



**EPRI**

ELECTRIC POWER  
RESEARCH INSTITUTE

## 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.
- Collect information about experiences and technologies for carbon-14 sampling.

# Calculation of Reactor Coolant C-14 Source Term

- Production reactions
- Target concentrations
- Neutron cross-sections
- Neutron fluxes
- Target quantity in neutron flux

# Production of C-14 in Coolant

Nuclear Reactions (in order of importance)

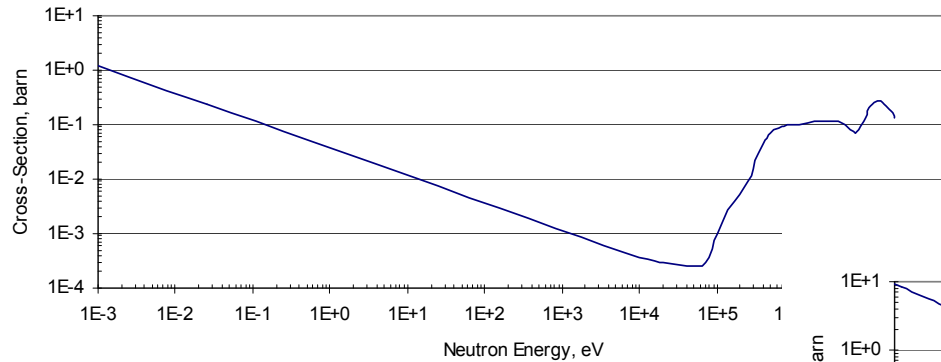
| Reaction                                 | Target Abundance (%) | Target Concentration                                        |
|------------------------------------------|----------------------|-------------------------------------------------------------|
| $^{17}\text{O} (n,\alpha) ^{14}\text{C}$ | 0.038                | 1.27E22 atoms $^{17}\text{O}/\text{kg H}_2\text{O}$         |
| $^{14}\text{N} (n,p) ^{14}\text{C}$      | 99.632               | 4.28E19 atoms $^{14}\text{N}/\text{kg H}_2\text{O}$ - ppm N |
| $^{13}\text{C} (n,\gamma) ^{14}\text{C}$ | 1.07                 | 5.36E17 atoms $^{13}\text{C}/\text{kg H}_2\text{O}$ - ppm C |

# Possible Chemical Forms Produced from In-Core $^{14}\text{C}$ Production Reactions

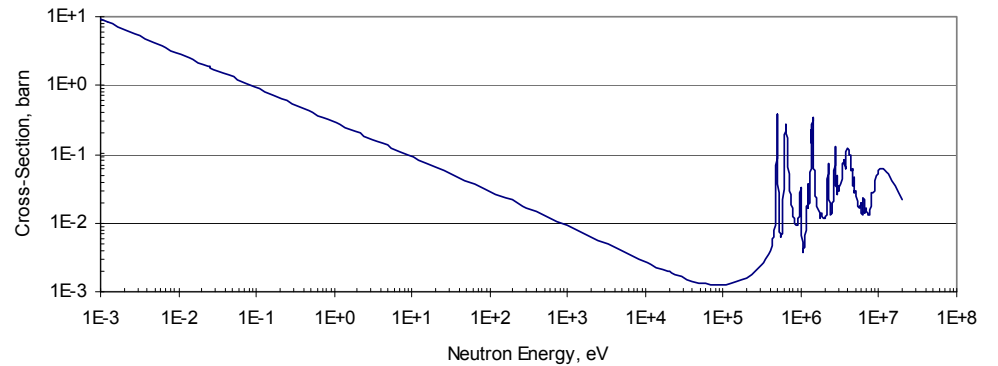
| Single Carbon Atom Species    |                 | Environment |
|-------------------------------|-----------------|-------------|
| $\text{CO}_2$                 | Carbon Dioxide  | Oxidizing   |
| $\text{CO}$                   | Carbon Monoxide | ↓           |
| $\text{HCOOH}$                | Formic Acid     |             |
| $\text{H}_2\text{C}=\text{O}$ | Formaldehyde    |             |
| $\text{CH}_3\text{OH}$        | Methanol        |             |
| $\text{CH}_4$                 | Methane         | Reducing    |

