REDUCTION OF RADIOLOGICAL ACCIDENT CONSEQUENCES

Title	**FP transport/behaviour in RCS**
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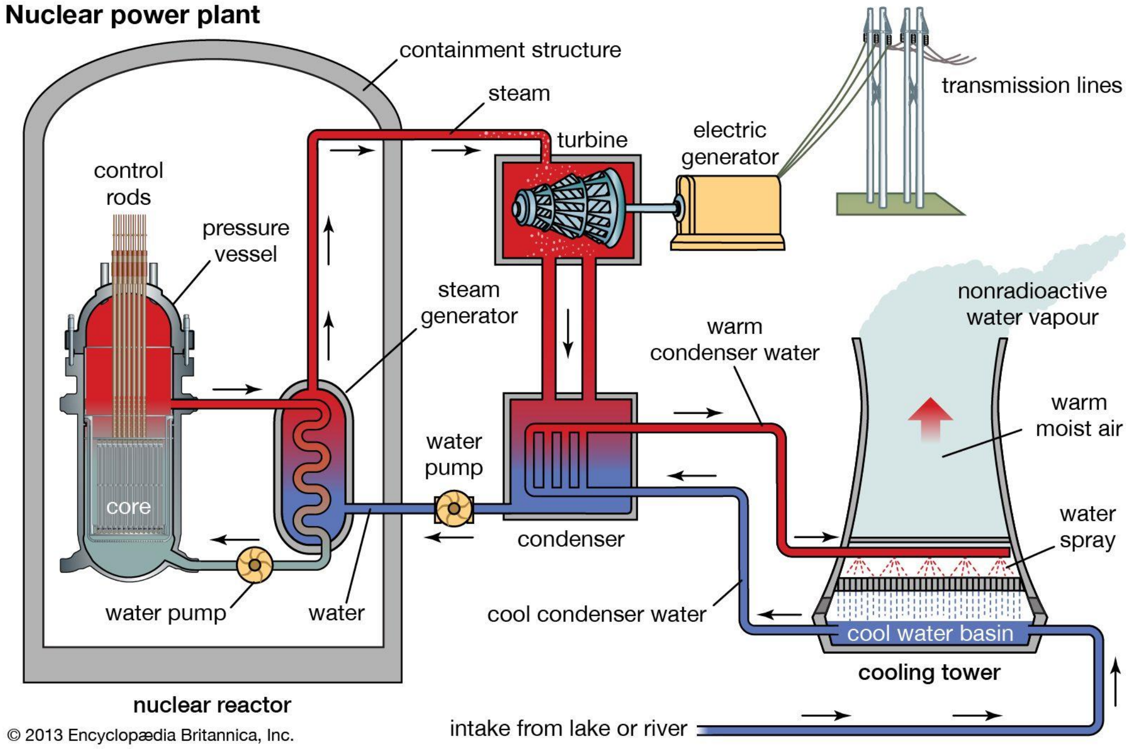
Contents

Introduction and motivation

REDUCTION OF RADIOLOGICAL CONSEQUENCES OF DESIGN BASIS & DESIGN EXTENSION ACCIDENTS

- Main FP release comes from the primary circuit, where two typical situations may occur
  - Containment bypass accidents
  - LOCAs inside containment
- **FP release from primary is an important initial and boundary condition**, which affects the estimation of the source term
- What should be the outcome
  - Target phase
  - Amount
  - Chemical form
  - All mentioned are often time dependent



