Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents

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EPRI Project Objectives

- Review international research and development & data related to Carbon-14 generation and release.

- Review Carbon-14 generation and release mechanisms in nuclear power plants.

- Provide utilities with methodology for accurately estimating carbon-14 generation and release from specific nuclear power plants.

- Benchmark 2-BWRs, 2-W PWRs, 1-CE PWR, 1-B&W PWR

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Calculation of Reactor Coolant C-14 Source Term

- Production reactions

- Target concentrations

- Neutron cross-sections

- Neutron fluxes

- Target quantity in neutron flux

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Production of C-14 in Coolant

Nuclear Reactions (in order of importance)

Reaction	Target Abundance (%)	Target Concentration
$^{17}O \, (n,\alpha) \, ^{14}C$	0.038	1.27E22 atoms ^{17}O/kg H_2O
$^{14}N \, (n,p) \, ^{14}C$	99.632	4.28E19 atoms ^{14}N/kg H_2O - ppm N
$^{13}C \, (n,\gamma) \, ^{14}C$	1.07	5.36E17 atoms ^{13}C/kg H_2O - ppm C

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Possible Chemical Forms Produced from In-Core ^{14}C Production Reactions

Single Carbon Atom Species		Environment
CO_2	Carbon Dioxide	Oxidizing
CO	Carbon Monoxide	
HCOOH	Formic Acid	
$H_2C=O$	Formaldehyde	
CH_3OH	Methanol	
CH_4	Methane	Reducing

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C-14 Production Cross-Sections

Cross-Section for $^{17}O(n,alpha)^{14}C$ Reaction
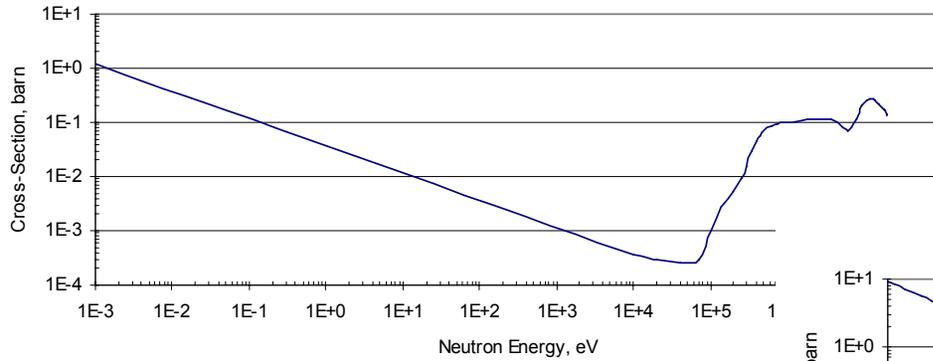