# C-14 Production Cross-Sections

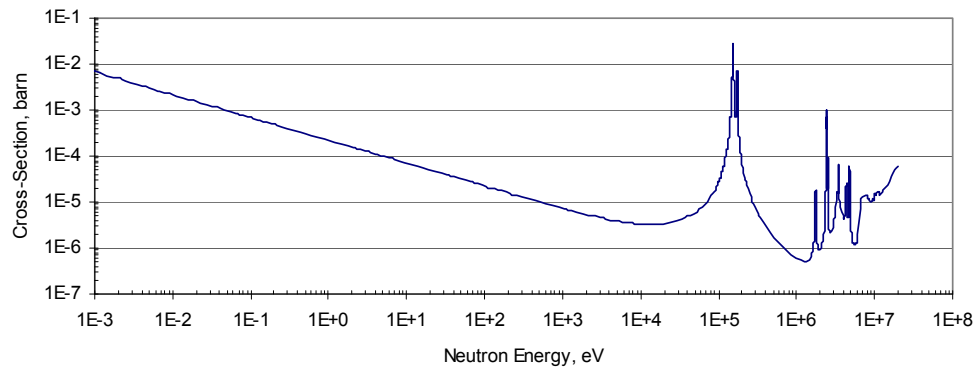
Cross-Section for  $^{17}\text{O}(n,\alpha)^{14}\text{C}$  Reaction



