Fission product behaviour in the containment

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Outline of the presentation

1. Introduction
   • Motivation
   • Source term to the containment
   • Iodine chemistry in the containment

2. ASTEC analysis example on fission product behaviour in the Nordic BWR containment

3. Summary
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Motivation:

- Fission product behaviour inside the containment has important role in defining the source term to the environment
- In severe accident scenarios with controlled pressure relief through Filtered Containment Venting (FCV) line, typically filters are able to retain:
  - Almost all aerosol particles
  - Only a small part of gaseous iodine
  - Negligible part of organic iodides and even less noble gases

→ It is essential to be able to define the form of fission products
  → All relevant phenomena should be taken into account
  → Iodine chemistry in the containment considered extremely complex
Gaseous fission products that are formed in the fuel will diffuse to gas gap between the fuel pellet and cladding (~ 5 % of the inventory)

- Diffusion rate increases when the fuel temperature increases
- Different fission products have different diffusion rates

Gas gap inventory will be released instantaneously when the cladding fails

Practically all noble gases (Xe, Kr), caesium (Cs) and iodine (I) will be released during the in-vessel phase

- Releasing Cs will react promptly with I forming CsI
  - In Phébus experiments with silver-indium-cadmium control rods CdI₂ was more common form
- Typically more Cs is released than I
  - Traditionally it has been assumed that excess Cs will form CsOH
  - Phébus experiments did show that Cs will more probably react with molybdenum forming Cs₂MoO₄

- CsI, CdI₂, CsOH and Cs₂MoO₄ will agglomerate as aerosol particles
Source term to the containment: Transportation of fission products

- Fission product aerosols
  - Transportation to primary circuit with steam or gas flows
  - Deposition of particles onto primary circuit structures
    - **Sedimentation**: suspended particles settle out from the fluid by gravitation
    - **Impaction**: suspended particles settle out from the fluid by momentum
    - **Diffusiophoresis**: motion of dispersed particles in a fluid induced by a diffusion gradient
    - **Thermophoresis**: motion of dispersed particles in a fluid induced by a temperature gradient
  - May re-vaporise if the structures become heated
- Noble gases remain in gas phase as well as part of iodine (5 %)
Principles of iodine behaviour:
Liquid phase chemistry

- The most of the metal iodides are soluble and will be dissolved to sump water → iodide ions → I₂ due radiative and thermal effects
  - The formation of I₂ depends extensively on pH level of the sump
  - If pH > 7 the formation of I₂ is considered to be very low
    - NUREG-1465: less than 1 % of the dissolved iodine will be converted to elemental iodine
- Other source for volatile iodine in the sump is the formation of organic iodine compounds
  - Organic radicals in the sump water originate for example from paints
- Silver will react with dissolved iodine species to form for example insoluble silver iodide (AgI)
  - This will limit the formation volatile I₂ regardless of the sump pH
Principles of iodine behaviour: Gas phase chemistry

- Volatile iodine will be absorbed and desorbed from surfaces in the containment
  - A painted surface is a sink for $I_2$ and a source for volatile organic iodine
    - Radiation induces fast radiochemical reactions between iodine and the paints
      - This phenomenon was observed in the Phébus-FP experiments by a formation of small steady-state concentration of iodine not dependent from pool pH
  - Also the air radiolysis products react with $I_2$ to form primarily non-volatile species
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ASTEC (Accident Source Term Evaluation Code)

- **ASTEC** is an integral code for simulating the entire severe accident sequence from the initiating event to the release of radioactive elements out of the containment.
- Jointly developed by IRSN and GRS since 1996.
- Progressive and continuous evolution towards a **state-of-the-art tool for source term calculations**.
ASTEC input

- **BREAK** = water, steam, H₂
- **NOBLES** = Xe, Kr
- **HALOGENS** = I, Br
- **ALKALI** = Cs, Rb

Leak size dependent on $P_{\text{drywell}}$

Venting begins when $P_{\text{drywell}} = 5$ bar

Will open if $P_{\text{wetwell}} > P_{\text{drywell}}$ more than 0.01 bar, size dependent on $dP$

