SOURCE TERMS
Na aerosols and fission products

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Source Terms: Na aerosols and fission products

Contents

- Main source terms and phenomena
- Overview of knowledge & experimental data
- Current code predictability & major needs

Source Terms: Na aerosols and fission products

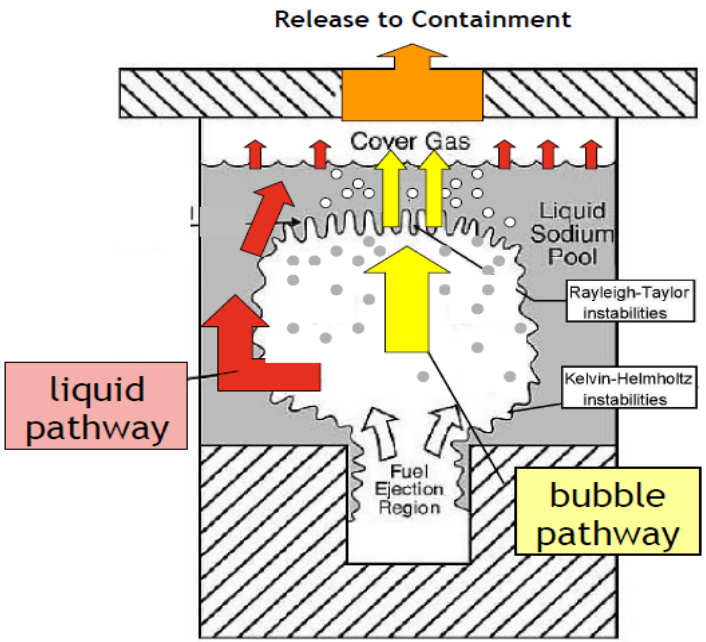
Context

- **Predicting the radiological consequences of SFR accidental scenarios is of paramount importance (public health and acceptance)**

- **Increased safety of NPPs is nowadays required (especially after FKS) implying better evaluation of Source Terms (Best Estimate calc. Plus Uncertainties) then**
  - ✓ **A better theoretical knowledge**
  - ✓ **More comprehensive models**
  - ✓ **Development of efficient numerical tools**

- **Realistic quantification of STs requires a good understanding of whole phenomena and characterization of main parameters impacting FP retention/releases**

Main sources terms: origin



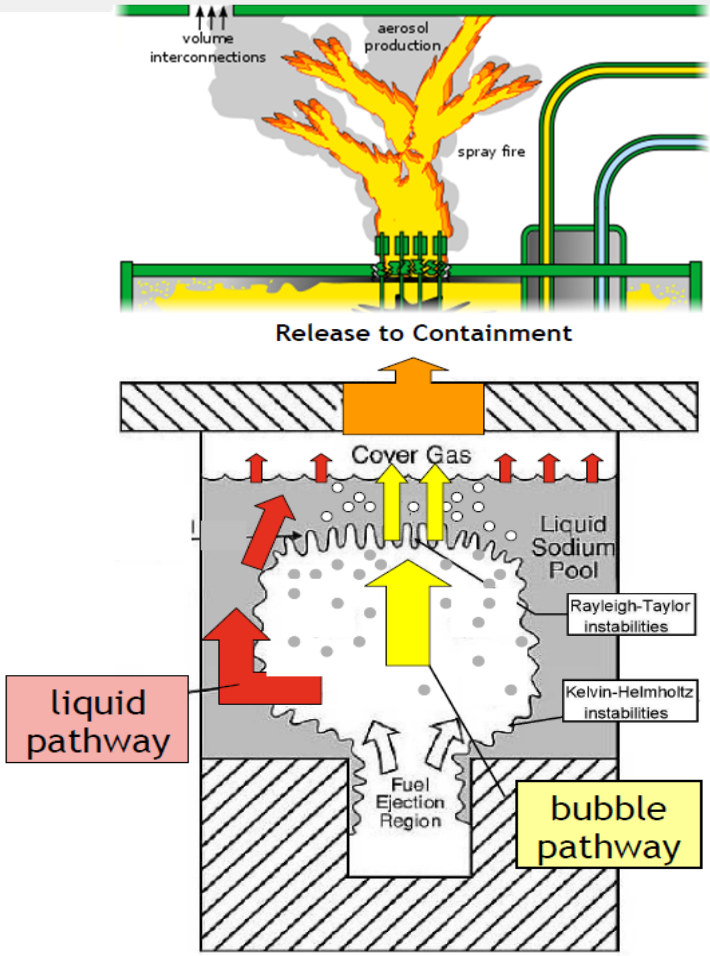
In-vessel : 2 transport pathways

✓Aerosol transport: the bubble pathway
A minority of safety studies have investigated bubble scrubbing