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

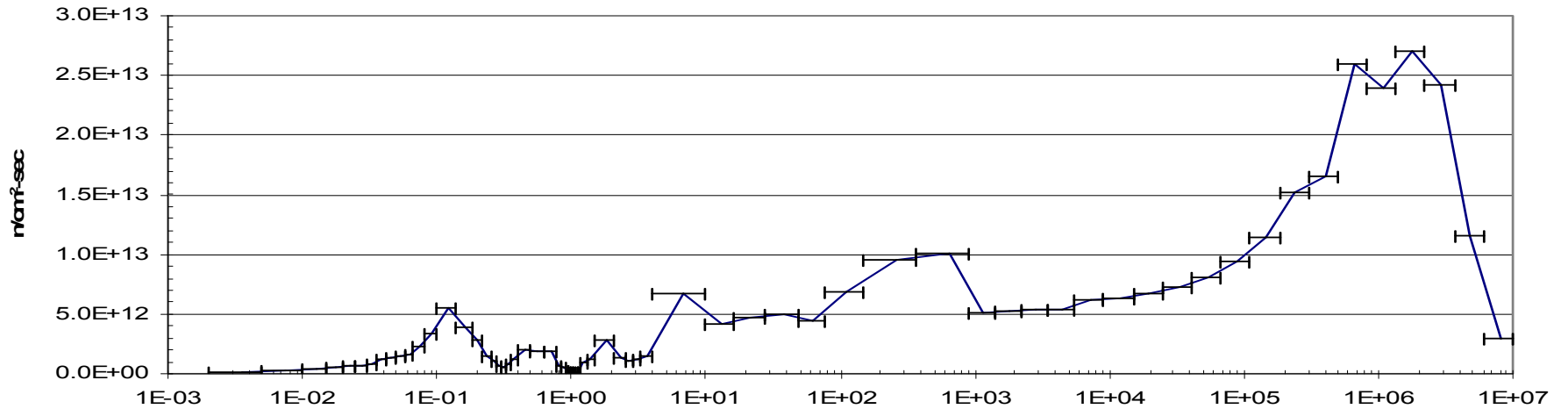


Cross-Section for  $^{13}\text{C}(n,\gamma)^{14}\text{C}$  Reaction

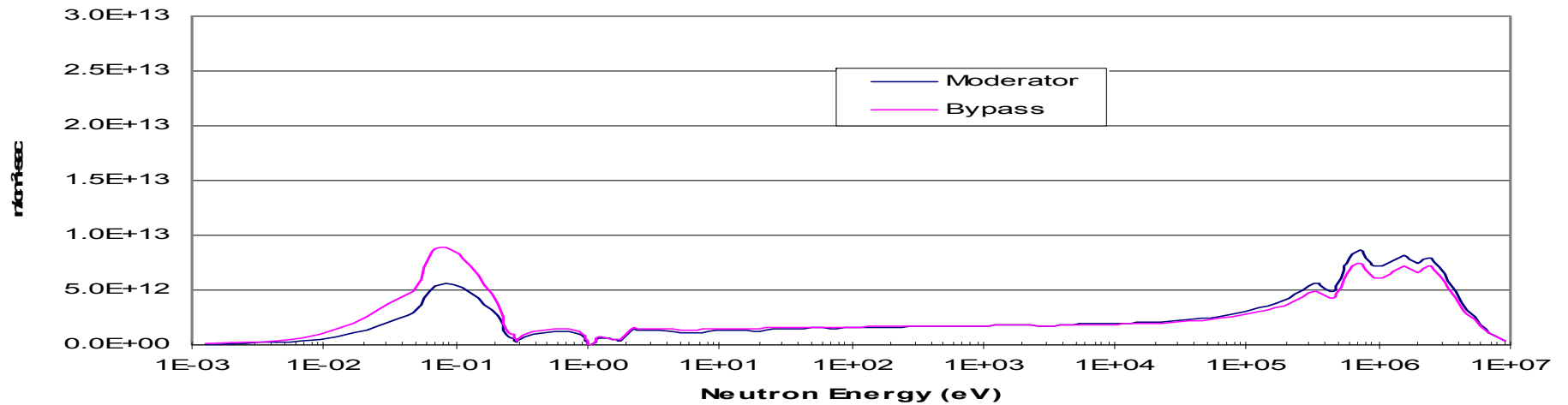


# Examples of Neutron Flux Spectra

**PWR Neutron Flux - 16 GWd/MTU**



**BWR Neutron Flux Distribution - 20 GWd/MTU**



# “Effective Mass” of Coolant in Active Core

|       |                                               |           |
|-------|-----------------------------------------------|-----------|
| • BWR | BWR-GALE Code                                 | 39,000 kg |
|       | Limerick UFSAR                                | 33,975 kg |
|       | GE Vessel Drawings                            |           |
|       | 2894 MW <sub>T</sub> , 624 Fuel Assemblies    | 24,630 kg |
|       | 3579 MW <sub>T</sub> , 748 Fuel Assemblies    | 29,755 kg |
|       | 3579 MW <sub>T</sub> , 732 Fuel Assemblies    | 27,805 kg |
|       | Oyster Creek, 1860 MWt, 560 Fuel Assemblies   | 26,254 kg |
|       | 1593 MW <sub>T</sub> , 368 Fuel Assemblies    | 15,830 kg |
|       | Bonka, 1972                                   | 33,000 kg |
|       | Fowler , 1976, 3579 MW <sub>T</sub> BWR/6     | 39,500 kg |
| • PWR | Helmholz (Braidwood 1)                        | 14,100 kg |
|       | Bonka (1976)                                  | 13,400 kg |
|       | Fowler (1976) - 3479 MW <sub>T</sub> CE Plant | 13,700 kg |