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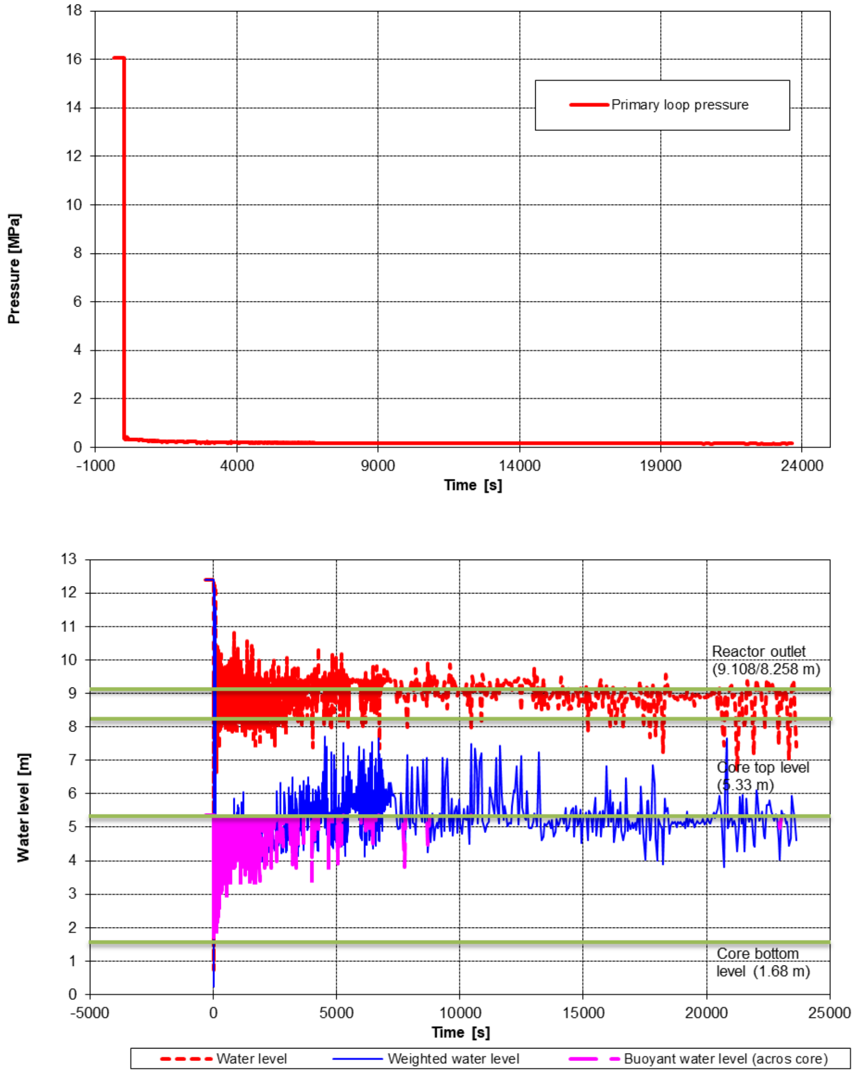
Primary circuit during LOCA

- Four phases of LOCA in the core
  - Blowdown ~30 s
  - Refill ~ 40 s
  - Reflood ~40 – 250 s
  - Quenching ~ 250 s and more
- Possibility to uncover the core
  - Release to both water and gas phase
- Rapid depressurisation during blowdown may lead to flashing, ie. FPs may entrain the steam phase
- Generally unstable conditions



T2.3 Report on LOCA simulations

Chemical situation in the primary

- The primary circuit does not contain only water
- H_3BO_3 boric acid
  - pH↓
  - Additive to control reactivity
- LiOH lithium hydroxide
  - pH↑
  - Additive to increase pH in western PWR
  - VVER use KOH
- NH_3 ammonia
  - pH↑
  - Formed from hydrazine (NH_4), mitigates oxide
- Impurities
- Dose rate
- Wide range of pH

Fission products in the primary

- Pre accidental presence of FPs from leaking fuel + spiking of some elements
- Three groups of FPs to be considered (different behaviour)
  - Generally volatile FPs considered to be released from fuel
    - Noble gasses
    - Aerosols
    - Iodine
- In a typical 900MWe PWR, following masses can be found in the fuel
  - ~ 300 kg of Xe
  - ~ 160 kg of Cs
  - ~ 12 kg of I
  - Not all of the FPs are important for radiological consequences

Existing Approaches

- How to deal with the FP source term from the primary circuit?

