Chemistry and transport of fission products (CATFIS) project

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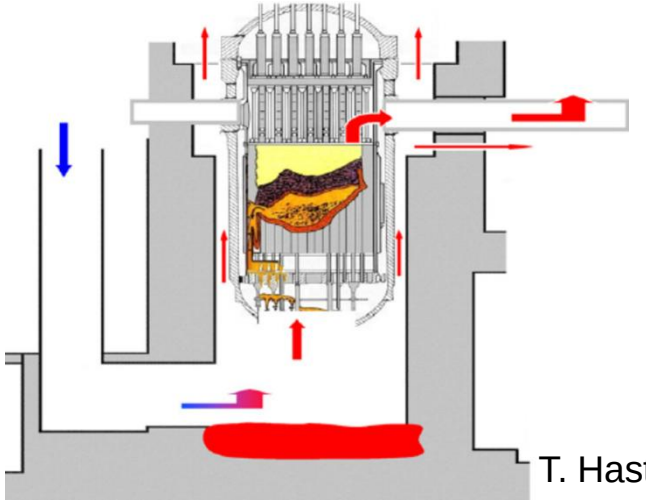
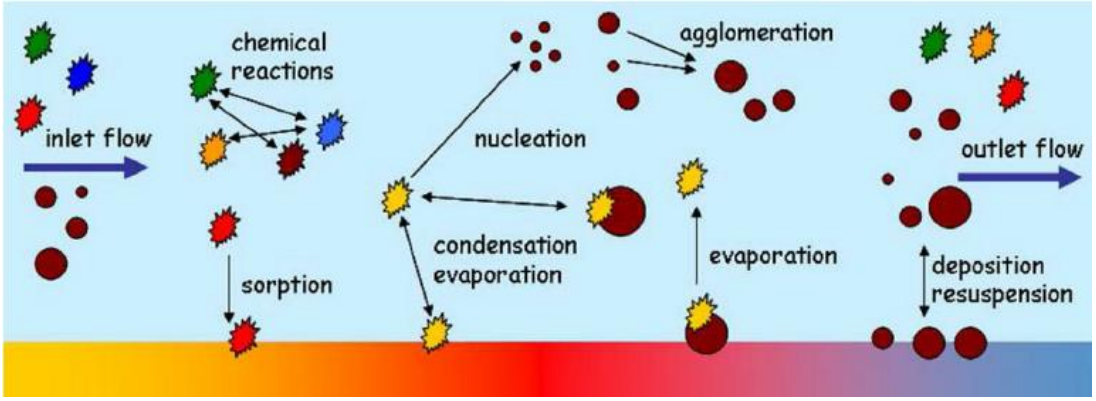
Objectives of CATFIS

- The aim is to investigate the **transport and chemistry** of gaseous and particulate **fission products** in severe accident conditions.

- The emphasis is on the **phenomena**, which are **poorly-known** internationally or **not considered in the** current severe accident **analysis codes** due to the lack of information.



T. Haste et al. NED 2009



L. Cantrel et al. NED 2014

Severe accident research priorities
(W. Klein-Heßling et al., SARNET2 / ERMSAR 2013)

- High priorities:
  - Quantify the source term, **in particular for Ru**, under oxidation conditions / air ingress for HBU and MOX.
  - Improve predictability of **iodine and ruthenium** species exiting RCS to provide the best estimate of the source into the containment.

- CATFIS project: The **main focus** is on the behaviour of **iodine and ruthenium** which are highly radiotoxic and the mitigation of their possible source term is of utmost importance.