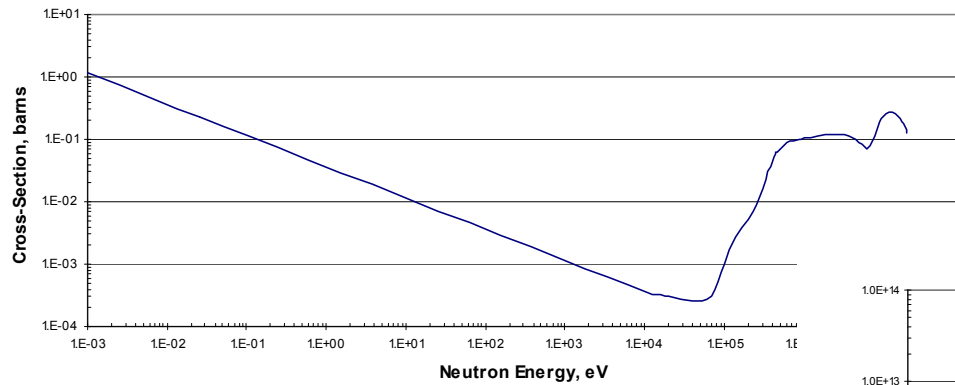
- Need site specific value to calculate individual unit source term.



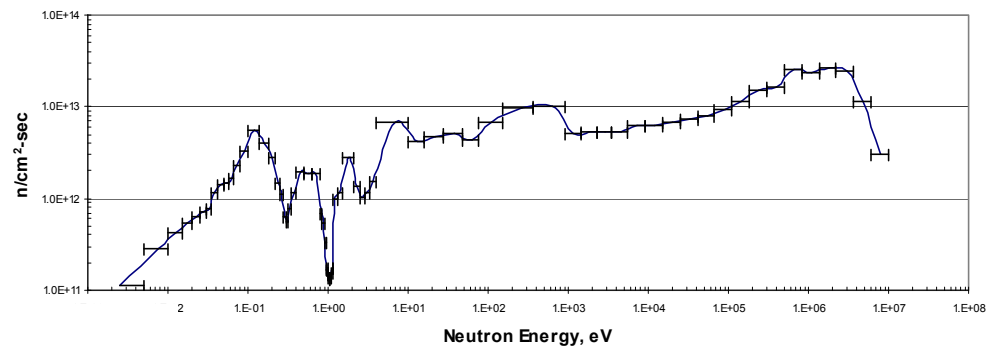
# Example Calculation at 16 GWd/MTU

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

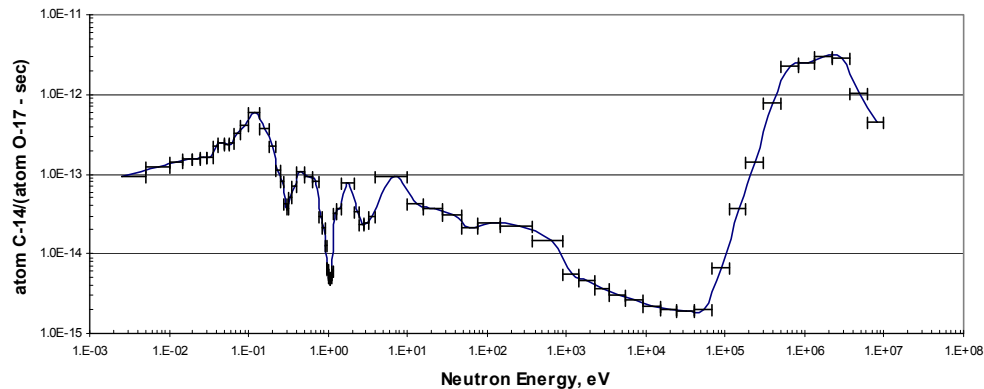
O-17 (n,alpha) C-14 Production Reaction



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



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



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

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

| Neutron Group | Group Energy             | “Effective Cross-Section”, b |
|---------------|--------------------------|------------------------------|
| Thermal       | $\leq 0.625$ eV          | 0.1189                       |
| Intermediate  | $> 0.625$ eV - $< 1$ MeV | 0.0183                       |
| Fast          | $\geq 1$ MeV             | 0.1504                       |

