Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents

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NWT Corporation
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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)

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Target Abundance (%)</th>
<th>Target Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{17}$O (n,α) $^{14}$C</td>
<td>0.038</td>
<td>1.27E22 atoms $^{17}$O/kg H$_2$O</td>
</tr>
<tr>
<td>$^{14}$N (n,p) $^{14}$C</td>
<td>99.632</td>
<td>4.28E19 atoms $^{14}$N/kg H$_2$O - ppm N</td>
</tr>
<tr>
<td>$^{13}$C (n,γ) $^{14}$C</td>
<td>1.07</td>
<td>5.36E17 atoms $^{13}$C/kg H$_2$O - ppm C</td>
</tr>
</tbody>
</table>
Possible Chemical Forms Produced from In-Core $^{14}$C Production Reactions

<table>
<thead>
<tr>
<th>Single Carbon Atom Species</th>
<th>Environment</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO$_2$</td>
<td>Carbon Dioxide</td>
</tr>
<tr>
<td>CO</td>
<td>Carbon Monoxide</td>
</tr>
<tr>
<td>HCOOH</td>
<td>Formic Acid</td>
</tr>
<tr>
<td>H$_2$C=O</td>
<td>Formaldehyde</td>
</tr>
<tr>
<td>CH$_3$OH</td>
<td>Methanol</td>
</tr>
<tr>
<td>CH$_4$</td>
<td>Methane</td>
</tr>
</tbody>
</table>
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
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

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

<table>
<thead>
<tr>
<th>PWR</th>
<th>Mass (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Helmholz (Braidwood 1)</td>
<td>14,100</td>
</tr>
<tr>
<td>Bonka (1976)</td>
<td>13,400</td>
</tr>
<tr>
<td>Fowler (1976) - 3479 MW_T CE Plant</td>
<td>13,700</td>
</tr>
</tbody>
</table>

- 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 - $^{17}$O(n,α)$^{14}$C Reaction

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

<table>
<thead>
<tr>
<th>Neutron Group</th>
<th>Group Energy</th>
<th>“Effective Cross-Section”, b</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal</td>
<td>$\leq 0.625$ eV</td>
<td>0.1189</td>
</tr>
<tr>
<td>Intermediate</td>
<td>$&gt;0.625$ eV - &lt;1 MeV</td>
<td>0.0183</td>
</tr>
<tr>
<td>Fast</td>
<td>$\geq 1$ MeV</td>
<td>0.1504</td>
</tr>
</tbody>
</table>

- C-14 production per kilogram of water is 2.475E-5 µCi/sec-kg
- For the 1178 MWₑ (~3549 MWₜₜ) PWR with an estimated coolant mass of 14,100 kg, the total production for this reactor would be approximately:

$$2.475\times10^{-5} \times 14,100 = 0.349 \mu\text{Ci/sec}$$
$$= 11.0 \text{ Ci/yr} = 9.35 \text{ Ci/GWₑ-yr}$$
$$= 13.1 \text{ kBq/MWhₜₜ} = 0.354 \mu\text{Ci/MWhₜₜ}$$
Example Calculation at 16 GWd/MTU
$^{14}\text{N}(n,p)^{14}\text{C}$ Reaction

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

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

PWR Neutron Flux
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

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<thead>
<tr>
<th>Neutron Group</th>
<th>Group Energy</th>
<th>“Effective Cross-Section”, b</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal</td>
<td>≤0.625 eV</td>
<td>0.9519</td>
</tr>
<tr>
<td>Intermediate</td>
<td>&gt;0.625 eV - &lt;1 MeV</td>
<td>0.0358</td>
</tr>
<tr>
<td>Fast</td>
<td>≥1 MeV</td>
<td>0.0505</td>
</tr>
</tbody>
</table>

- C-14 production rate per kilogram of water with 1.0 ppm dissolved nitrogen:
  
  $2.10\text{E-7}\ \mu\text{Ci/sec-kg-ppm N}$

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

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

• For the 1178 MWₑ (~3549 MWₜʰ) PWR with an estimated coolant mass of 14,100 kg and 2 ppm N:
  
  – \( ^{17}\text{O}(\text{n},\alpha)^{14}\text{C} \) Reaction = 11.0 Ci/yr
  – \( ^{14}\text{N}(\text{n},p)^{14}\text{C} \) 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

<table>
<thead>
<tr>
<th>Neutron Group</th>
<th>Group Energy</th>
<th>Moderator</th>
<th>Bypass</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal</td>
<td>$\leq 0.625$ eV</td>
<td>0.1328</td>
<td>0.1387</td>
</tr>
<tr>
<td>Intermediate</td>
<td>$&gt;0.625$ eV - $&lt;1$ MeV</td>
<td>0.0238</td>
<td>0.0222</td>
</tr>
<tr>
<td>Fast</td>
<td>$\geq 1$ MeV</td>
<td>0.1106</td>
<td>0.1106</td>
</tr>
</tbody>
</table>

- C-14 Production: $^{17}\text{O}(n,\alpha)^{14}\text{C}$
  
  **Moderator Region**
  
  $1.75\times 10^{-5}$ µCi/sec-kg

  **Bypass Region**
  
  $2.05\times 10^{-5}$ 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:

  
  
  $1.75\times 10^{-5} \times 12,655 + 2.05\times 10^{-5} \times 17,100 = 0.572$ µCi/sec

  
  
  $= 18.0$ Ci/yr

  
  
  $= 21.3$ kBq/MWhth

  
  
  $= 0.575$ µCi/MWhth
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

<table>
<thead>
<tr>
<th>Neutron Group</th>
<th>Group Energy</th>
<th>&quot;Effective Cross-Section&quot;, b</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal</td>
<td>$\leq 0.625$ eV</td>
<td>1.0560</td>
</tr>
<tr>
<td>Intermediate</td>
<td>$&gt;0.625$ eV - $&lt;1$ MeV</td>
<td>0.0384</td>
</tr>
<tr>
<td>Fast</td>
<td>$\geq 1$ MeV</td>
<td>0.0479</td>
</tr>
</tbody>
</table>

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

<table>
<thead>
<tr>
<th>Moderator Region</th>
<th>Bypass Region</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.15E-7 $\mu$Ci/sec-kg-ppm N</td>
<td>3.23E-7 $\mu$Ci/sec-kg-ppm N</td>
</tr>
</tbody>
</table>

- 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:

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

- However, the concentration of N$_2$ in BWR coolant is very low and C-14 production from this reaction is considered to be negligible.
BWR Source Term Example

• 3579 MW\textsubscript{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\textsuperscript{O}(n,\alpha)^{14}\textsuperscript{C} Reaction = 18.0 Ci/yr
  – 14\textsuperscript{N}(n,p)^{14}\textsuperscript{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 \( \leq 0.625 \text{ eV} \)
    • Intermediate \( >0.625 \text{ eV} - <1.0 \text{ MeV} \)
    • Fast \( \geq 1.0 \text{ MeV} \)
  • 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 \( \rightarrow \) 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

**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% CO₂

- Remainder, Organic

- 0.5 – 2.9 Ci/yr CO₂

  (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
Simplistic BWR Transport Model

Generation Rate
15 – 20 Ci/yr*

95% to 99%

< 0.5%

<1% to 5%

Gaseous Release
14 to 19.8 Ci/yr total
Building Vent: ~3%
Off-gas Vent: ~97%
Chemical Form, ~95% CO₂

Liquid Release
< 0.1 Ci/yr

Solid Release
IX Resins
HWC < NWC
0.2 to 1 Ci/yr
10CFR61 Documentation for estimate.

*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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