Cross-Section for $^{14}N(n,p)^{14}C$ Reaction

Cross-Section for $^{13}C(n,gamma)^{14}C$ Reaction

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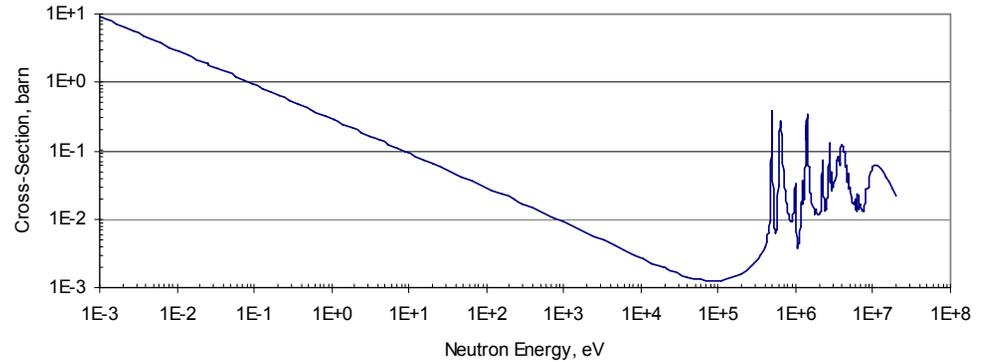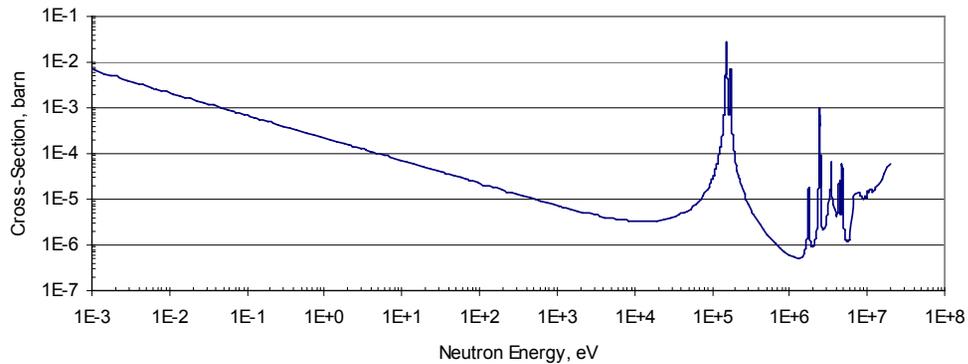
Examples of Neutron Flux Spectra

PWR Neutron Flux – 16 GWd/MTU
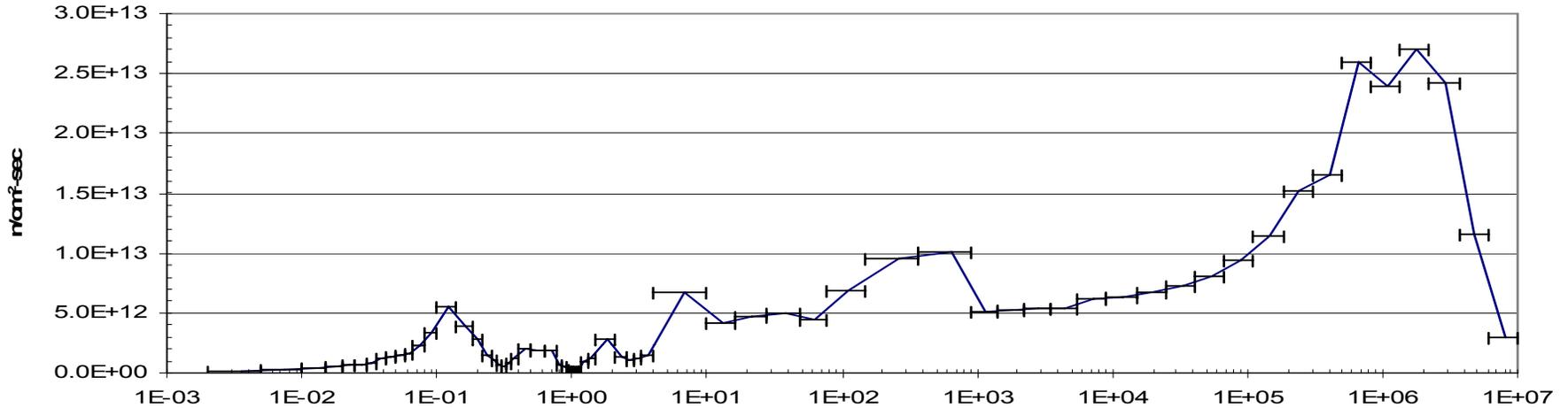
BWR Neutron Flux Distribution – 20 GWd/MTU