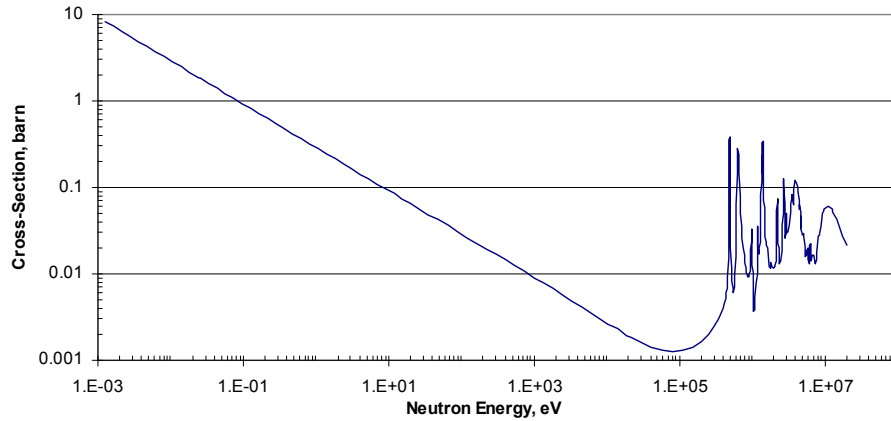
- C-14 production per kilogram of water is  $2.475\text{E-}5$   $\mu\text{Ci/sec-kg}$
- For the  $1178 \text{ MW}_e$  ( $\sim 3549 \text{ MW}_{\text{TH}}$ ) PWR with an estimated coolant mass of 14,100 kg, the total production for this reactor would be approximately:

