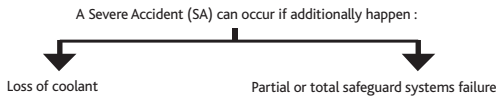




Main achievements of the European R&D on the issue of air ingress during a LWR severe accident

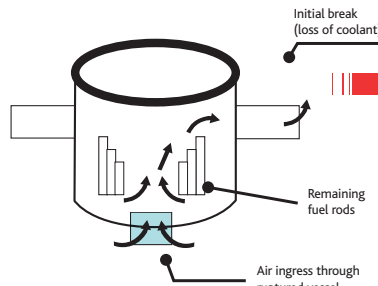
(C. Mun, T. Kärkelä, N. Vër, D. Corbin, G. Brillant, M. Steinbrück)

Context



Consequence: core fusion and possible core vessel lower head failure.

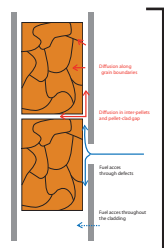
1. Air can then penetrate in the reactor vessel evidenced by simulations with calculation codes (ASTEC, MAAP-SATURNE).
2. Remaining zirconium cladding is oxidised by air and may undergo large degradation.
3. Remaining fuel is also oxidised as well as fission products (FP's) which form volatile species.



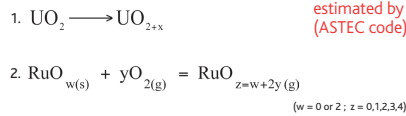
Potential high release of volatile ruthenium oxides from the reactor vessel to the reactor containment, and possibly to the outside environment.

- Important part of ruthenium produced in a LWR core (15.8 % of the ²³⁵U fission reactions).
- High radiotoxicity of ruthenium through ¹⁰³Ru and ¹⁰⁶Ru.

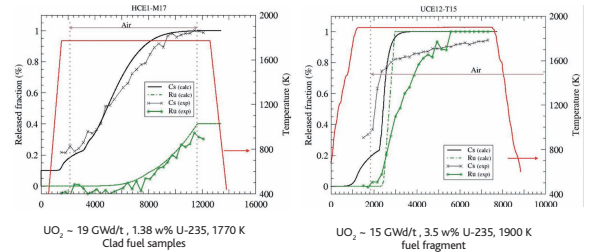
Ruthenium release from fuel



Ruthenium release kinetics depends on both fuel oxidation kinetics and ruthenium oxidation kinetics.

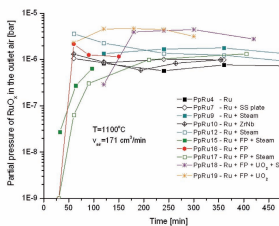


Under air atmosphere, high ruthenium oxides releases were experimentally observed, and now are well estimated by calculations (ASTEC code).



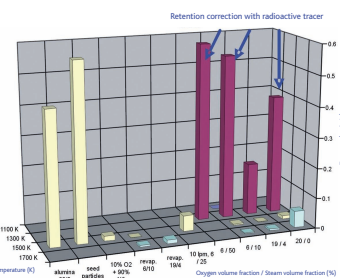
Ruthenium transport in the RCS

All $RuO_4(g)$ inventory is not trapped by the RCS, leading to the presence of a significant fraction of gaseous tetroxide in the containment.



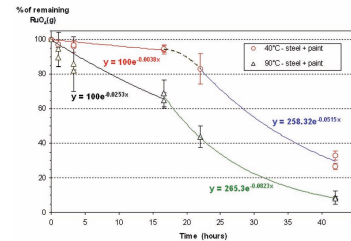
- Partial pressure of RuO_4 in the outlet air was in the range of 10^{-6} bar, far above the value that would be expected from thermodynamic equilibrium (10^{-10} bar).
- Presence of steam, other fission products and UO_2 influenced the concentration of RuO_4 in the ambient temperature escaping gas, and caused a time delay in appearance of its maximum values.