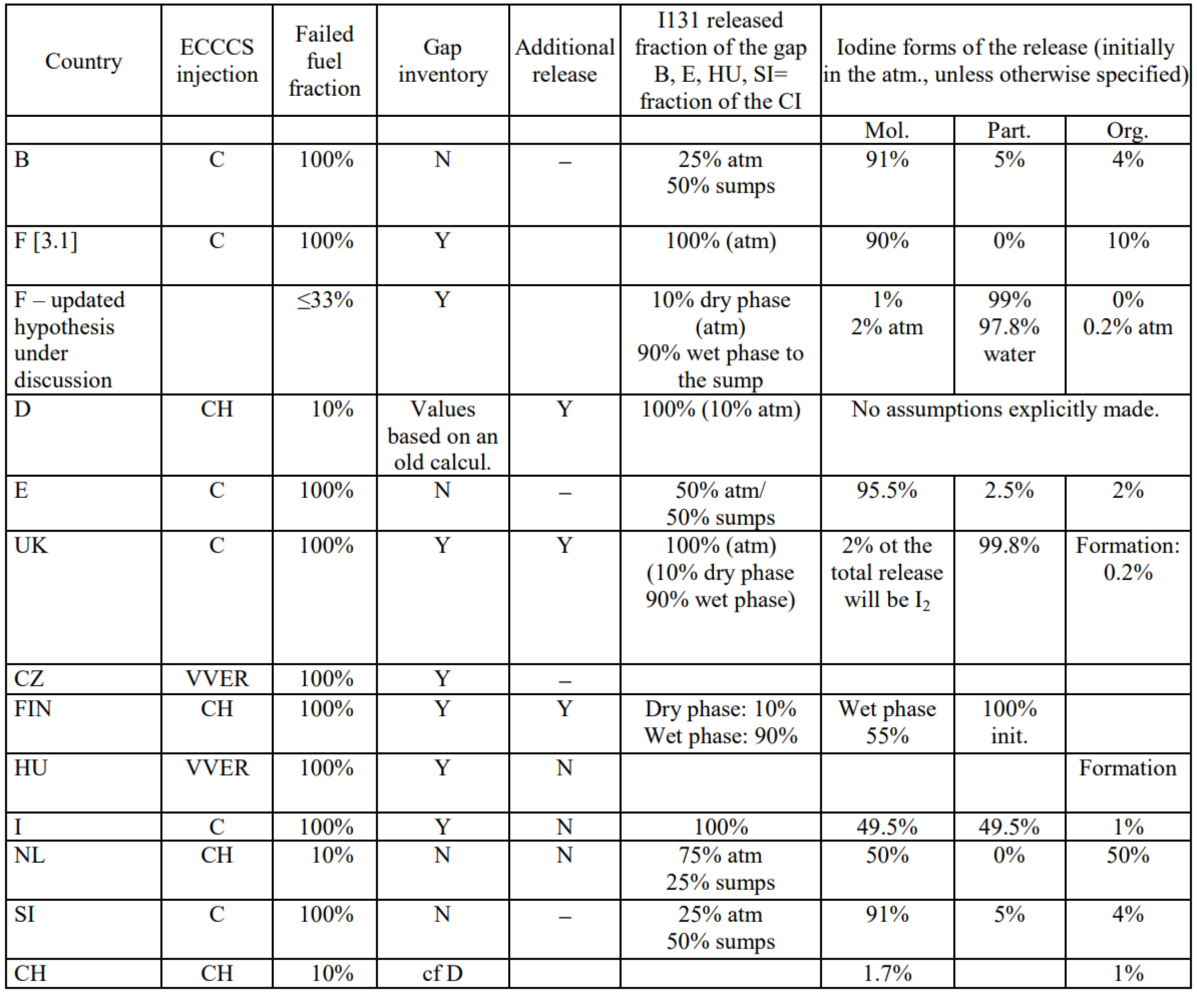
  - „Methodical approach“
    - Simplified approach, which does not calculate the real FP transport in the primary
    - Depending on design, experience and national habits, different methodologies are adopted
    - Typically widely accepted R.G. 1.183, 1.195
    - Generally very conservative, problem of determination correct ratio between iodine forms

  - „Computational approach“
    - Realistic approach, which calculates the transport and retention of FPs in the primary circuit
    - More complex, demanding both on computational power and manpower
    - Need of validation data

Existing Approaches

- Significant variation between the countries
- Regulatory guides

R.G. 1.183	R.G. 1.195
95 % part 4.85 % elem 0.15 % org	5 % part 91 % elem 4 % org