✓Evaporation: the liquid pathway
Main trend in safety studies has been to base primary-system transfer reactions on this pathway

Common implicit or explicit assumption has been that everything remains in liquid Na
Not obvious: can at best be partly true since, after energetic FCI, a heterogeneous mixture is formed
For it to be ientrely valid, the pool scrubbing DF should tend to ∞

Main sources terms: origin



In-containment : 2 releases pathways

✓ Spray fires
High T(Na) and specific surface:
Early and fast RN vaporization

✓ Pool fires
Lower T(Na), O_2 access/pool surface limitation
Late releases over o longer period of time

Two different source terms to be considered

- Instantaneous ST (bubble pathway + spray fire)
  Fast release of RN (volatiles to non volatiles), in-vessel FP retention/partition, remaining FP ejection with Na

- Delayed ST (liquid pathway + pool fire)
  Additional releases of volatiles & semi volatiles from RN vaporisation, longer duration

Main sources terms: nature

$Na/Na_2O/Na_2O_2/NaOH/Na_2CO_3$ aerosols (from Na fires)

- ✓ Na activation:

$$^{23}Na + n \xrightarrow{❶} {}^{24}Na \xrightarrow{15h} {}^{24}Mg + \beta^- + \gamma$$
$$\xrightarrow{❷} {}^{22}Na + 2n \xrightarrow{2,6a} {}^{22}Ne\ (+\beta^+ \text{ ou } - e^-) + X/\gamma$$
$$\xrightarrow{❸} {}^{23}Ne + p \xrightarrow{37s} {}^{23}Na + \beta^-$$

Specific activity of Na in SFRs low but large amount (source IAEA, 1993) →

In case of primary Na release: potential high β- + γ activity during several days

- ✓ High chemical toxicity of some species (NaOH)

Immediately Dangerous for Life and Health: 10 mg (NaOH)/m^3 for 30 min

isotope half-life reactor	ref \ units	^{24}Na 15 h TBq/kg	^{22}Na 2.6 a MBq/kg
EBR-II	[27]	0.1	5
FFTF	[27]	0.41	20
KNK II	[13],[28]	2.5	25
PFR	[104],[24]		14
RAPSODIE	[10]		40
PHENIX	[10],[143]		22
BOR-60	[25],[109]		22
BN-350	[149]	0.37	28
BN-600	[109]	0.75	26

FP dissolved/suspended in Na

- ✓ Higher specific activity for some FP (^{131}I) but lower releases
- ✓ Releases from Na vaporisation/combustion dependent on their physico-chemical characteristics (volatility, solubility, chemistry)

Main phenomena & status

Aerosol/FP bubble scrubbing

✓ Few studies (FAUST, CARAVELLE….), challenging experiments & models

FP vaporization and partition from hot Na pools/Na fires

✓ Some experiments available for FP released from hot Na pools (NALA) few for FP releases from Na pool fires (FANAL), detailed data (release kinetics) often missing

✓ Current past codes often use correlations with fitted parameters (CONTAIN), mechanistic modelling is missing → SCOPE of ESFR-SMART R&D work

Na aerosol generation, chemical transformation & transport

✓ Na fires largely studied in the past (FAUNA, EMIS…) for combustion and aerosol generation, more recent experiments focused on Na aerosol speciation *(studied in past JASMIN project)*

✓ Combustion is generally well predicted (pool fires?), some codes embedded ageing models (ASTEC-Na..)

