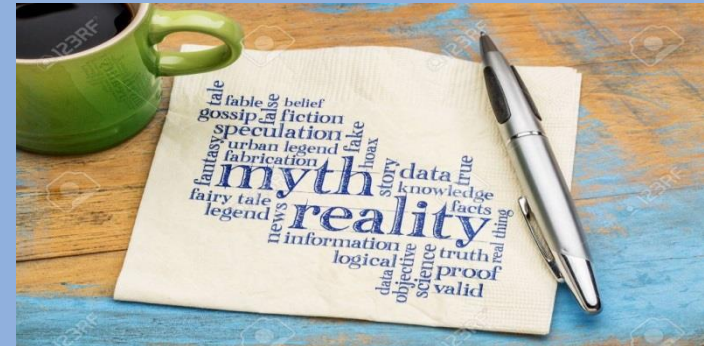# CORE DEGRADATION PARADIGM: MYTHS AND REALITY

IMPACT OF REMAINING UNCERTAINTIES ON SAFETY EVALUATION
PERSPECTIVES FOR ACCIDENT TOLERANT FUELS

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MODELLING IN NUCLEAR SCIENCE AND
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A lot has been done on the code validation as evidenced by the conclusions of the NUGENIA/CORSOAR Project (Haste et al., 2018)

- OECD/CSNI TMI-2 (2015) exercise and benchmark OECD/CSNI/ISP-46 (2013) on Phébus-FPT1 and NUGENIA/SARNET/QUENCH-11 have led to an evaluation the quality of the codes for PWR transients

- Results are in good agreement, with the calculated states of the core and the primary circuit being very similar; the main discrepancies concern the void fraction in the core, but these are considered to be acceptable

- For the degradation phase, up to core reflooding, results show a rather good agreement between the different codes for integral parameters such as total H_2 production and mass of molten materials

- The variability in these results is much lower than the results obtained in a benchmark exercise performed 25 years ago, showing a significant improvement of the codes in the last two decades
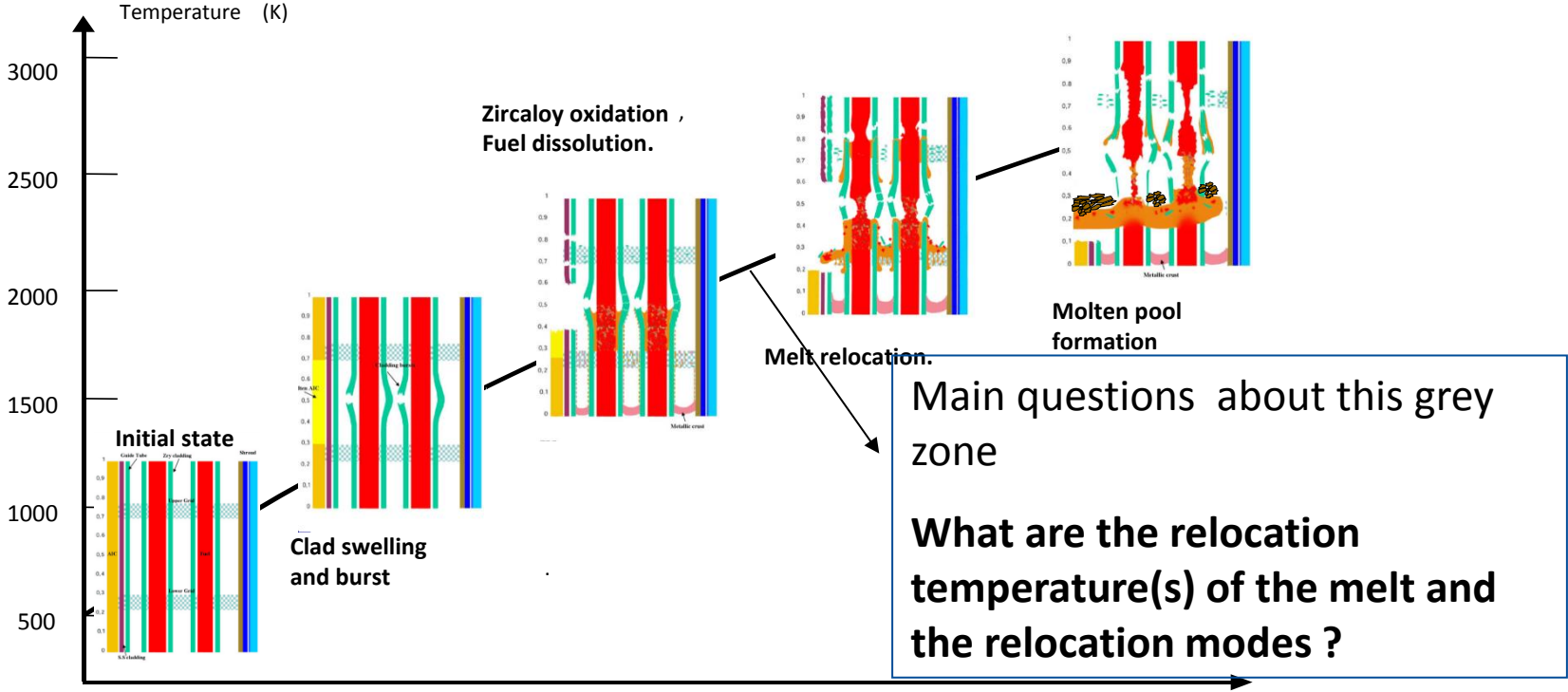
But some work remains to be done …