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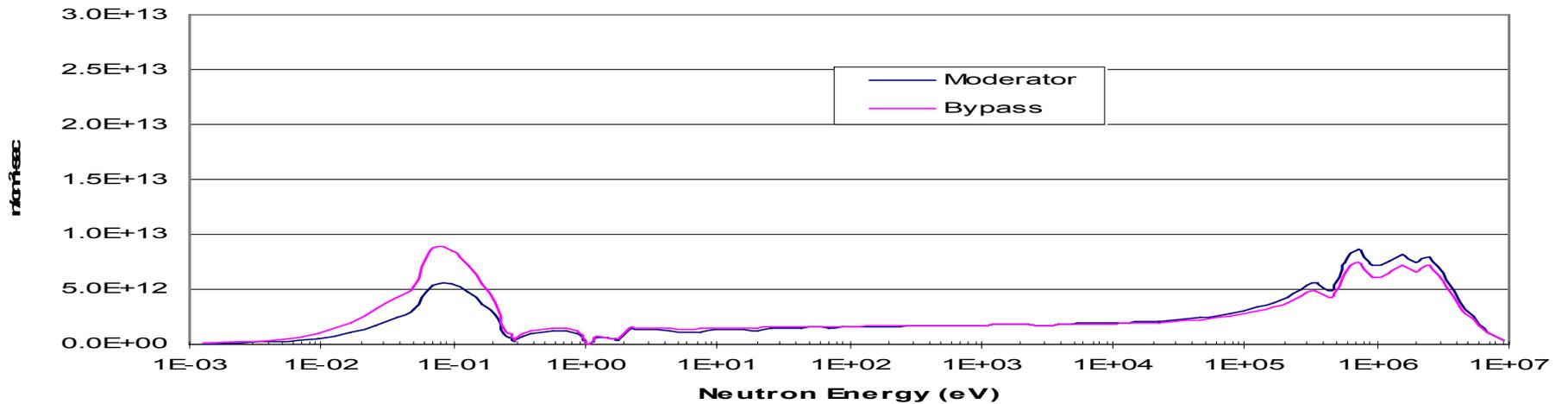
"Effective Mass" of Coolant in Active Core

- BWR

BWR-GALE Code	39,000 kg
Limerick UFSAR	33,975 kg
GE Vessel Drawings	
2894 MW_T, 624 Fuel Assemblies	24,630 kg
3579 MW_T, 748 Fuel Assemblies	29,755 kg
3579 MW_T, 732 Fuel Assemblies	27,805 kg
Oyster Creek, 1860 MWt, 560 Fuel Assemblies	26,254 kg
1593 MW_T, 368 Fuel Assemblies	15,830 kg
Bonka, 1972	33,000 kg
Fowler , 1976, 3579 MW_T BWR/6	39,500 kg

- PWR

Helmholz (Braidwood 1)	14,100 kg
Bonka (1976)	13,400 kg
Fowler (1976) - 3479 MW_T CE Plant	13,700 kg

- <u>Need</u> site specific value to calculate individual unit source term.

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# Example Calculation at 16 GWd/MTU $^{17}O(n,\alpha)^{14}C$ Reaction