Country	ECCCS injection	Failed fuel fraction	Gap inventory	Additional release	I131 released fraction of the gap B, E, HU, SI= fraction of the CI	Iodine forms of the release (initially in the atm., unless otherwise specified)		
						Mol.	Part.	Org.
B	C	100%	N	–	25% atm 50% sumps	91%	5%	4%
F [3.1]	C	100%	Y		100% (atm)	90%	0%	10%
F – updated hypothesis under discussion		≤33%	Y		10% dry phase (atm) 90% wet phase to the sump	1% 2% atm	99% 97.8% water	0% 0.2% atm
D	CH	10%	Values based on an old calcul.	Y	100% (10% atm)	No assumptions explicitly made.		
E	C	100%	N	–	50% atm/ 50% sumps	95.5%	2.5%	2%
UK	C	100%	Y	Y	100% (atm) (10% dry phase 90% wet phase)	2% ot the total release will be I_2	99.8%	Formation: 0.2%
CZ	VVER	100%	Y	–				
FIN	CH	100%	Y	Y	Dry phase: 10% Wet phase: 90%	Wet phase 55%	100% init.	
HU	VVER	100%	Y	N				Formation
I	C	100%	Y	N	100%	49.5%	49.5%	1%
NL	CH	10%	N	N	75% atm 25% sumps	50%	0%	50%
SI	C	100%	N	–	25% atm 50% sumps	91%	5%	4%
CH	CH	10%	cf D			1.7%		1%

NEA (2009), Nuclear Fuel Behaviour in Loss-of-coolant Accident (LOCA) Conditions: State-of-the-art Report, OECD Publishing, Paris

What to choose

- Which approach is the best?
  - DBA (depending on country) often use conservative approach, i.e. maximisation of evaluated results, using rather conservative (sometimes extreme) initial and boundary conditions
  - For DBA „methodical approach“ used to be acceptable
  - With increasing demands on NPPs – power uprate, reduction of safety limits and criteria etc., using conservative approach hits the legislative limits on radiological consequences
  - Possibility to reduce the results by application of „computational approach“, which may lead to retention of the FPs in the primary and better estimation of the FPs mass entraining the containment

Transport of the FPs

- Complex problem dealing with physical and chemical nature of the elements as well as harsh conditions of the primary circuit
  - Pressure, temperature, pH, dose rate
  - Transport with water and steam + interaction on phase interface
- Groups of FPs
  - Volatile FPs – NGs, Cs, Te, I …
  - Semi-volatile FPs – Mo, Ba
  - Low-volatile FPs – Sr, Ru …
  - Non-volatile FPs –Zr, Nd …
- From the radiological consequences point of view
  - Most important FPs are those having significant high conversion factors
  - Cs, **I**, NGs
  - For DBA LOCA iodine impact is enormous, up to 95 % of the dose

Experimental programme

- During the last 25 years, numerous experiments aiming at FP and iodine transport representing various conditions were conducted
- PHEBUS FPT
  - Experiments revealed high elemental iodine concentrations entraining the containment (CsI was expected to be dominant)
  - Highest values obtained for FPT3, where B_4C control rods were used (similar composition is used at VVER-1000 in Temelin)
- CHIP, GAEC
  - Extensive test campaign using boron and other compounds to investigate release of I_2
- Majority of the R&D activities aimed at severe accidents
  - Application of the results to DBA is generally limited

Iodine in primary circuit (I-O-H)

- Oxidation states ranging from -1 to +7
- Low solubility of I_2 in water
- Iodine I_2 is chemically stable in environments with slightly acid pH
- When reaching the neutral pH, iodine hydrolises

$$I_2 + H_2O \rightarrow HOI + I^- + H^+$$

$$3HOI \rightarrow IO_3^- + 2I^- + 3H^+$$

- For pH > 8 iodine dismutes rapidly and totally (inverse Dushman reaction)

$$3I_2 + 3H_2O \Leftrightarrow IO_3^- + 5I^- + 6H^+$$

- What it means?