FP interactions with Na-AER

✓ Few experiments only (FANAL, ATF), on going experiments (ATF) & model developments

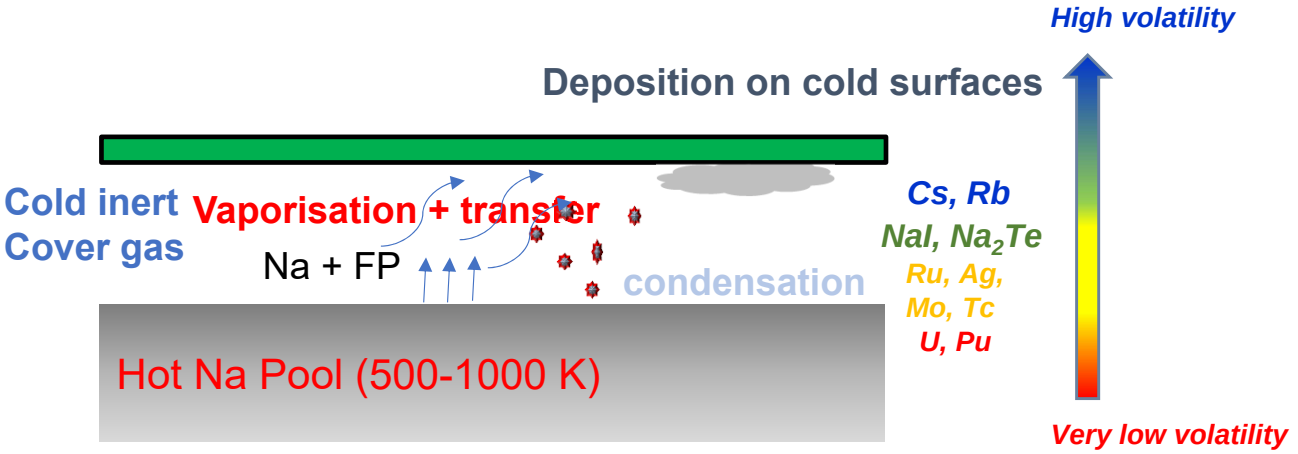
FP vaporisation from hot Na

FP releases function of pool surface and bulk conditions → impact of

- ✓ Na evaporation (i.e. Na temperature)
- ✓ FP characteristics (volatility, solubility, interaction with Na): I releases lower than Cs due to NaI

But also on

- ✓ Mass transfer in gas phase (natural or forced convection)
- ✓ Condensation on aerosols and cold surfaces



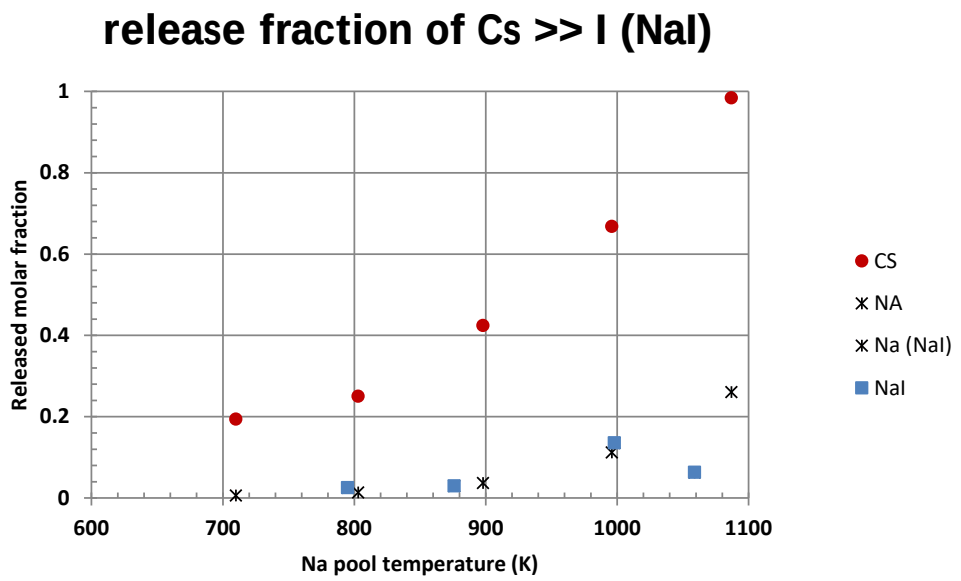
$$K_d = \frac{\dot{m}_i / m_{i,pool}}{\dot{m}_{na} / m_{na,pool}}$$