Begins at 1800 s from scram
Source term to the containment based on NUREG-1465 report

<table>
<thead>
<tr>
<th>Category</th>
<th>Substance</th>
<th>Total mass [kg]</th>
<th>Gap release</th>
<th>Early In-vessel release</th>
</tr>
</thead>
<tbody>
<tr>
<td>1) Noble gases</td>
<td>Xe</td>
<td>0.753</td>
<td>5 %</td>
<td>95 %</td>
</tr>
<tr>
<td></td>
<td>Kr</td>
<td>2.266</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2) Halogens</td>
<td>I</td>
<td>14.656</td>
<td>5 %</td>
<td>25 %</td>
</tr>
<tr>
<td></td>
<td>Br</td>
<td>0.001</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3) Alkali metals</td>
<td>Cs</td>
<td>159.211</td>
<td>5 %</td>
<td>20 %</td>
</tr>
<tr>
<td></td>
<td>Rb</td>
<td>0.001</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Timing of the release: 20 min – 50 min, 50 min – 2 h 20 min
Pressure in the containment

- Pressure increases rapidly in the containment after the pipe break and stabilizes
  - Controlled with blowdown pipes
- Increase again when the fission product release to the containment begins at 20 min
  - Around the same time hydrogen production starts
- The containment rupture disk venting begins at around 3 h 48 min decreasing the pressure effectively
- The peak at 4 h 4 min is caused by a steam burst from the primary circuit caused by a small amount of debris slumping to the lower plenum
Temperature in the containment

- Decaying fission products heat the containment atmosphere
- When the fission product input ends, the decay heat produced in the containment is 2.38 MW
- The total decay heat should be around 25 MW at that time
  - $P_{th} = 2\,500\,\text{MW}$
- Nearly 10% of the total decay heat is produced in the containment after 2 h 20 min
Produced dose rates

- Gas phase dose rate maximum occurs before all fission products enter to the containment (short lived nuclides)
- Dose rates on walls higher than dose rates on gas phase due to the amount of deposited aerosols
- For cavity the change is more notable
  - The temperature is lower and therefore there is assumed to be less gaseous fission products compared to aerosols
  - More deposited aerosols per wall area
- CsOH present also in gaseous form during temperature peaks in drywell
- To the wetwell remains a nearly constant inventory of noble gases
- Practically no pressure difference between the compartments
- Gaseous FPs from drywell and cavity are directed to the venturi scrubber
  - Cavity-drywell flow path: 1.9 m²
  - Drywell-wetwell leak: 0.01 m²
Aerosols in the atmosphere

On average, more fission products are present as aerosols than in gaseous form.
Deposited aerosols

- Deposition mostly occurred before containment venting begun (3h 48min)
- 49 % deposited by thermophoresis, 11 % by settling and 40 % by diffusiophoresis
- In the wetwell there is no deposition of aerosols
  - Assumed to be a result of relatively low temperature and condensation of water on walls
Fission products in sump

- In total 97.2 % of the fission products were retained in the containment
- Most of them were dissolved to water (at 6h):
  - 85 % of Cs
  - 86 % of I
  - 93 % of Kr and Xe
Iodine on paint

- Mass of organic iodides remained very small in the analysis.
- The maximum value of CH$_3$I registered was in the drywell gaseous phase: 0.6 g
  - Wetwell pool walls were assumed to have steel surface
- Organic iodine is destroyed by the gamma radiation which could explain partly the relatively small amount of CH$_3$I
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When considering the possible source term to the environment, gaseous fission products are challenging.

In the analysed scenario:

- More fission products were present as aerosols than in gaseous form
  - In addition to noble gases only CsOH present in gaseous form during temperature peaks
- 97.2 % of the fission products were retained in the containment
  - Most of deposition occurred already before controlled containment pressure relief
  - The mass of noble gases in water surprisingly high
- Mass of organic iodides remained very small
  - Destroyed by the γ-radiation
  - Painted wall area defined too small
- Mass of gaseous iodine remained also very small despite water pH was 5
  - Relatively small temperatures
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