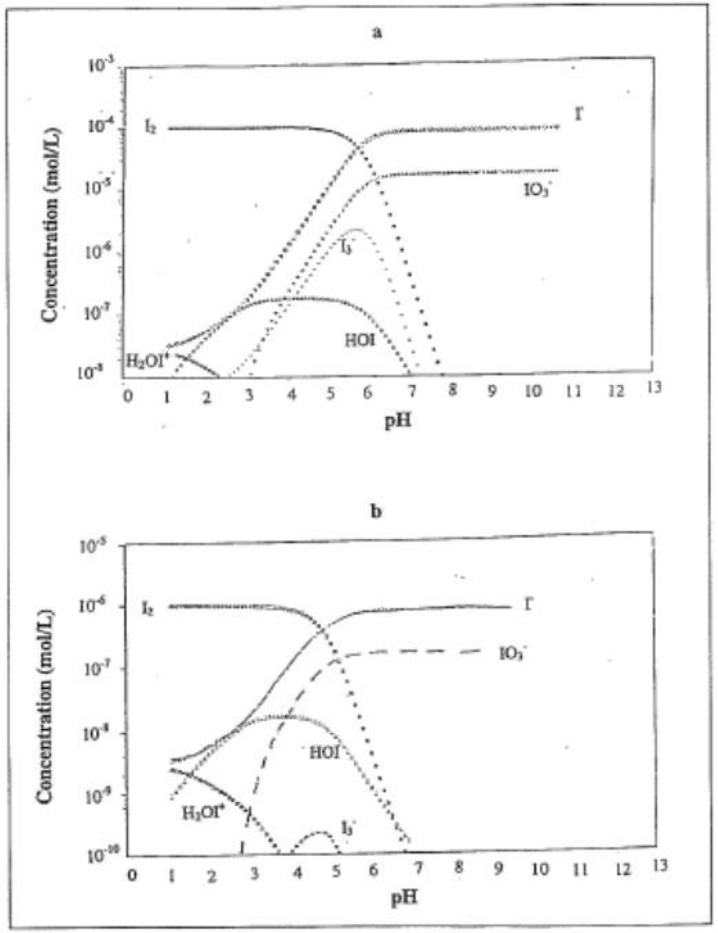


Figure II-5 : Courbes de répartition à 25°C des espèces formées par hydrolyse de l'iode en fonction du pH du milieu.

A : concentration initiale d'iode : 10^{-4} mol.L^{-1}
B : concentration initiale d'iode 10^{-6} mol.L^{-1}

From C. Cau Dit Coumes,phD Thesis

Iodine in primary circuit (Cs-I-O-H)

- Cs abundance in fuel is 10 times higher than I
- In standard fuel conditions iodine is released as a CsI vapour
- Depending on conditions, following situations may occur
  - Reducing or mildly oxidizing steam
    - CsI is stable to 1600 °C, below 1300°C it will plate on surface and will be removed with oxygen and water. If the CsI/steam mixture is condensed, iodine will be converted to I^-
  - Steam/air
    - Oxidizing conditions, where CsI may be oxidized to Cs_2O and I_2, with further reaction of Cs_2O to CsOH
  - High temperature water
    - CsI will dissolve to Cs and I, where the present dose rate may oxidize I to IO_3^-

Assessment of iodine behaviour in reactor containment buildings from a chemical perspective
Lemire, R.J.; Paquette, J.; Torgerson, D.F.; Wren, D.J. Fletcher, J.W.
Atomic Energy of Canada Ltd., Pinawa, Manitoba. Whiteshell Nuclear Research Establishment 1981

Iodine in primary circuit (B-Cs-I-O-H)

REDUCTION OF RADIOLOGICAL CONSEQUENCES OF DESIGN BASIS & DESIGN EXTENSION ACCIDENTS

- Several other species are present within the RCS
    - This makes the estimation and simulation even more difficult
- Experimental activities (e.g. PHEBUS FPT3) revealed that in B-Cs-I-O-H systems, boron serves as an effective sink for cesium, which yields to massive increase of I_2 fraction
    - Unfortunately, boron is a common specie in the primary (boric acid used to control the reactivity)
- Boron – cesium reactions mentioned in NUREG/CR-6193

$$CsOH + HBO_2 \Leftrightarrow CsBO_2 + 2H_2O$$

$$CsI + HBO_2 \cdot nB_20_3 \Leftrightarrow CsBO_2 \cdot nB_20_3 + HI$$

- Furthermore NUREG/CR-4817 (partitioning in steam generator) mentions that for given boric acid concentration, iodine volatility is pH independent
- Experimental acitvities are necessary

Summary

- RCS chemistry during DBA is very complicated due to chemical situation and rapid changes of the thermal hydraulic conditions
- Transport in primary circuit affects the in-containment source term significantly, having impact to further transport processes in containment and source term determination
- Methodical vs. computational approach
- Limited experimental activities dedicated to DBA conditions
  - This area becomes even more important for the SMRs
- Transport of iodine is affected by the complex chemistry, which depends on the presence of other chemical compounds
  - Significance of pH / boric acid concentration

Thank you!

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