$\dot{m}_{na}$, $\dot{m}_i$ — Na aerosol & RN release rates

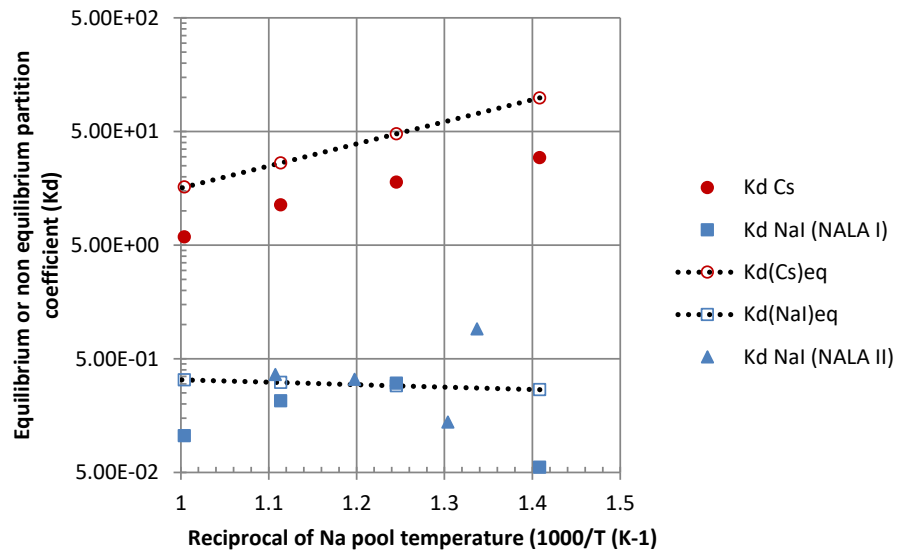
FP vaporisation from hot Na

NALA experiments

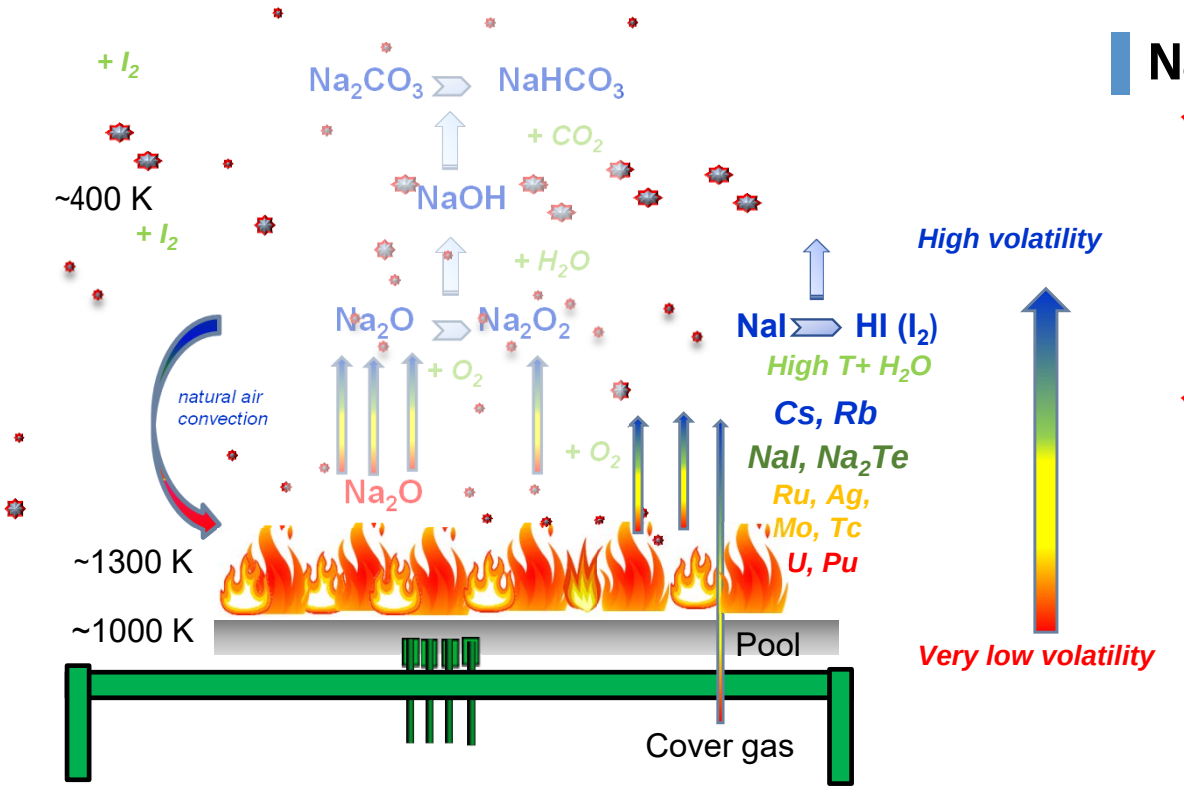
- In eq. conditions



- In non eq. conditions



FP vaporisation from Na fires



Na combustion/FP vaporization function of

- ✓ Na vaporisation then combustion: higher T(Na)
  - *Increased release of FP (function volatility..)*
  - *Enhanced release in case of concrete heating (effect of sparging gases (i.e.CO_2) on liquid MT*
- ✓ Chemical reactions within containment O_2-steam rich atmosphere
  - *Increased released of I & Ru* (NaI + H_2O → NaOH…)