- OECD/BSAF benchmark on Fukushima Daiichi NPP Unit 1(Nagase et al. ,2016)
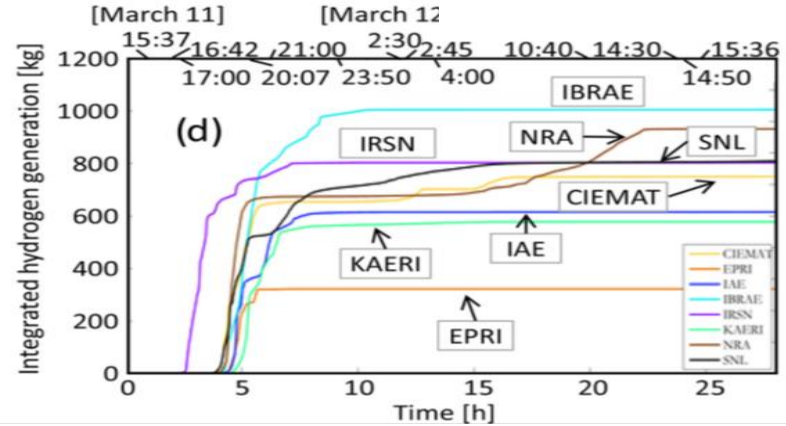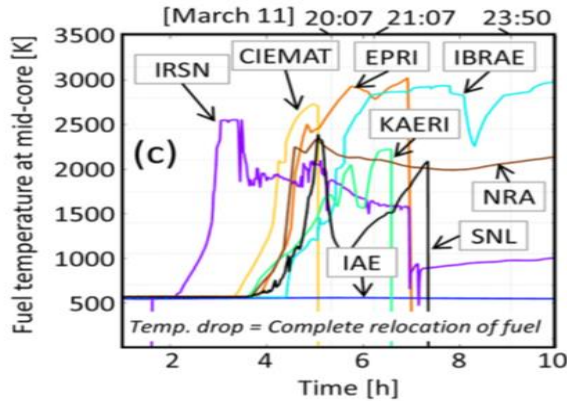
- Codes diverge as soon as the core loses its geometry, differences in modelling lead to significant differences for

  - H_2 (from 300 to 1000 kg)

  - Max. temperature reached in the vessel (from 2000° to 3000°C)

  - %. solid/liquid

Transition from the intact to the degraded geometries : a grey zone

Temperature (K)

Zircaloy oxidation,
Fuel dissolution.

Molten pool
formation

Melt relocation.

Initial state

Clad swelling
and burst

Main questions about this grey zone

What are the relocation temperature(s) of the melt and the relocation modes ?

Myths or reality about this grey zone

- **Relocation mode**

  - solid debris slumping/progressive relocation of melt ?

  - what is the impact of relocation mode on the late phase of the accident ?

- **Relocation temperature**

  - many experimental observations in **tests of PWR geometries** tend to indicate that temperature of melt relocation was much lower that UO_2-ZrO_2 eutectics (2800 K), about 2500-2600 K

  - some codes use this reduced temperature as input parameter in reactor calculations

  - is it justified whatever the transient ?

  - does this reduction correspond to "not modelled" interactions in codes ?