Two examples of research topics

- Primary circuit chemistry of **iodine**
  - *Surface reactions – formation of gaseous iodine*

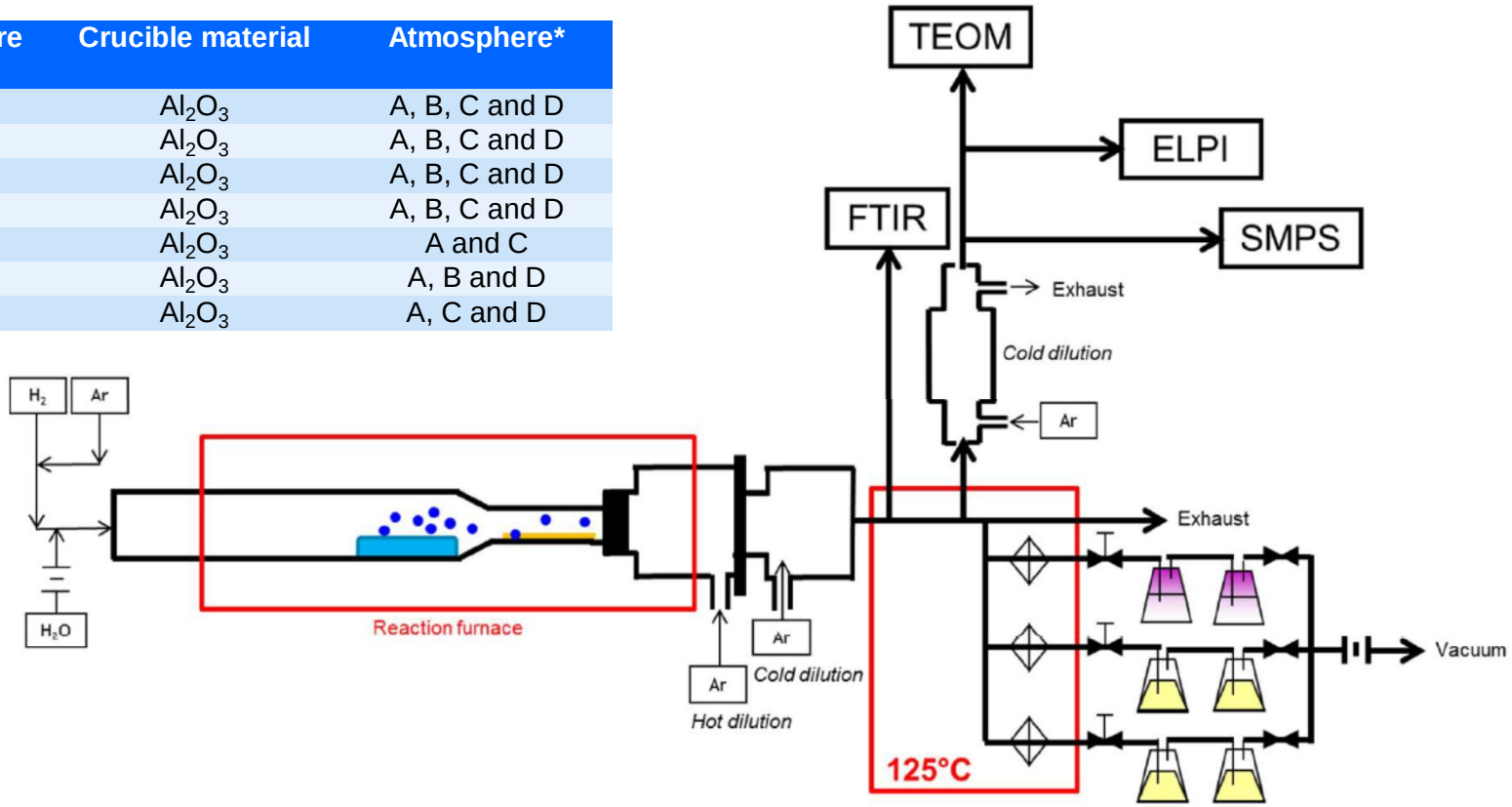
- Primary circuit chemistry of **ruthenium**
  - *Gaseous and aerosol compounds – formation of gaseous RuO_4*