O-17 (n,alpha) C-14 Production Reaction
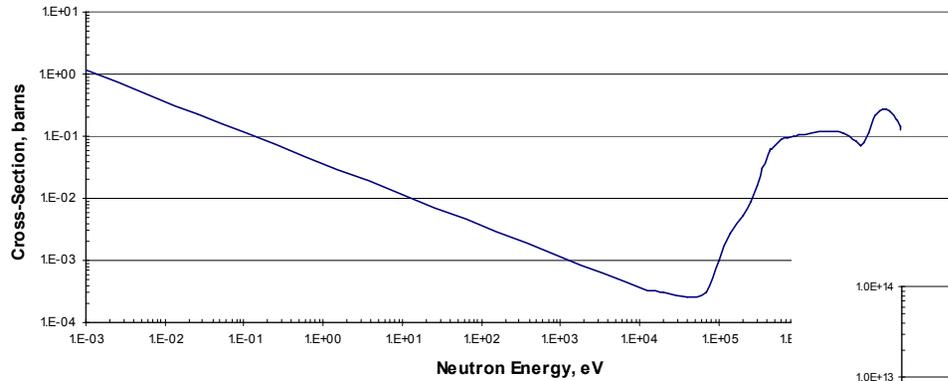
Average Neutron Flux (16,000 MWD/MTU) - PWR
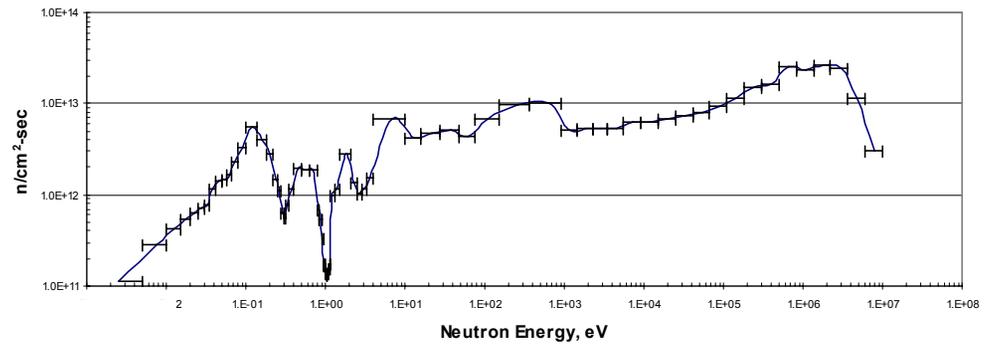
C-14 Production from the O-17(n,alpha)C-14 Reaction

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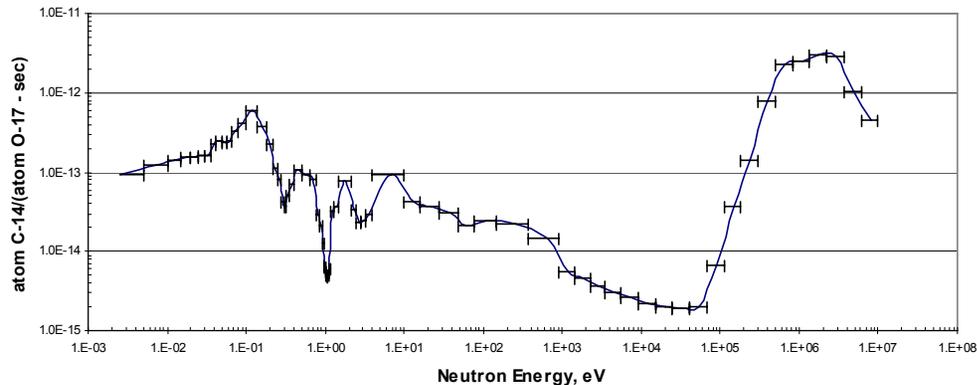
PWR Source Term Example - $^{17}O(n,\alpha)^{14}C$ Reaction

PWR "Effective Cross-Sections" for the $^{17}O(n,\alpha)^{14}C$ Reaction

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.1189
Intermediate	>0.625 eV - <1 MeV	0.0183
Fast	≥1 MeV	0.1504

- C-14 production per kilogram of water is 2.475E-5 µCi/sec-kg

- For the 1178 MW_e (~3549 MW_{TH}) PWR with an estimated coolant mass of 14,100 kg, the total production for this reactor would be approximately:

$$2.475E\text{-}5 * 14{,}100 = 0.349 \text{ µCi/sec}$$
$$= 11.0 \text{ Ci/yr} \qquad = 9.35 \text{ Ci/GW}_e\text{-yr}$$
$$= 13.1 \text{ kBq/MWh}_{TH} \qquad = 0.354 \text{ µCi/MWh}_{TH}$$

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# Example Calculation at 16 GWd/MTU
$^{14}N(n,p)^{14}C$ Reaction