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

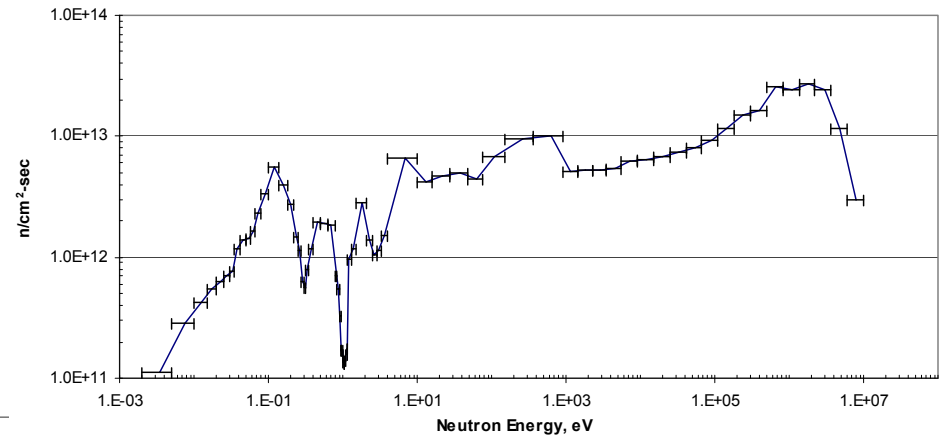
# Example Calculation at 16 GWd/MTU

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

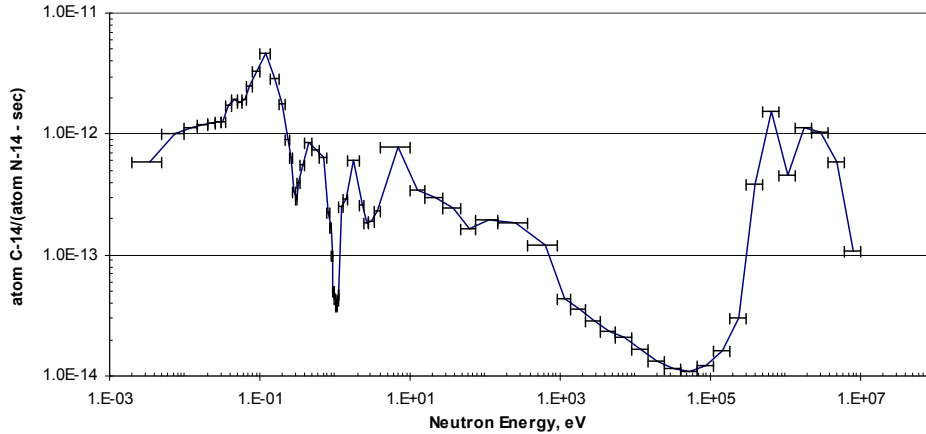
N-14 (n,p) C-14 Cross Section



PWR Neutron Flux



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



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

## PWR “Effective Cross-Sections” for the $^{14}\text{N}(n,p)^{14}\text{C}$ Reaction

| Neutron Group | Group Energy             | “Effective Cross-Section”, b |
|---------------|--------------------------|------------------------------|
| Thermal       | $\leq 0.625$ eV          | 0.9519                       |
| Intermediate  | $> 0.625$ eV - $< 1$ MeV | 0.0358                       |
| Fast          | $\geq 1$ MeV             | 0.0505                       |

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

$$2.10\text{E-}7 \text{ } \mu\text{Ci/sec-kg-ppm N}$$

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

$$2.10\text{E-}7 * 14,100 = 2.96\text{E-}3 \text{ } \mu\text{Ci/sec-ppm N or } 0.09 \text{ Ci/yr-ppm N}$$

## PWR Source Term Example

- For the 1178 MW<sub>e</sub> (~3549 MW<sub>TH</sub>) PWR with an estimated coolant mass of 14,100 kg and 2 ppm N:
  - $^{17}\text{O}(n,\alpha)^{14}\text{C}$  Reaction = 11.0 Ci/yr
  - $^{14}\text{N}(n,p)^{14}\text{C}$  Reaction = 0.18 Ci/yr
  - Total = 11.18 Ci/yr

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

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

|               |                          | “Effective Cross-Section”, b |        |
|---------------|--------------------------|------------------------------|--------|
| Neutron Group | Group Energy             | Moderator                    | Bypass |
| Thermal       | $\leq 0.625$ eV          | 0.1328                       | 0.1387 |
| Intermediate  | $> 0.625$ eV - $< 1$ MeV | 0.0238                       | 0.0222 |
| Fast          | $\geq 1$ MeV             | 0.1106                       | 0.1106 |

- C-14 Production:
 

|                                        |                                        |
|----------------------------------------|----------------------------------------|
| <u>Moderator Region</u>                | <u>Bypass Region</u>                   |
| $1.75\text{E-}5$ $\mu\text{Ci/sec-kg}$ | $2.05\text{E-}5$ $\mu\text{Ci/sec-kg}$ |
- For example, one large  $3579 \text{ MW}_{\text{TH}}$  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:

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

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

## “Effective Cross-Sections” for the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction

| Neutron Group | Group Energy             | “Effective Cross-Section”, b |        |
|---------------|--------------------------|------------------------------|--------|
|               |                          | Moderator                    | Bypass |
| Thermal       | $\leq 0.625$ eV          | 1.0560                       | 1.0903 |
| Intermediate  | $> 0.625$ eV - $< 1$ MeV | 0.0384                       | 0.0423 |
| Fast          | $\geq 1$ MeV             | 0.0479                       | 0.0478 |

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

### Moderator Region

2.15E-7  $\mu\text{Ci}/\text{sec}\cdot\text{kg}\cdot\text{ppm N}$

### Bypass Region

3.23E-7  $\mu\text{Ci}/\text{sec}\cdot\text{kg}\cdot\text{ppm N}$

- For example, one large 3579 MW<sub>th</sub> 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:

$$2.15\text{E-}7 \cdot 12,655 + 3.23\text{E-}7 \cdot 17,100 = 8.24\text{E-}3 \mu\text{Ci}/\text{sec}\cdot\text{ppm N} \text{ or } 0.26 \text{ Ci}/\text{yr}\cdot\text{ppm N}$$

- However, the concentration of N<sub>2</sub> in BWR coolant is very low and C-14 production from this reaction is considered to be negligible.