Primary circuit chemistry of iodine

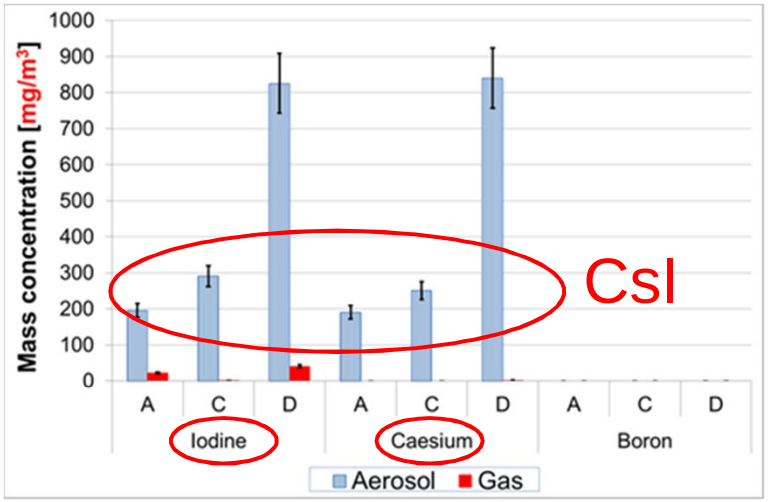
- Effect of **fission product deposits** reactions on the RCS surface for the **formation of gaseous iodine** and the **transport** as gas and particles.

Precursors	Temperature [°C]	Crucible material	Atmosphere*
CsI	650	Al_2O_3	A, B, C and D
CsI	400	Al_2O_3	A, B, C and D
CsI + B_2O_3	650	Al_2O_3	A, B, C and D
CsI + B_2O_3	400	Al_2O_3	A, B, C and D
CsI + CsOH	650	Al_2O_3	A and C
CsI + CsOH + B_2O_3	650	Al_2O_3	A, B and D
CsI + CsOH + B_2O_3	400	Al_2O_3	A, C and D

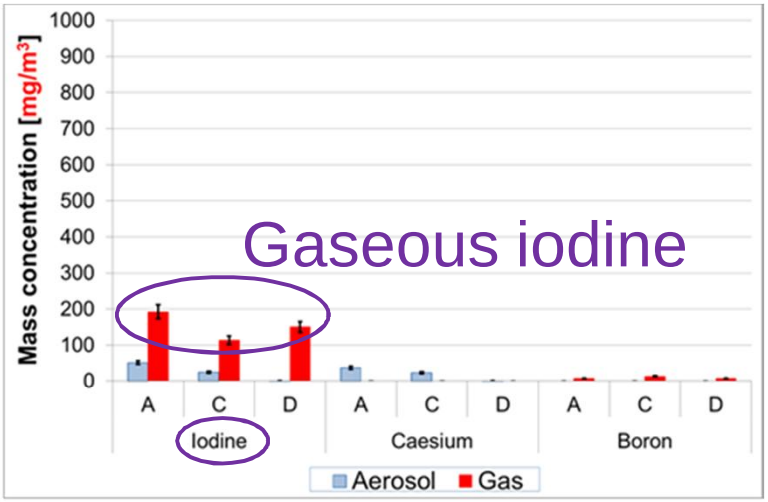
Gas	Atmosphere composition vol-%			
	A	B	C	D
Argon	86.7	83.9	76.1	86.7
Steam	13.3	13.5	13.4	0
H_2	0	2.6	10.5	0
Air	0	0	0	13.3



