet 2.293E14 + 0.0436 \bullet 8.394E13] \bullet 1.0E-24 \bullet 3.833E-12}{3.7E4}$$

$$= 2.164E-7 \mu Ci/sec-kg-ppm \cdot N$$

EOC calculation:

$$PR = \frac{4.284E19 \bullet [0.951 \bullet 4.296E13 + 0.0379 \bullet 2.337E14 + 0.0436 \bullet 8.8555E13] \bullet 1.0E-24 \bullet 3.833E-12}{3.7E4}$$

$$= 2.372E-7 \mu Ci/sec-kg-ppm \cdot N$$

The calculated production rate using the above data is:

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

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

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

$$2.21E\text{-}7\ \mu Ci/sec\text{-}kg\text{-}ppm\ N \bullet 13{,}868\ kg \bullet 3.156E7\ sec/yr = 9.67E4\ \mu Ci/yr\text{-}ppm\ N$$

$$= 0.097\ Ci/yr\text{-}ppm\text{-}N$$

or

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

## 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 ºF.

Site personnel provided the following neutron flux data:

**Table D-31**
**CE-B Core Average Neutron Flux**

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

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

**Table D-32**
**"Effective" Cross-Section for the $^{17}O(n,\alpha)^{14}C$ 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 μCi/sec-kg for the $^{17}O(n,\alpha)^{14}C$ reaction.

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux (≤0.625 eV), n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | Intermediate neutron flux (0.625 eV to 1 MeV), n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | 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 |

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 $^{14}C$ production rate using the above data is:

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

| | Production Rate, µCi/sec-kg |
|---|---|
| BOC | 2.633E-5 |
| MOC | 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 $^{17}O(n,\alpha)^{14}C$ reaction is:

2.79E-5 µCi/sec-kg • 15,610 kg • 3.156E7 sec/yr = 1.37E7 µCi/yr (13.7 Ci/yr)

or

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

or at an assuming 34% efficiency

0.422 µCi/$MW_{th}$-h • $MW_{th}$/0.34 $MW_e$ • 8,766 h/yr = 1.09E4 µCi/$MW_e$-yr (10.9 Ci/$GW_e$-yr)

$^{14}N(n,p)^{14}C$ Reaction Source Term Calculation:

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

**Table D-34**
**"Effective" Cross-Section for the $^{14}N(n,p)^{14}C$ 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 |

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_i$ | = | "effective" intermediate cross-section, b |
| $\varphi_i$ | = | Intermediate neutron flux (0.625 eV to 1 MeV), n/cm$^2$-sec |
| $\sigma_f$ | = | "effective" fast cross-section, b |
| $\varphi_f$ | = | fast neutron flux, n/cm$^2$-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-µCi |

BOC calculation:

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

$$= 2.330\,E-7\,\mu Ci/sec-kg-ppm \cdot N$$

MOC calculation:

$$PR = \frac{4.284\,E19 \bullet [0.951 \bullet 4.61\,E13 + 0.0379 \bullet 2.18\,E14 + 0.0436 \bullet 8.06\,E13] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$

$$= 2.468\,E-7\,\mu Ci/sec-kg-ppm \cdot N$$

EOC calculation:

$$PR = \frac{4.284\,E19 \bullet [0.951 \bullet 5.10\,E13 + 0.0379 \bullet 2.33\,E14 + 0.0436 \bullet 8.591\,E13] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$

$$= 2.711\,E-7\,\mu Ci/sec-kg-ppm \cdot N$$

The calculated production rate using the above data is:

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

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

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

2.50E-7 µCi/sec-kg-ppm N • 15,610 kg • 3.156E7 sec/yr = 1.23E5 µCi/yr-ppm N

= 0.123 Ci/yr-ppm N

or

$$[2.50E\text{-}7\ \mu Ci/sec\text{-}kg\text{-}ppm\ N \bullet 15{,}610\ kg \bullet 3.6E3\ sec/hr]/3716\ MW_{th}$$

$$= 3.78E\text{-}3\ \mu Ci/MW_{th}\text{-}h\text{-}ppm\ N$$

## 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 ºF.

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

| | **"Core Average" Neutron Flux, n/cm²-sec** | |
|---|---|---|
| | ≤0.625 eV | >0.625 eV |
| BOC | 4.202E13 | 3.148E14 |
| MOC | 4.518E13 | 3.237E14 |
| EOC | 5.069E13 | 3.260E14 |
| **Reaction** | **"Effective" Cross-Section[a], b** | |
| $^{17}O(n,\alpha)^{14}C$ | 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.

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

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

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

where:

| | | |
|---|---|---|
| N | = | 1.27E22 atoms $^{17}O$/kg $H_2O$ |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux (≤0.625 eV), n/cm²-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | Intermediate plus fast neutron flux (>0.625 eV), n/cm²-sec |
| 1.0E-24 | = | Conversion factor, 1.0E-24 cm²/b |
| $\Lambda$ | = | $^{14}C$ decay constant, 3.833E-12/sec |
| 3.7E4 | = | Conversion factor, 3.7E4 d/sec-µCi |