N-14 (n,p) C-14 Cross Section
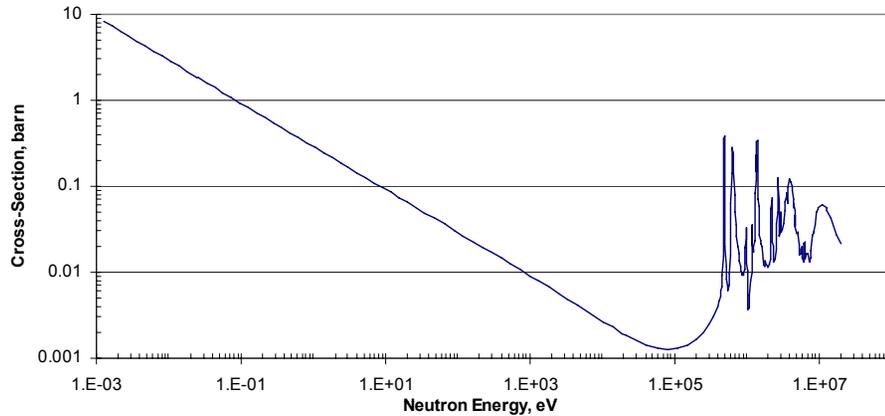
PWR Neutron Flux
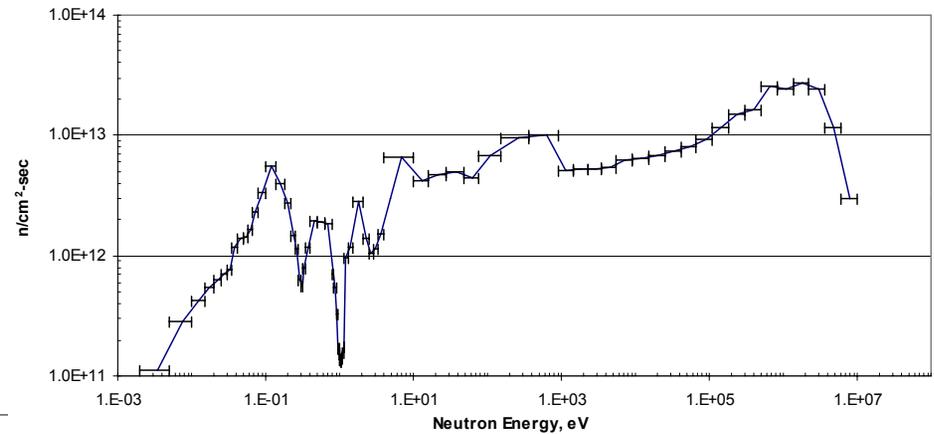
Production of C-14 by the N-14 (n,p) C-14 Reaction in a PWR

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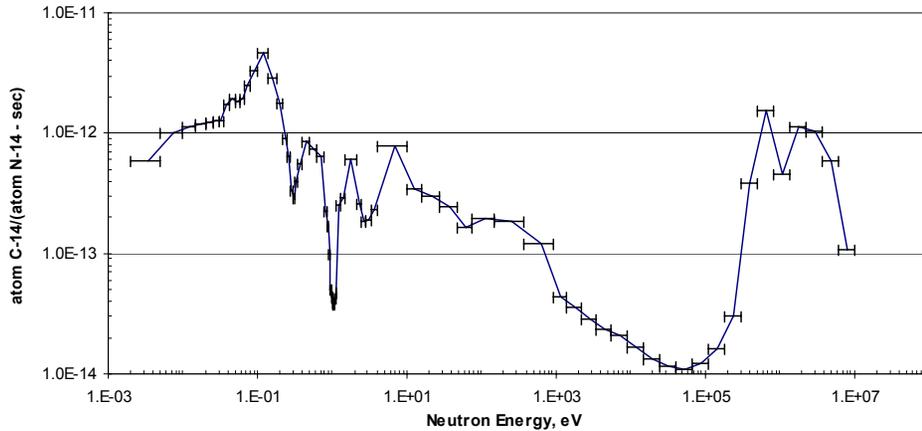
PWR Source Term Example - $^{14}N(n,p)^{14}C$ Reaction