$$K_c = \frac{\dot{m}_i / m_{i,pool}}{\dot{m}_{na} / m_{na,pool}}$$

$\dot{m}_{na}$, $\dot{m}_i$ — Na aerosol & RN release rates

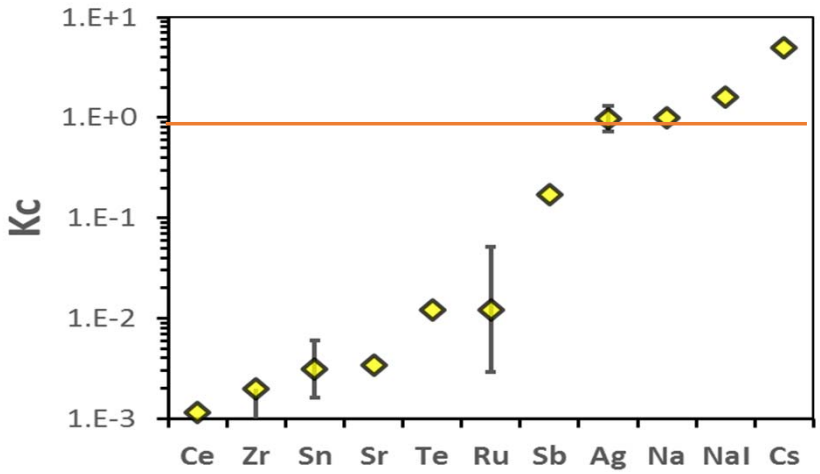
FP vaporisation from Na fires

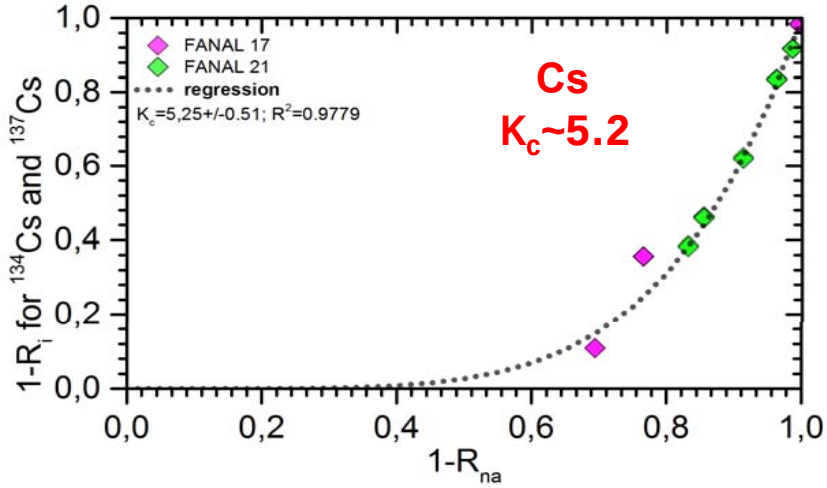
FANAL experiments

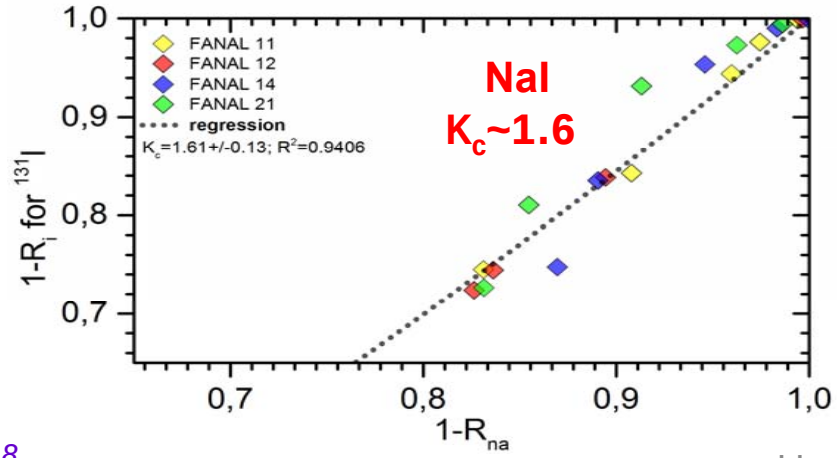
Enrichment or depletion of dissolved radionuclides *generally* linked to volatility excepted for iodine (as NaI)

- **Cs - more volatile → enrichment**