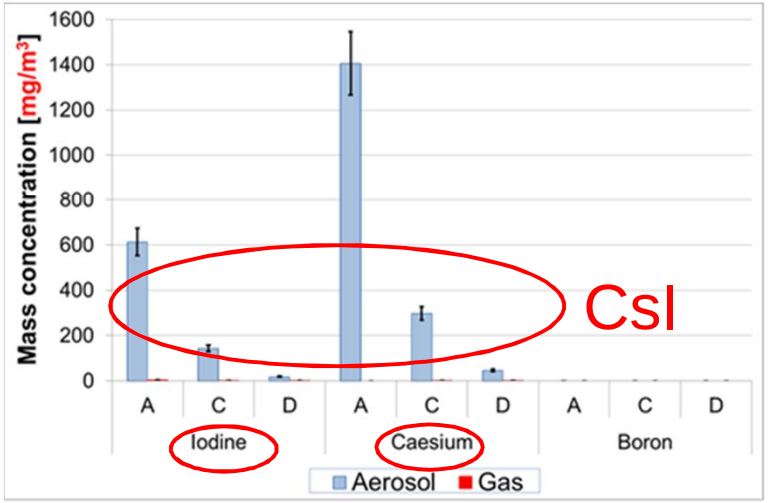
Primay circuit chemistry of iodine



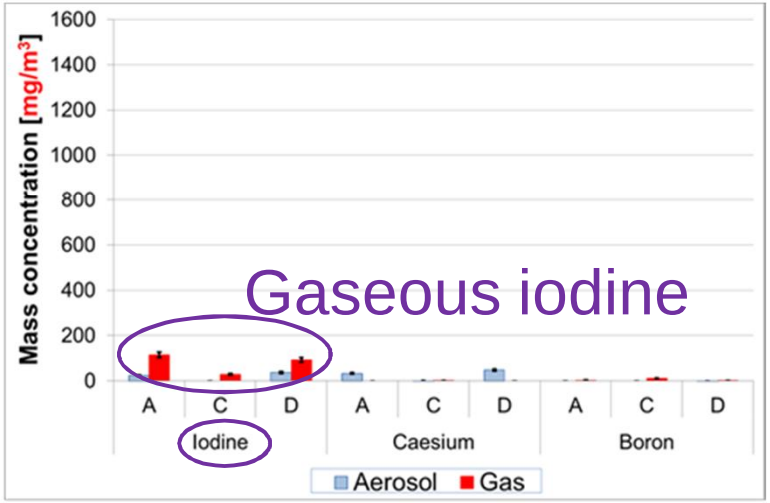
CsI - 650°C



CsI + B_2O_3 - 650°C