Gaseous RuO_4 transport rate through temperature gradient tube and atmosphere composition.



Ruthenium behaviour in the reactor containment

Due to radiolytic reactions, $RuO_4(g)$ may be present in the containment atmosphere for several days.



$$-\frac{d[RuO_4]_{(g)}}{dt} = (k_d + k_H) [RuO_4]_{(g)}$$

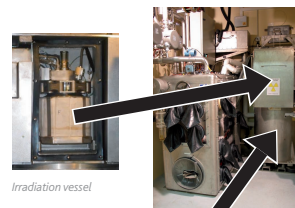
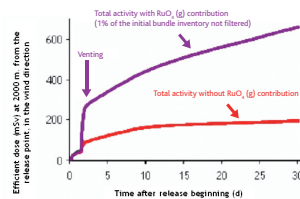
- $RuO_4(g)$ is not as unstable as indicated by the literature.
- Half-life time was experimentally evaluated to around 5 hours in representative conditions of a SA (temperature, % steam).
- $RuO_4(g)$ decomposition process into RuO_2 deposits is catalysed by steam and ruthenium dioxide deposits.

$$\frac{d[RuO_4]_{(g)}}{dt} = (k_{O_3} + k_{H_2O_2}) X(H_2O) \left(\frac{n_{[RuO_2]_{(s)}}}{V} \right) [O_3]$$

- Revolatilisation phenomenon from ruthenium oxide deposits at low temperature (40°C/90°C) has been experimentally highlighted.
- Revolatilisation is induced by the oxidising effect of ozone producing $RuO_4(g)$ species.
- The oxidation reaction is modelled as first-order with respect to $[O_3]$ and $[H_2O_{(aq)}]$.

Preliminary evaluation of off-site radiological impact

- Example for a LOCA scenario : filtered releases from reactor containment (beyond 24h after core fusion).
- In this scenario, vessel failure occurred 6h after the reactor scram and the French Ultimate procedure of containment venting ("U5") occurred after 36h.
- Leakages rates from containment: before vessel failure $\rightarrow 0.156\%$ vol./d, after $\rightarrow 0.624\%$ vol./d.
- Containment spray system is considered out of service.
- Surface activities (soil) and volumetric activities (atmosphere) are calculated by a dispersion model taking into account meteorological data.
- 4 radio-nuclides are considered: ¹⁰³Ru, ^{103m}Ru, ¹⁰⁶Ru, ^{106m}Ru, and dose coefficients associated are linked with all exposition mode (internal contamination, external irradiation).



Irradiation sources of ⁶⁰Co

$$\frac{d[RuO_4]_{(g)}}{dt} = k_{(T)} \cdot DR \cdot (P_{vap})^3 \cdot N_{(Ru-dep)} \cdot \frac{S}{V}$$

- Radiolytic oxidation phenomenon was also detected during radiolysis experiments in the EPICUR irradiation facility.
- Temperature and steam are two key parameters.
- The high oxidation enhancement resulting from an increased humidity rate is assumed to be caused by hydroxyl radical (OH).

Conclusions: main achievements in SARNET

- Air ingress scenarios have been simulated: such scenarios have significant implication for the source term and specifically towards ruthenium FP, which is highly radiotoxic.
- Under very oxidising conditions, like those induced by air, ruthenium can form volatile oxide species from metallic ruthenium still present in the fuel.
- These ruthenium oxides can be transported through the RCS and are able to reach the containment building.
- Due to various radio-chemical reactions in the containment, the species RuO_4 can persist in the gaseous phase during several days.
- Potential release of $RuO_4(g)$ in the environment may occur due to the French ultimate procedure of containment venting, leading thus to a probable increase of the total efficient dose in the environment.

The assessment of $[RuO_4(g)]$ remaining in the reactor containment atmosphere before containment venting procedure is of primary importance!