- **Ag ,Sn, Sb, Ru, Ce, Sr, Te, Zr - less volatile → depletion**
- **I (NaI) - less volatile but enrichment**

$$\left(1 - R_i(t)\right) = \left(1 - R_{na}(t)\right)^{K_c}$$

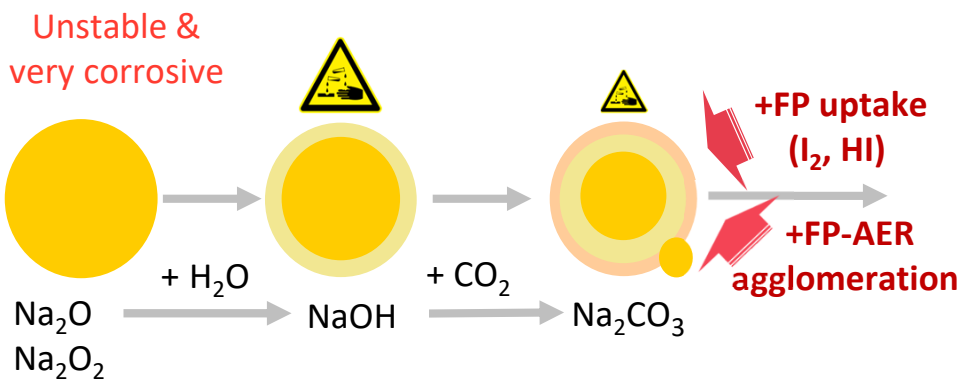


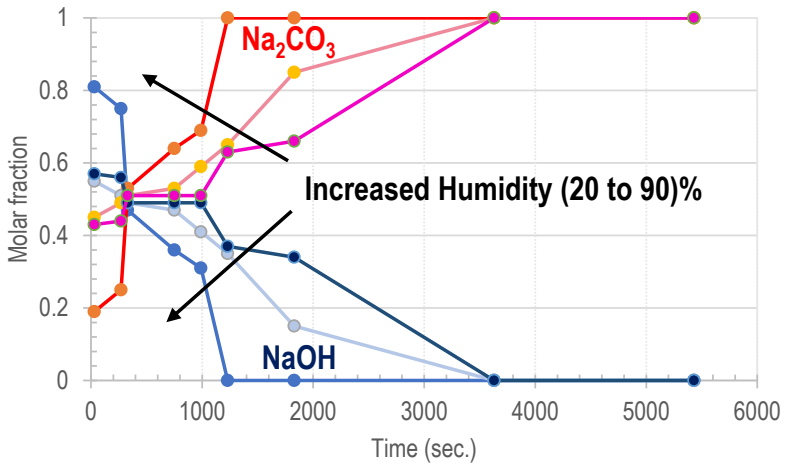




L. Lebel & al., NED 327, 2018

Na-AER behaviour





Na-AER ageing

✓ Kinetics of carbonation process enhanced in high relative humidity (90%): complete carbonation in 20mn (90% RH)/1h (20-50% RH) (*Narayanan & al., ANE 80, 2015*)