CsI + CsOH - 650°C



CsI + B_2O_3 + CsOH - 650°C

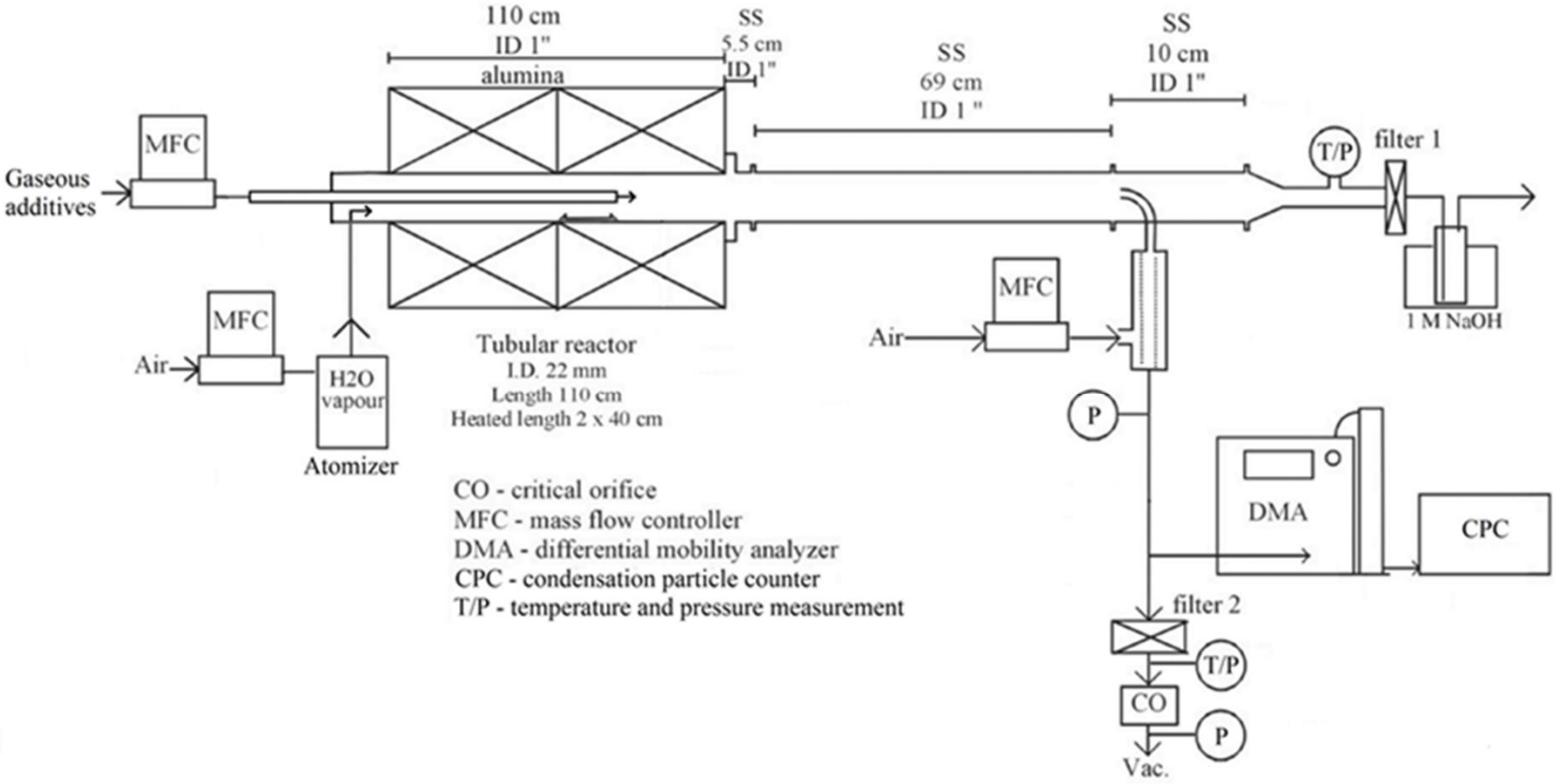
Primary circuit chemistry of iodine - Outcomes

