Research on the Transport and Chemistry of Fission Products in Primary Circuit and Containment Conditions at VTT

Nuclear 2012, Pitesti, Romania, 17.5.2012
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Introduction

- During a severe accident in a nuclear power plant (NPP), in which the normal core cooling is lost and the fuel rods are melting, the release of fission products (FPs) from fuel will take place.

- In that event it is possible, that FPs are transported from the damaged fuel rods into the primary circuit and further to the containment building.
Introduction

- The released fission products, such as ruthenium and iodine, are radiotoxic and they may form gaseous compounds.

- Therefore, it is important to know in which form (gas, aerosol) the released FPs are transported in severe accident conditions.
Background

- The release and transport of fission products have internationally been one of the main interests in a field of nuclear safety research.

- The objectives of the studies have been to gather information on the behaviour of FPs and utilize it in the validation of severe accident simulation codes.
Background

- Phebus-FP has been one of the largest experimental programs where the release of FPs from the real fuel rods, with various burn-ups, has been studied [2].

- These large scale experiments have resulted in a wide database.


Background

- However, there are still open questions e.g. on the behaviour of iodine.

- In the Phebus-FP experiments, the gas phase concentration of iodine was found to be on a stable level in a long term, regardless of the experimental conditions and the water chemistry in a sump.

- The chemical processes for the observed generation and depletion of gaseous iodine in the containment are not known.
Background

- While the processes leading to this are not known the following hypothesis has been formulated*:
  - Gaseous iodine and particulate iodine swept to painted condensers by steam condensation
  - Iodide ion (or some other soluble chemical form of iodine) rapidly absorbs from water film onto paint before water film can drain and be discharged to sump
  - Iodine on paint desorbs as a volatile species whether a water film is present or not
  - Gaseous iodine species radiolytically destroyed to form fine particulate iodine oxides or iodine nitrogen oxides
  - Iodine particulate sediments from containment atmosphere

*R.Y. Lee and M. Salay, Phébus-FP Findings on Iodine Behaviour in Design Basis and Severe Accidents, Presented to the Advisory Committee on Reactor Safeguards 8.5.2008, U.S.NRC
Background

- In addition to large-scale facilities, there are several intermediate and small scale facilities which are dedicated on studying the various phenomena taking place in the primary circuit and the containment building during a severe accident.

- Studies at VTT have focused on the transport and chemistry of FPs at described conditions. In recent years the interest in experiments has been on ruthenium and iodine, because of their high radiotoxicity.
Experiments at VTT

- Ruthenium transport in primary circuit
- Iodine transport in primary circuit
- Radiolytical oxidation of iodine in containment conditions
- Desorption of iodine from IOx deposition on painted surface
Ruthenium transport in primary circuit

- In experiments the formation and transport of volatile ruthenium oxides was studied by exposing RuO$_2$ powder to diverse oxidising atmospheres at a relatively high temperature.

- Varied parameters:
  - Temperature 1100-1700 K
  - Oxygen volume fraction in flow 6-21 %
  - Steam volume fraction in flow 0-50 %
  - Total flow rate 5 – 10 l/min
Ruthenium transport in primary circuit

Tubular reactor
I.D. 22 mm
Length 110 cm
Heated length 2 x 40 cm

CO - critical orifice
MFC - mass flow controller
ESP - electrostatic precipitator
DMA - differential mobility analyzer
CNC - condensation nucleus counter

CO - vapour
H2O - water
Argon
Air
1 M NaOH

filter 1
1 M NaOH

filter 2
CNC
0.3 lpm
Ruthenium transport in primary circuit

![3D chart showing Ruthenium transport in primary circuit with release rate, oxygen volume fraction, and steam volume fraction at different temperatures: 1100 K, 1300 K, 1500 K, 1700 K.](chart.png)
Ruthenium transport in primary circuit

RuO₂ transport rate [mg/min]
Ruthenium transport in primary circuit

$\text{RuO}_4$ transport rate [mg/min]
Ruthenium transport in primary circuit

Ru deposition profile in the facility

- Exp 6 1500K Air saturated
- Exp 7 1300K 10% steam
- Exp 8 1300K 50% steam
- Exp 9 1300K Dry air
- Exp 16 1500K Air sat + RuO4
- Exp 17 1500K Air sat + RuO4
Ruthenium transport in primary circuit

- It was noticed that a very large fraction of ruthenium may be transported in gaseous form.