- *Higher CO_2 diffusivity in liquid core (at high RH) compared to solid core particles* (*Gilardi, IAEA-CN 2013*)

Na-AER growth

✓ Particle size increases with RH (0.9 to 2.1 µm from 20 to 90% RH) enhancing coagulation/settling processes

- *Chemical/radiological toxicity function on suspended aerosol concentration and thus on % RH*

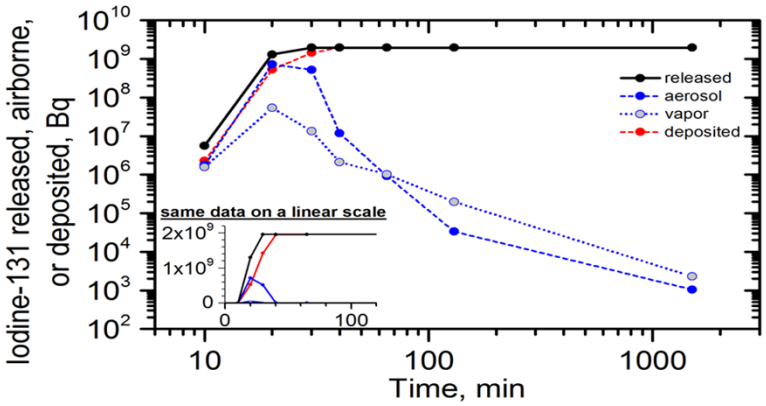
FP interaction with Na-AER

I_2 quickly reacted with Na combustion aerosols

✓ Simplified kinetic model (pseudo first order law) from experiments

$$N_{I_2}^{uptake} = \frac{q_{I_2}^0}{k^I}$$

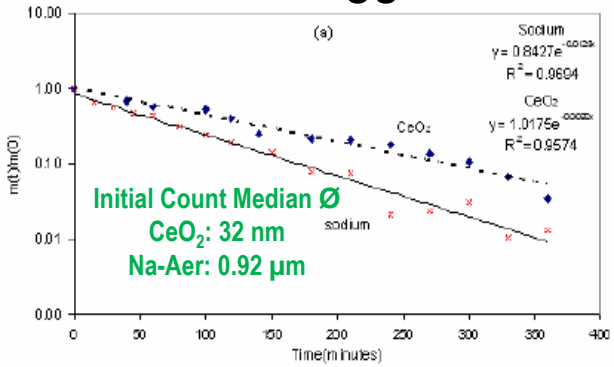
k~3 10^{-3} s^{-1}

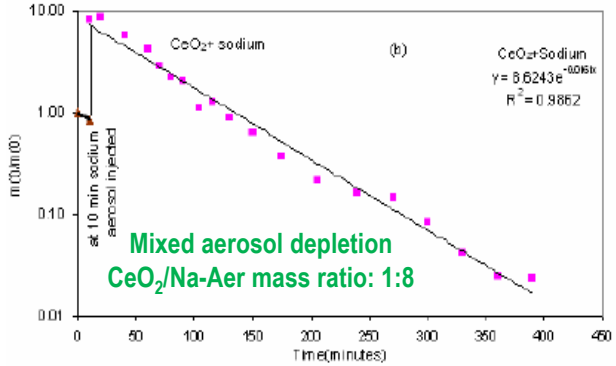


- FANAL+ FTR experiments + ab initio modelling of the adsorption isotherm

L. Lebel & al., NED 327, 2018
A. Jadon & al., J. Phys.Chem., 2018

Faster settling of FP-AER in aerosol mixtures due to co-agglomeration





- Aerosol Test Facility (ATF)

V. Subramanian & al, Nucl. Tech. 176, 2011

Main take aways

Cs release fraction is much higher than Na and I/Te from hot evaporating Na

but Cs not emitted from a Na fire as quick as from Na evaporation in a inert atm.