- FP deposits on the RCS surfaces can act as a source of gaseous iodine in a SA.
  - May be an important source of gaseous iodine in a long term.
  - It should be considered also when mitigating the possible releases to the environment (e.g. FCVS).

- Atmosphere composition has an effect on the release of iodine from the FP deposits on RCS surfaces.
  - Increasing concentration of oxygen (e.g. air atmosphere) seemed to enhance the formation of gaseous iodine.

Primary circuit chemistry of ruthenium

- Effect of air radiolysis products and aerosols on the formation and transport of ruthenium as gas and particles in RCS.
  - NKS-R collaboration with CHALMERS.



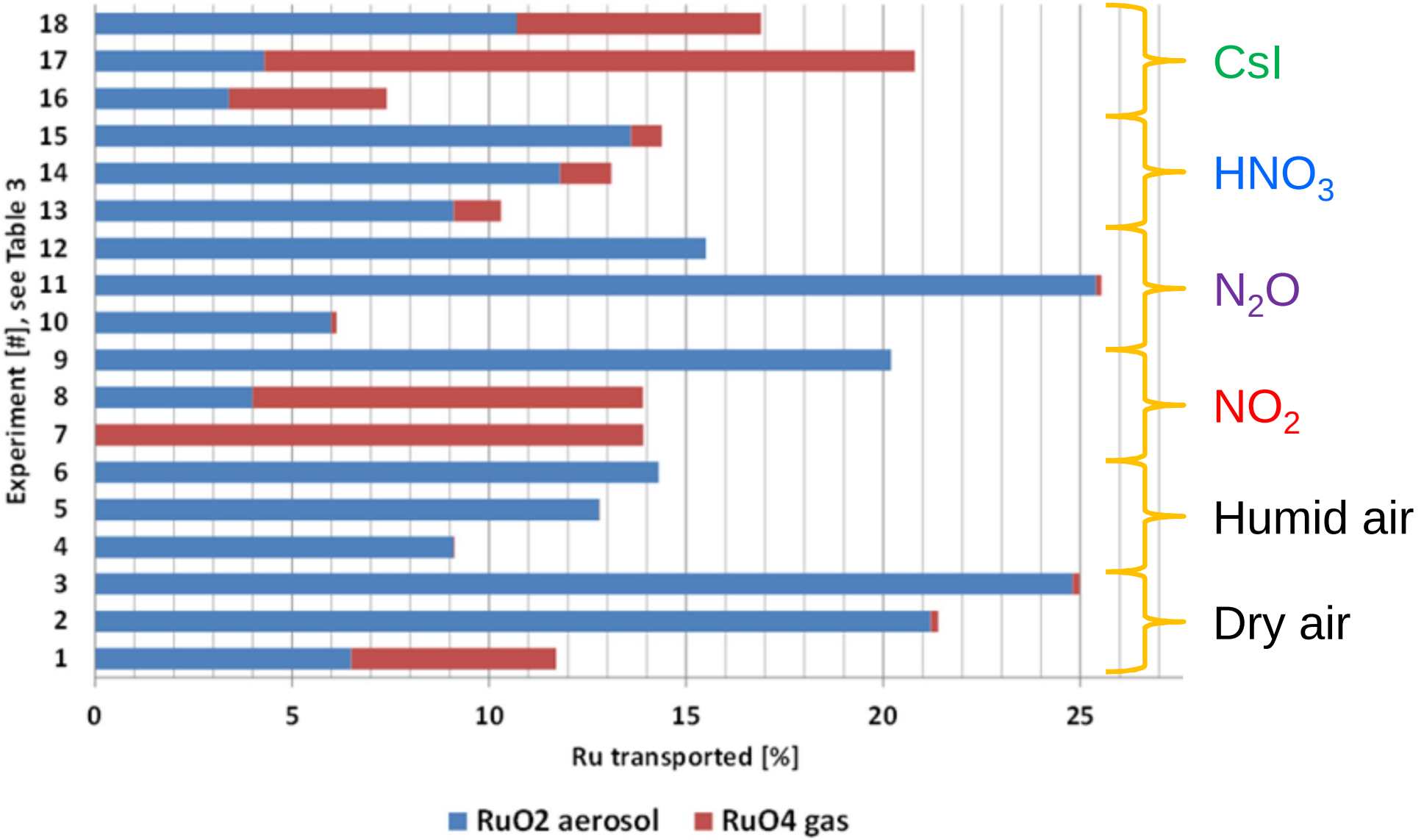
Primary circuit chemistry of ruthenium

Exp	T [K]	Precursor	Gas phase	Additive precursor conc.	Humidity
1	1300	RuO_2	Air	-	dry
2	1500	RuO_2	Air	-	dry
3	1700	RuO_2	Air	-	dry
4	1300	RuO_2	Air	-	humid
5	1500	RuO_2	Air	-	humid
6	1700	RuO_2	Air	-	humid
7	1300	RuO_2	Air+NO_2	50 ppmV NO_2	humid
8	1500	RuO_2	Air+NO_2	50 ppmV NO_2	humid
9	1700	RuO_2	Air+NO_2	50 ppmV NO_2	humid
10	1300	RuO_2	Air+N_2O		
11	1500	RuO_2	Air+N_2O		
12	1700	RuO_2	Air+N_2O		
13	1300	RuO_2	Air+HNO_3	5 ppmV HNO_3	humid
14	1500	RuO_2	Air+HNO_3	5 ppmV HNO_3	humid
15	1700	RuO_2	Air+HNO_3	5 ppmV HNO_3	humid
16	1300	RuO_2	Air+CsI	4 wt.% of CsI solution	humid
17	1500	RuO_2	Air+CsI	4 wt.% of CsI solution	humid
18	1700	RuO_2	Air+CsI	4 wt.% of CsI solution	humid