## BWR Source Term Example

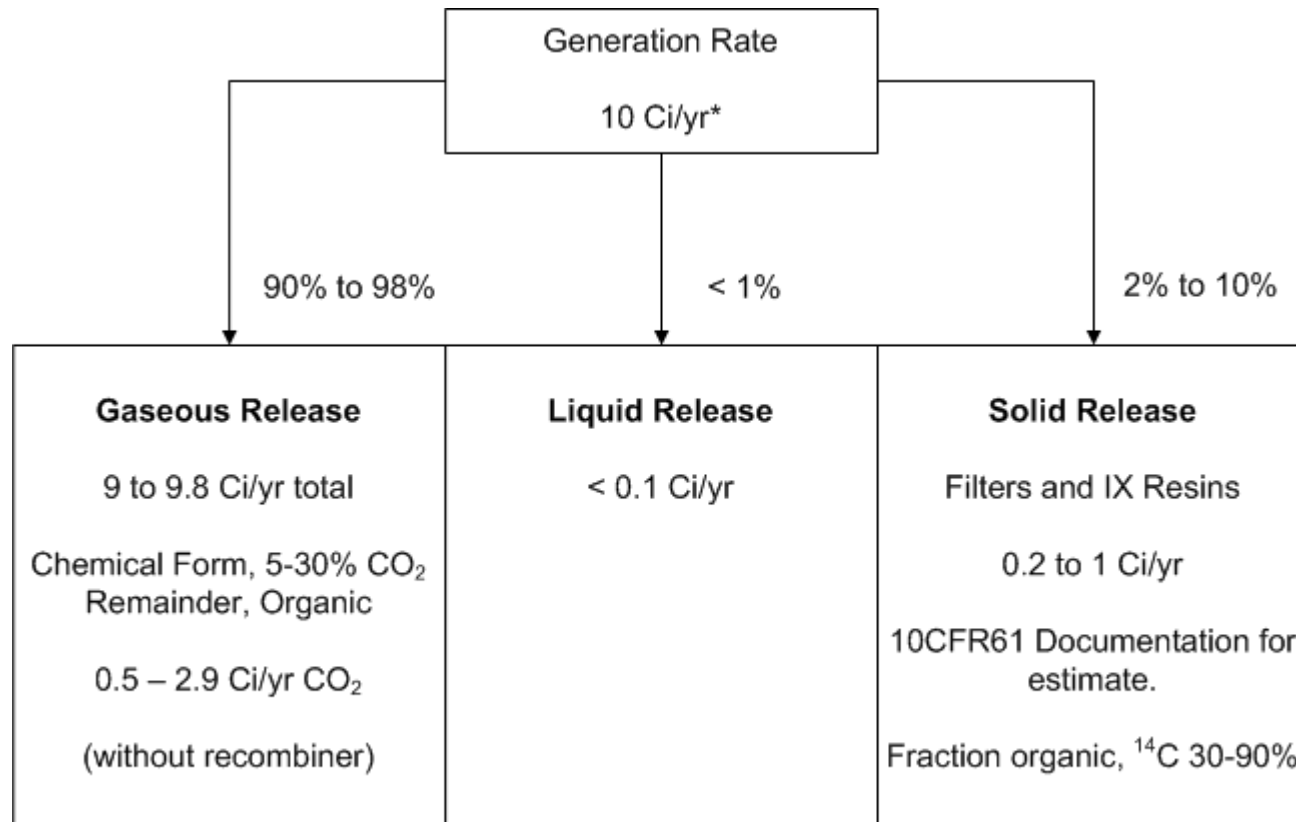
- 3579 MW<sub>TH</sub> BWR, is estimated to have 17,100 kg of coolant in the bypass region and 12,650 kg of coolant in the moderator region:
  - $^{17}\text{O}(n,\alpha)^{14}\text{C}$  Reaction = 18.0 Ci/yr
  - $^{14}\text{N}(n,p)^{14}\text{C}$  Reaction = Negligible
  - Total = 18.0 Ci/yr