- ✓ Partition coefficient (K_d) in inert atmosphere ~ 4 times higher
- ✓ Impact of gas phase chemistry ? (Cs oxidation in Cs_2O)& condensation on Na-Aer?)

NaI release fraction slightly higher than Na

but only during sodium fire

- ✓ Partition coefficient in inert atmosphere ~ 3 times lower (i.e. depletion of NaI in Na-AER)
- ✓ NaI not reactive with O_2 may diffuse over flame front wo condensing onto Na-Aer flowing back to pool
- ✓ NaI thermal decomposition in the flame : **$NaI + H_2O \rightarrow NaOH + HI$; $HI + HI \rightarrow I_2 + H_2$**

Others radionuclides have a very small partitioning

- ✓ Higher partitioning of soluble RNs (except Te : Na_2Te formation?) compared to non soluble RNs (except Ru: reacts with O_2

FP interactions with Na-AER

- ✓ Strong impact of Na-AER on FP behavior: gas. iodine efficient/fast uptake, FP-AER co-agglomeration
- ✓ Na-AER concentration and Initial size distribution thus very critical for FP behaviour

Current code predictability and major needs

- **For SFRs contrarily to PWRs where several tools and established methodology available only few integral severe accident tools (ASTEC-Na, CONTAIN-LMR, SOCRAT-BN….)**
  - ✓ Missing validated models for RN partitioning in core bubble (in vessel instantaneous ST)
  - ✓ For RN partitioning between gas and condensed phases (from hot Na pools and Na fires) at the best correlations with fitted parameters are available for FP release
  - ✓ Chemical reactivity of FPs are often not taken into account (real FP mixtures, reaction between FP & Na, containment gas phase chemistry ..)

- **For SFRs appropriate to focus on the development of more mechanistic models and integral codes for in-vessel and in-containment ST modelling**
  - ✓ EU projects (JASMIN, CP-ESFR, ESFR-SMART)
  - ✓ CRP AIEA on PFBR source term evaluation

Source Terms: Na aerosols and fission products

Conclusions and pending issues

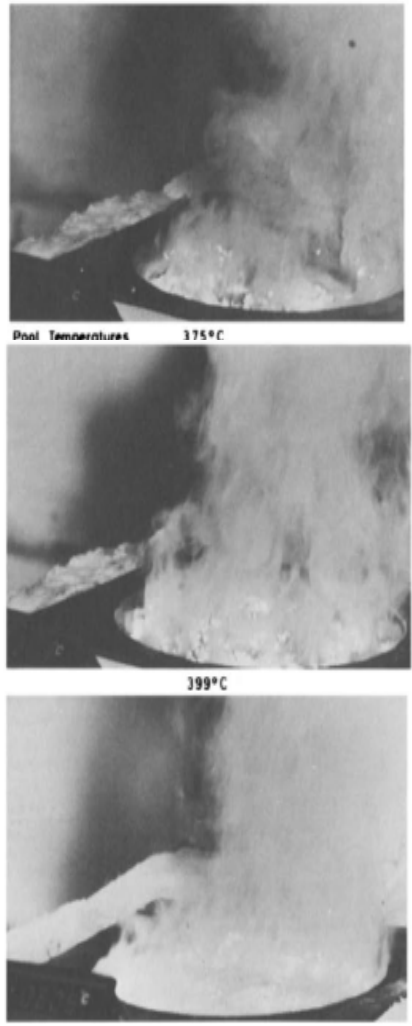
- **Few experimental tests on FP releases from hot Na pool, missing kinetics data**

- **Considerable amount of work focused on sodium fire combustion**
  - ✓ Comprehensive, large-scale experiments, difficult to replicate today
  - ✓ Only poorly investigated Na-AER ageing & FP behavior

- **Recent exp. programs allowed to improve our understanding on both Na-AER & FP during fires and to develop new models**
  - ✓ Kinetics of Na-AER ageing
  - ✓ Interactions between Na-AER and FP-AER or volatile iodine

- **Past code predictability regarding Na-AER and FP behavior generally poor (fitted parameter correlations)**

- **Analytical thermodynamical models from ab-initio calc. could greatly help to extensively study FP interactions with Na-AER**



Thank you!