Air radiolysis products

CsI aerosol

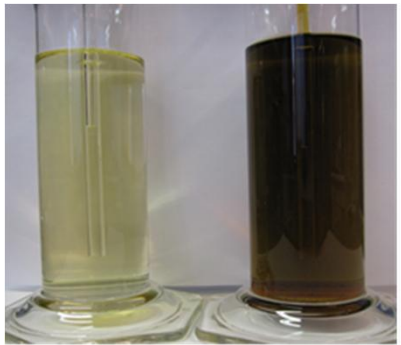
Primary circuit chemistry of ruthenium

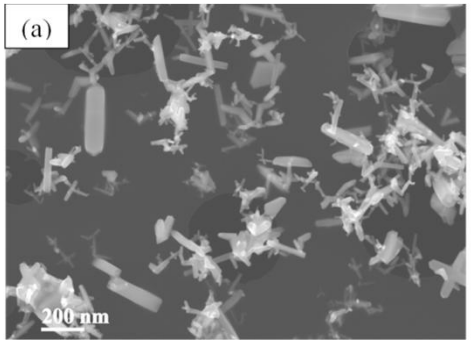


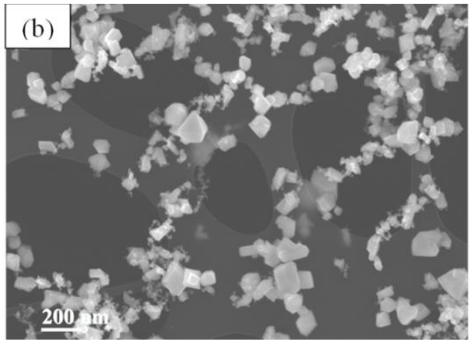
Primary circuit chemistry of ruthenium - Outcomes

- Air radiolysis products may significantly increase the transport of (low volatile) ruthenium as a gaseous compound into the containment building (air ingress conditions).
  - Internationally, separate effect experiments on Ru transport in RCS has usually been performed in pure air-steam atmospheres.

- The highest transport of ruthenium as a gaseous compound was observed when CsI reacted with Ru oxides.
  - **Important observation**, not discussed internationally before.
  - Detailed analysis of the reasons behind the observation is ongoing.







Conclusions

- **New information** for the improvement of SA analysis codes / FP transport models has been produced.
  - Enhancement of **nuclear safety**.

- The performed experiments have also been noticed internationally.
  - **Iodine chemistry:** Further joint project with Japanese JAEA organization on the effect of boron on iodine chemistry in the RCS.
  - **Ruthenium chemistry:** OECD/NEA STEM-2 project will verify these findings and perform complementary experiments.