- At 1300 K ruthenium was primarily transported in gaseous form (as high as 89% of transported Ru) and increasing steam partial pressure increased the fraction of gaseous ruthenium.

- At 1500 K and above, ruthenium transport took place almost entirely as aerosol particles (~99% of transported Ru).

- Gaseous ruthenium seemed to react on the surface of the stainless steel tube, on top of which RuO$_2$ particles had been deposited.
Iodine transport in primary circuit

- Reactions of deposited fission products on primary circuit surfaces can modify the amount, composition and timing of fission product release to the containment.

- Studies to investigate the effects of reactions with primary circuit surfaces on transport of iodine have been conducted. The effects of different precursor mixtures on iodine transport have been investigated with EXSI PC facility
Iodine transport in primary circuit

- Caesium iodide (CsI) was used as the source of iodine in all experiments. Silver, molybdenum oxide and boron oxide were used as additives.

- Reaction furnace temperature was varied between 400 and 650 °C in order to determine the effect of temperature in the release of gaseous and aerosol phase iodine.

- Each experiment contained three different atmosphere conditions, with different fractions of steam, argon and hydrogen
Addition of silver to the CsI precursor at 650 °C decreased the release of iodine as well as the fraction of gaseous iodine.
At 400 °C, Ag + CsI as well as Ag + MoO$_3$ + CsI precursor significantly increased the release of gaseous iodine, where almost no aerosol particles were released.
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Iodine transport in primary circuit

- Generally in the experiments, it was observed that the hydrogen in the atmosphere decreased the fraction of released gaseous iodine.

- As the temperature was lowered, less iodine was released, but the fraction of gaseous iodine from the overall released iodine was increased.
Radiolytical oxidation of iodine in containment conditions

- During a severe accident, iodine may react with painted surfaces of containment to form organic iodine species.

- These organic species are a possible source of volatile iodine, which may increase the fraction of iodine in the gas phase.

- Therefore, it is important to study the transport of organic iodine in containment conditions.

- Another question is, whether the organic iodides are transported as gaseous molecules or as aerosol particles resulting from reactions with air radiolysis products.
Radiolytical oxidation of iodine in containment conditions

- In experiments methyl iodide was fed into the EXSI CONT facility in an air mixture. In some experiments the flow contained also humidity.

- The reactions took place in a quartz tube heated either to 50°C, 90°C or 120°C.

- UV-light was used as a source of radiation to produce ozone from oxygen. A separate generator was also applied to reach higher ozone concentrations.
Radiolytical oxidation of iodine in containment conditions
Radiolytical oxidation of iodine

- Irradiation of oxygen molecules in air leads to the subsequent formation of ozone, which is keen to react with other substances.

- Both gaseous elemental and organic iodine tends to react with air radiolysis product ozone and form iodine oxide aerosol particles.

- Air radiolysis:

  \[ \text{UV (185nm)} + \text{O}_2 \rightarrow \text{O} + \text{O} \]
  \[ \text{O} + \text{O}_2 \rightarrow \text{O}_3 \]
  \[ \text{I}_2 + \text{O}_3 \rightarrow \ldots \text{final products: e.g. I}_2\text{O}_5, \text{I}_4\text{O}_9 \text{ aerosols} \]
Radiolytical oxidation of iodine
Radiolytical oxidation of iodine
Radiolytical oxidation of iodine

![Graph showing the radiolytical oxidation of iodine]

- **CH₃I**
- **CH₃OH**
- **HCHO**

100% radiation intensity - 50% radiation intensity
Desorption of iodine from IOx deposition on painted surface
The effect of radiation dose on the desorption of iodine from IOx on paint

The fraction of iodine desorbed [%]
Gamma radiation dose [kGy]

Humid flow
Conclusions

- A significant fraction of ruthenium and iodine may transport as gas to the containment atmosphere.

- The radiolytical oxidation of iodine in the air atmosphere produces IOx particles.

- Gaseous iodine releases from the IOx particles deposited on the painted surface of containment.

- This work was funded by the Finnish Research Programme on Nuclear Power Plant Safety (SAFIR2010 and SAFIR2014), Nordic Nuclear Safety Research (NKS-R), European Commission / EU SARNET2 programmes.