BOC calculation:

$$PR = \frac{1.27\,E22 \bullet [0.121 \bullet 4.202\,E13 + 0.0479 \bullet 3.148\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$

$$= 2.653\,E-5\,\mu Ci/sec-kg$$

MOC calculation:

$$PR = \frac{1.27\,E22 \bullet [0.121 \bullet 4.518\,E13 + 0.0479 \bullet 3.237\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$

$$= 2.759\,E-5\,\mu Ci/sec-kg$$

EOC calculation:

$$PR = \frac{1.27\,E22 \bullet [0.121 \bullet 5.069\,E13 + 0.0479 \bullet 3.260\,E14] \bullet 1.0E-24 \bullet 3.833\,E-12}{3.7E4}$$

$$= 2.861\,E-5\,\mu Ci/sec-kg$$

The calculated cycle average $^{14}C$ production rate using the above data is:

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

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

The active core volume was calculated to be 710.9 $ft^3$. At the specified core average temperature of 589.5ºF and operating 2250 psi, the "active core" mass is ~14,132 kg ($\rho$ = 0.702 kg/L assumed).

The total $^{14}C$ produced by the $^{17}O(n,\alpha)^{14}C$ reaction is:

2.76E-5 µCi/sec-kg • 14,132 kg • 3.156E7 sec/yr = 12.3E6 µCi/yr (12.3 Ci/yr)

or

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

or

0.387 µCi/$MW_{th}$-h • 8.766E3 h/yr • $MW_{th}$/0.34 $MW_e$ • 1E3 $MW_e$/$GW_e$ • Ci/1E6 µCi

= 9.98 Ci/$GW_e$-yr

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

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

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

where:

| | | |
|---|---|---|
| N | = | 4.284E19 atoms $^{14}N$/kg-ppm N |
| $\sigma_{th}$ | = | "effective" thermal cross-section, b |
| $\varphi_{th}$ | = | thermal neutron flux, n/cm$^2$-sec |
| $\sigma_{i+f}$ | = | "effective" intermediate plus fast cross-section, b |
| $\varphi_{i+f}$ | = | intermediate plus fast neutron flux, n/cm$^2$-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-µCi |

BOC calculation:

$$PR = \frac{4.284\ E19 \bullet [0.951 \bullet 4.202\ E13 + 0.0392 \bullet 3.148\ E14] \bullet 1.0E-24 \bullet 3.833\ E-12}{3.7E4}$$
$$= 2.321\ E-7 \mu Ci/sec-kg-ppm \cdot N$$

MOC calculation:

$$PR = \frac{4.284\ E19 \bullet [0.951 \bullet 4.518\ E13 + 0.0392 \bullet 3.237\ E14] \bullet 1.0E-24 \bullet 3.833\ E-12}{3.7E4}$$
$$= 2.470\ E-7 \mu Ci/sec-kg-ppm \cdot N$$

EOC calculation:

$$PR = \frac{4.284\ E19 \bullet [0.951 \bullet 5.069\ E13 + 0.0392 \bullet 3.260\ E14] \bullet 1.0E-24 \bullet 3.833\ E-12}{3.7E4}$$
$$= 2.707\ E-7 \mu Ci/sec-kg-ppm \cdot N$$

The calculated production rate using the above data is:

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

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

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

2.50E-7 μCi/sec-kg-ppm N • 14,132 kg • 3.156E7 sec/yr = 1.11E5 μCi/yr-ppm N

= 0.111 Ci/yr-ppm-N

or

[2.50E-7 μCi/sec-kg-ppm N • 14,132 kg • 3.6E3 sec/hr]/3625.6 $MW_{th}$ =

= 3.51E-3 μCi/$MW_{th}$-h–ppm N

= 0.090 Ci/$GW_e$-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:

$$P_i = H \bullet X_i \qquad \textbf{Eqn. E-1}$$

where

| | | |
|---|---|---|
| H | = | Henry's Law Constant, atm/mole fraction |
| $P_i$ | = | Partial Pressure of Species i, 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 $N_2$ can be calculated from the following relations based on the values developed from Reference E-1:

$$H = -11.672\ (ºC)^2 + 1897.3\ (ºC) + 46710 \text{ at } 20ºC \text{ to } 50ºC \qquad \textbf{Eqn. E-2}$$

or

$$H = -3.6024\ (ºF)2 + 1284.6\ (ºF) + 9290.5 \text{ at } 68ºF \text{ to } 122ºF \qquad \textbf{Eqn. E-3}$$

## 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 35ºC is calculated to be:

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

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

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

The mole fraction of 3.12E-6 is equivalent to ~4.85 ppm $N_2$, e.g.:

$$\frac{3.12E-6 \text{ moles } N_2}{\text{mole } H_2O} \bullet \frac{\text{mole } H_2O}{18.02 \text{ g } H_2O} \bullet \frac{28.01 \text{ g } N_2}{\text{mole } N_2} = 4.85E\text{-}6 \text{ g N/g } H_2O = 4.85 \text{ ppm}$$

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 $^{14}C$ source term. For example, if the coolant contained 1 ppm $NH_3$, the total nitrogen concentration would be

$$\text{Total Nitrogen} = 4.85 + 1\left(\frac{14}{17}\right) = 5.67 \text{ ppm}$$

## 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 $^{14}C$ (primarily methane) in the RCS and the VCT will be reduced.
- The formation of $^{14}C$ 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 $^{14}C$ gaseous releases will be from the gas treatment systems.
- For PWRs without recombiners in the gas treatment system, a greater fraction of the released $^{14}C$ will be organic which will reduce local environmental radiation exposures.
- The gas phase concentration of $^{14}C$ 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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