EPRI | ELECTRIC POWER RESEARCH INSTITUTE

Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents

2010 TECHNICAL REPORT

Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents

1021106

Final Report, December 2010

EPRI Project Manager
K. Kim

ELECTRIC POWER RESEARCH INSTITUTE
3420 Hillview Avenue, Palo Alto, California 94304-1338 • PO Box 10412, Palo Alto, California 94303-0813 • USA
800.313.3774 • 650.855.2121 • askepri@epri.com • www.epri.com

DISCLAIMER OF WARRANTIES AND LIMITATION OF LIABILITIES

THE FOLLOWING ORGANIZATION, UNDER CONTRACT TO EPRI, PREPARED THIS REPORT:

NWT Corporation

NOTE

For further information about EPRI, call the EPRI Customer Assistance Center at 800.313.3774 or e-mail askepri@epri.com.

- 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

ACKNOWLEDGMENTS

The following organization, under contract to the Electric Power Research Institute (EPRI), prepared this report:

NWT Corporation
7015 Realm Drive
San Jose, CA 95119

Principal Investigators
G. Palino
H. Helmholz
S. Sawochka

This report describes research sponsored by EPRI.

The following nuclear industry representatives supported the development of this report:

Ron Chrzanowski, Exelon
Drew Odell, Exelon
Ken Sejkora, Entergy
Steve Sandike, Entergy
Greg Barley, Progress
Mike Millinor, Progress
Kevin O'Hare, FPL
Dale Holden, Duke Energy
Richard Motko, Entergy
Kathy Yhip, Southern California Edison
Don E. Adams II, TVA
John Doroski, Dominion
Ralph Andersen, NEI
Jeff Waites, Southern Company
Mary Beth Lloyd, Southern Company
Martin Wright, Pacific Gas & Electric
Doo-Ho Lee, KHNP
Chris Eastus, Entergy
Ken Watson, Transware, Inc.
Paul Prejean, Entergy
Aimey Tregre, Entergy
John Knemeyer, Pacific Gas & Electric
Kjell Johansen, FPL
Bernt Bengtsson, Vattenfall

This publication is a corporate document that should be cited in the literature in the following manner:

Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents. EPRI, Palo Alto, CA: 2010 1021106.

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%.

CONTENTS

LIST OF FIGURES

LIST OF TABLES

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 800°C 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 3-13
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).

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.

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 $\mu Ci/cm^3$. 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 $\mu Ci/ml$ with an average of 1.1E-4 $\mu Ci/ml$. Approximately 0.008 Ci/yr was released to containment via a primary coolant leak.

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

4.4.2 Indian Point Unit 3

At Indian Point Unit-3, all gases were discharged through the plant vent. Continuous samplers were used to measure total ^{14}C and $^{14}CO_2$ 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.

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