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

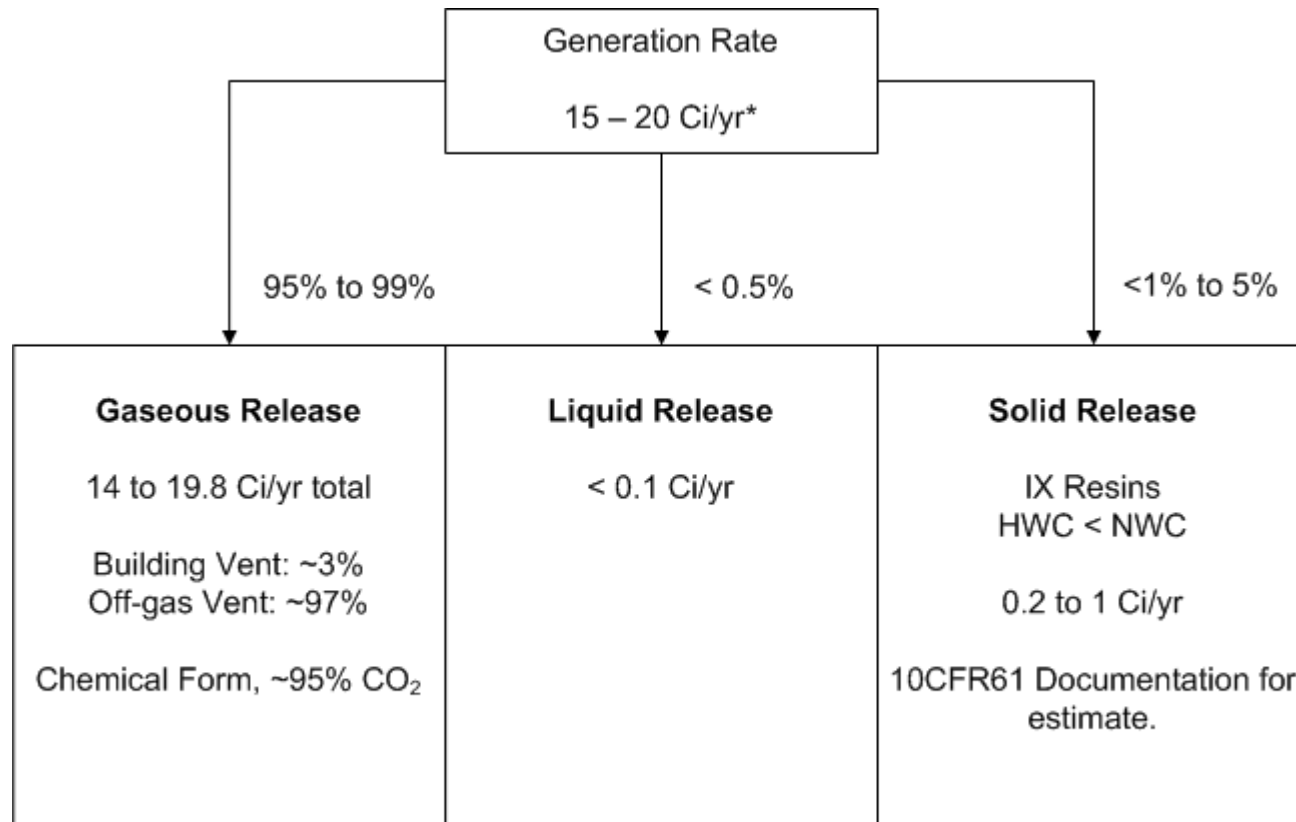
- Core “Average” Neutron Flux
  - BOC, Mid-cycle, EOC
  - Three energy groups
    - Thermal  $\leq 0.625$  eV
    - Intermediate  $> 0.625$  eV -  $< 1.0$  MeV
    - Fast  $\geq 1.0$  MeV
- Use “Effective Cross-Sections” in the three neutron energy groups
- “Effective Mass” of coolant in active core
  - 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

# Simplistic PWR Transport Model



\*Unit Specific

# Simplistic BWR Transport Model



\*Unit Specific

# 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.
  - 2-BWRs, 2-W PWRs, 1-CE PWR, 1-B&W PWR
- Draft methodology to be available in Fall 2010.



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