PWR "Effective Cross-Sections" for the $^{14}N(n,p)^{14}C$ Reaction

Neutron Group	Group Energy	"Effective Cross-Section", b
Thermal	≤0.625 eV	0.9519
Intermediate	>0.625 eV - <1 MeV	0.0358
Fast	≥1 MeV	0.0505

- C-14 production rate per kilogram of water with 1.0 ppm dissolved nitrogen:

  2.10E-7 µCi/sec-kg-ppm N

- At a coolant mass in the core flux of 14,100 kg, the production rate for this reactor would be:

  2.10E-7*14,100 = 2.96E-3 µCi/sec-ppm N or 0.09 Ci/yr-ppm N

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PWR Source Term Example

- For the 1178 MW_e (~3549 MW_{TH}) PWR with an estimated coolant mass of 14,100 kg and 2 ppm N:

  - $^{17}O(n,\alpha)^{14}C$ Reaction = 11.0 Ci/yr
  - $^{14}N(n,p)^{14}C$ Reaction = 0.18 Ci/yr
  - Total $\qquad$ = <u>12.8 Ci/yr</u>

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BWR Source Term Example - $^{17}O(n,\alpha)^{14}C$ Reaction

"Effective Cross-Sections" for the $^{17}O(n,\alpha)^{14}C$ reaction

Neutron Group	Group Energy	"Effective Cross-Section", b	
		Moderator	Bypass
Thermal	≤0.625 eV	0.1328	0.1387
Intermediate	>0.625 eV - <1 MeV	0.0238	0.0222
Fast	≥1 MeV	0.1106	0.1106

- C-14 Production:
  - Moderator Region: 1.75E-5 µCi/sec-kg
  - Bypass Region: 2.05E-5 µCi/sec-kg

- For example, one large 3579 MW_{TH} BWR, is estimated to have 17,100 kg of coolant in the bypass region and 12,650 kg of coolant in the moderator region. The total production for this reactor would be:

$$1.75E-5*12,655 + 2.05E-5*17,100 = 0.572 \text{ µCi/sec}$$
$$= 18.0 \text{ Ci/yr}$$
$$= 21.3 \text{ kBq/MWhth}$$
$$= 0.575 \text{ µCi/MWhth}$$

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BWR Source Term Example - $^{14}N(n,p)^{14}C$ Reaction

"Effective Cross-Sections" for the $^{14}N(n,p)^{14}C$ reaction

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

• C-14 production per kilogram of water with 1.0 ppm dissolved nitrogen

Moderator Region	Bypass Region
2.15E-7 µCi/sec-kg-ppm N	3.23E-7 µCi/sec-kg-ppm N

• For example, one large 3579 MW_{th} BWR is estimated to have 17,100 kg of water in the bypass region and 12,655 kg of water in the moderator region.

• The total production for this reactor would be:

2.15E-7*12,655 + 3.23E-7*17,100 = 8.24E-3 µCi/sec-ppm N or 0.26 Ci/yr-ppm N

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BWR Source Term Example

- 3579 MW$_{TH}$ BWR, is estimated to have 17,100 kg of coolant in the bypass region and 12,650 kg of coolant in the moderator region:

  – $^{17}O(n,\alpha)^{14}C$ Reaction = 18.0 Ci/yr
  – $^{14}N(n,p)^{14}C$ Reaction = Negligible
  – Total $\qquad$ = <u>18.0 Ci/yr</u>

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What Do We Need To Do To Estimate the C-14 Source Term?

- Core "Average" Neutron Flux
  - BOC, Mid-cycle, EOC
  - Three energy groups
    - Thermal ≤ 0.625 eV
    - Intermediate >0.625 eV - <1.0 MeV
    - Fast ≥ 1.0 MeV
- Use "Effective Cross-Sections" in the three neutron energy groups

- "Effective Mass" of coolant in active core
  - Suggest use mass from "bottom" of active core to "top" of active core
  - BWR → must consider moderator and bypass regions

- Concentration of Nitrogen in the coolant (for PWR; BWR negligible.)