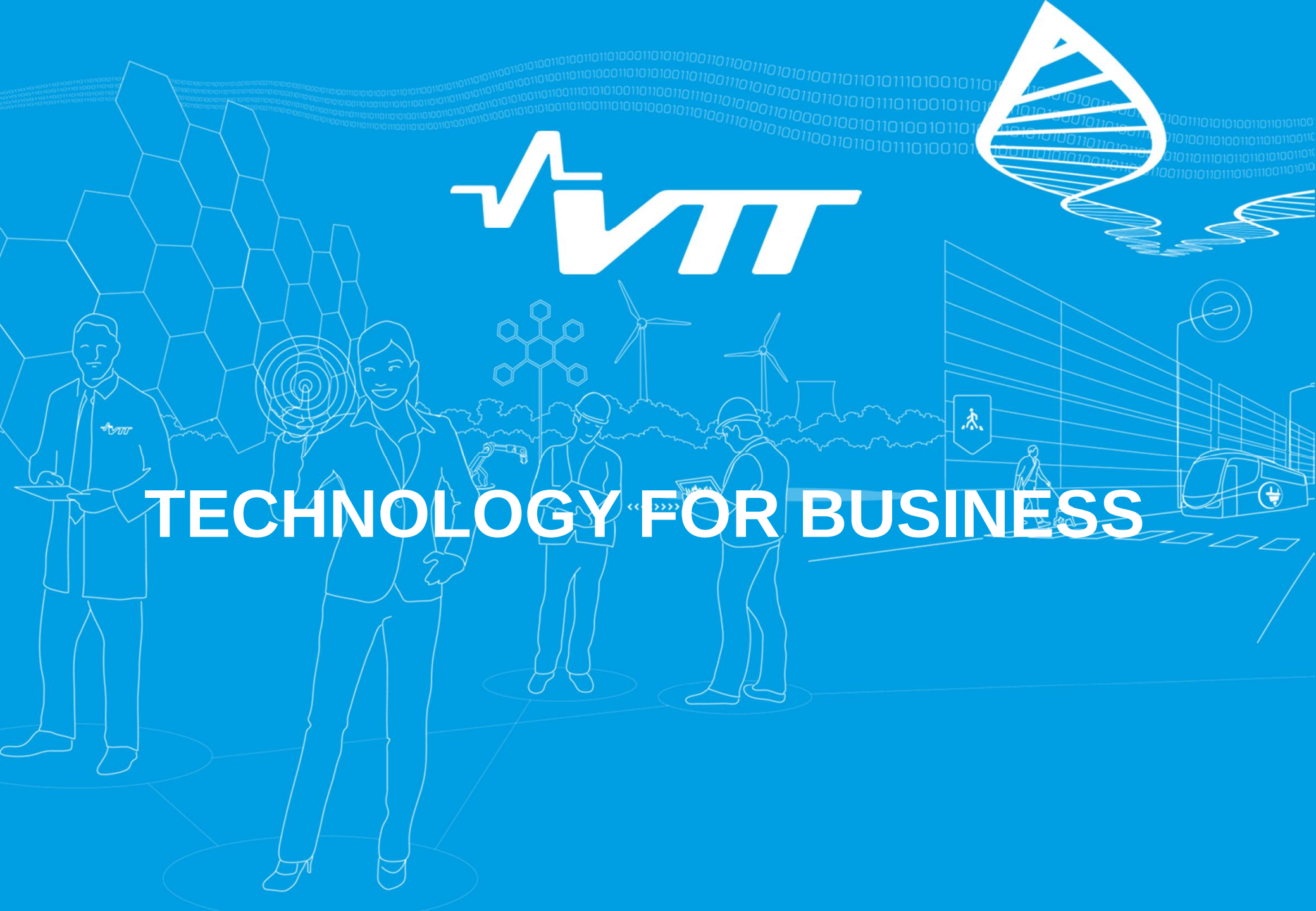
Further studies

- In CATFIS 2017, also experiments on the retention of FPs in FCVS pool.
  - Increasing interest on European level:
    - EU PASSAM project in 2013-2016.
    - NUGENIA IPRESCA project starting in 2017.

- In general, collaboration with other organisations is being looked for all the time.

References

- Gouëllo, M., Hokkinen, J. 2016a. Experimental Study on the Behaviour of CsI on Primary Circuit Surfaces: Effects of Boron and Air Ingress on Iodine Transport during a Severe Nuclear Accident, VTT report VTT-R-00046-16 (2016).
- Gouëllo, M., Hokkinen, J. 2016b. Experimental Study on the Behaviour of CsI on Primary Circuit Surfaces of a Nuclear Power Plant: Effects of Boron and Air Ingress on Iodine Transport at 400°C, VTT report VTT-R-03242-16 (2016).
- Gouëllo, M., Hokkinen, J., Kärkelä, T., Auvinen, A. 2016c. Experimental Study of the Boron and Air Effects on Iodine Transport in the Primary Circuit during Severe Nuclear Accident, International Congress on Advances in Nuclear Power Plants, San Francisco, USA, April 17-20, 2016.
- Kajan, I., Kärkelä, T., Auvinen, A., Ekberg, C. 2017. Effect of nitrogen compounds on transport of ruthenium through the RCS, J Radioanal Nucl Chem, Published online 11th of January, 2017, DOI 10.1007/s10967-017-5172-7.
- Kärkelä, T., Auvinen, A. 2009. Experimental Study on Iodine Chemistry (EXSI) - Facility for Primary Circuit experiments. Espoo, Finland, Research report VTT-R-02791-09.
- Kärkelä, T., Vér, N., Haste, T., Davidovich, N., Pyykönen, J. & Cantrel, L. 2014. Transport of ruthenium in primary circuit conditions during a severe NPP accident. Ann. Nucl. En. 74, 173-183.
- Kärkelä, T., Kajan, I., Tapper, U., Auvinen, A., Ekberg, C. 2016. Ruthenium transport in a RCS with airborne CsI, Submitted to Progress in Nuclear Energy.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