- Calculate BOC, mid-cycle and EOC C-14 source term
  - Average the three values

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Simplistic PWR Transport Model

Generation Rate

10 Ci/yr*

90% to 98%   < 1%   2% to 10%

Gaseous Release

9 to 9.8 Ci/yr total

Chemical Form, 5-30% CO_2
Remainder, Organic

0.5 – 2.9 Ci/yr CO_2

(without recombiner)

Liquid Release

< 0.1 Ci/yr

Solid Release

Filters and IX Resins

0.2 to 1 Ci/yr

10CFR61 Documentation for estimate.

Fraction organic, ^{14}C 30-90%

*Unit Specific

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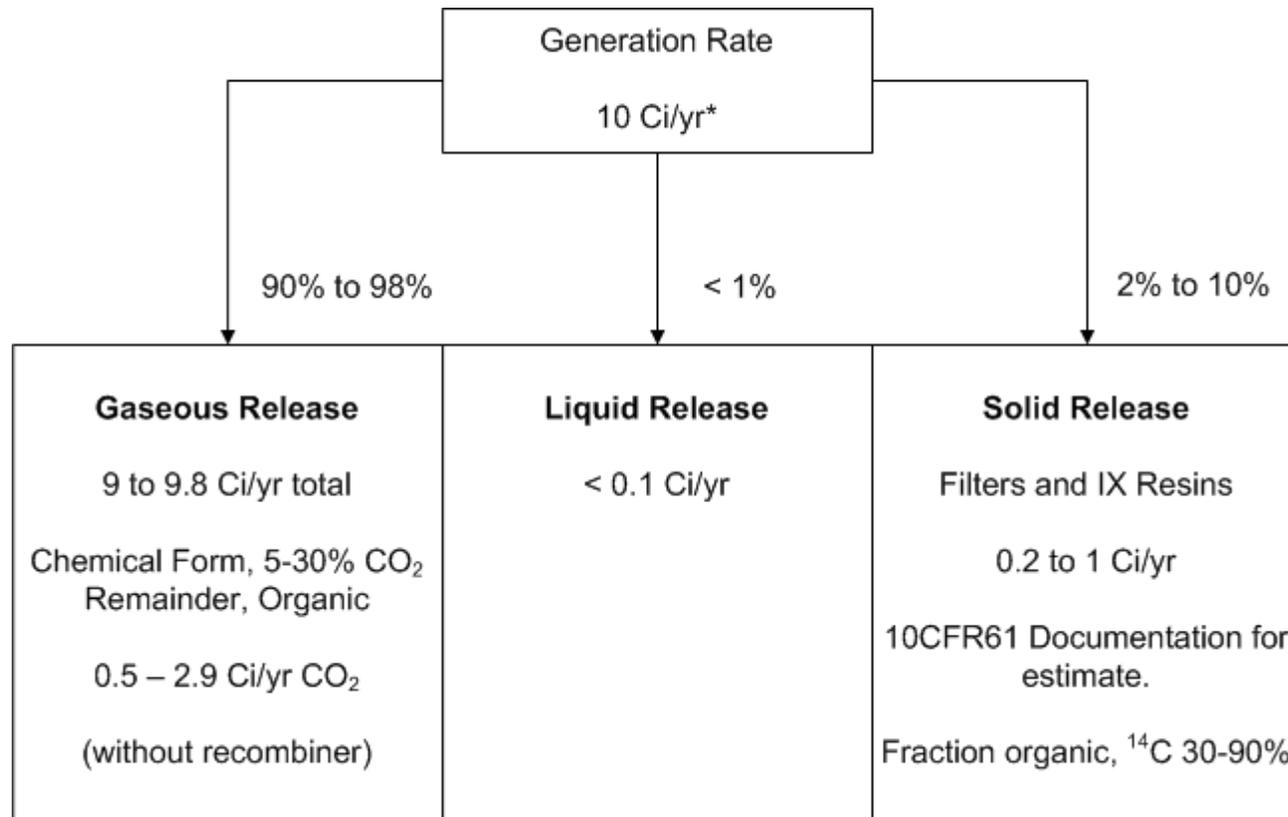
Simplistic BWR Transport Model
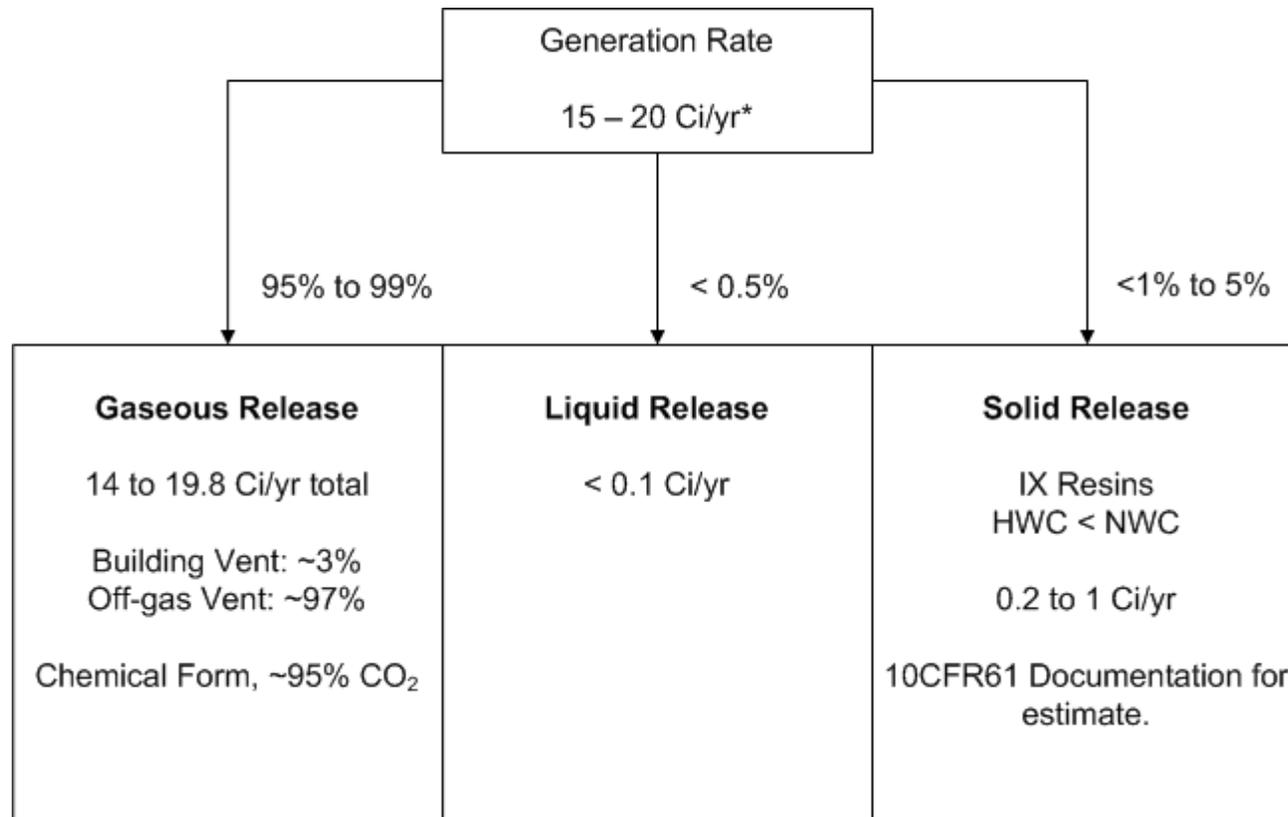
Current Status

- Calculation methodology for generation established.

- Collecting operational data from U.S. utilities to conduct example estimations and compare with available carbon-14 data.

- Draft methodology to be available in Fall 2010.

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