  - **BWR/PWR the same reality ?**

Relocation mode : solid slumping/progressive? (1/2)

- Highly dependent on the criterion of cladding failure (Veshchunov et al., 2013)

- Most widely assumptions used in the codes → failure at a given temperature (between 2100 and 2300°C) or for a given ZrO_2 thickness

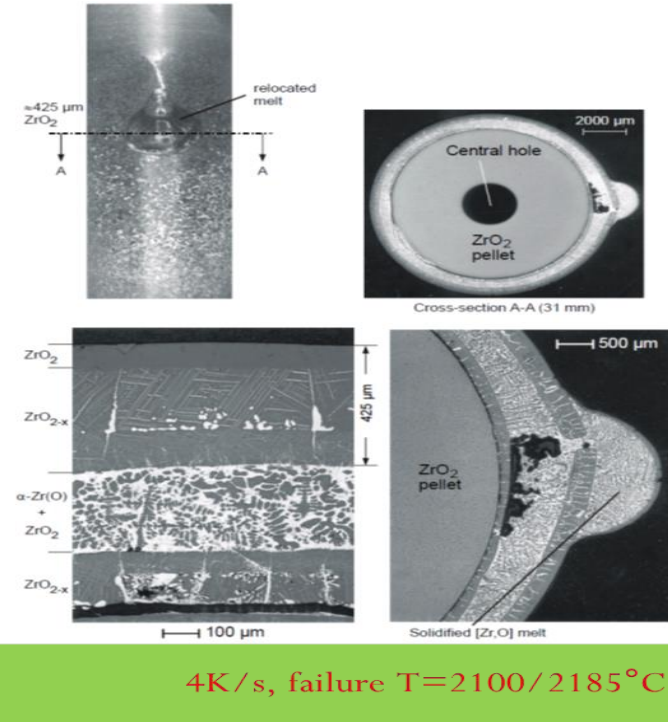- Not consistent with separate effect tests (Hofmann et al., 1999, EC CIT

Relocation mode : solid slumping/progressive? (2/2)

8K/s, failure T = 2030°C

4K/s, failure T = 2100/2185°C
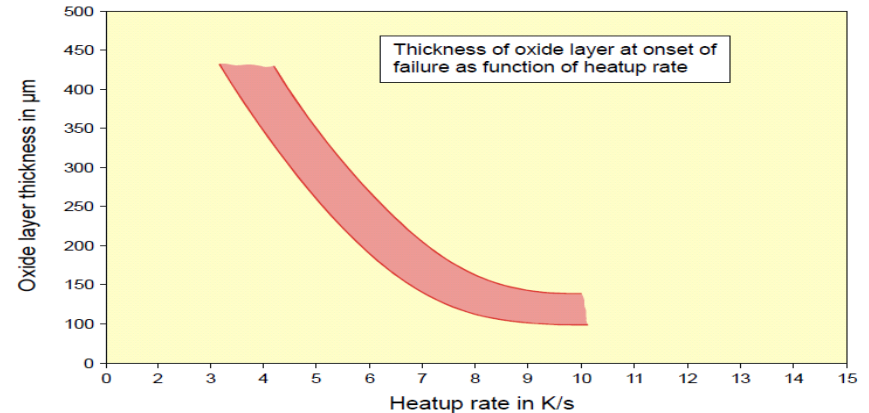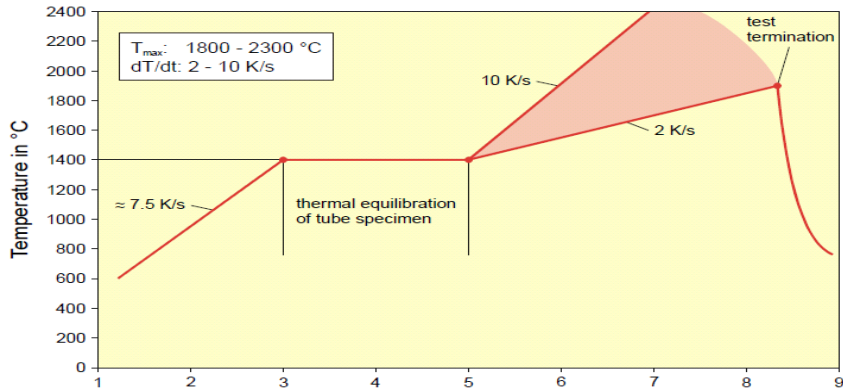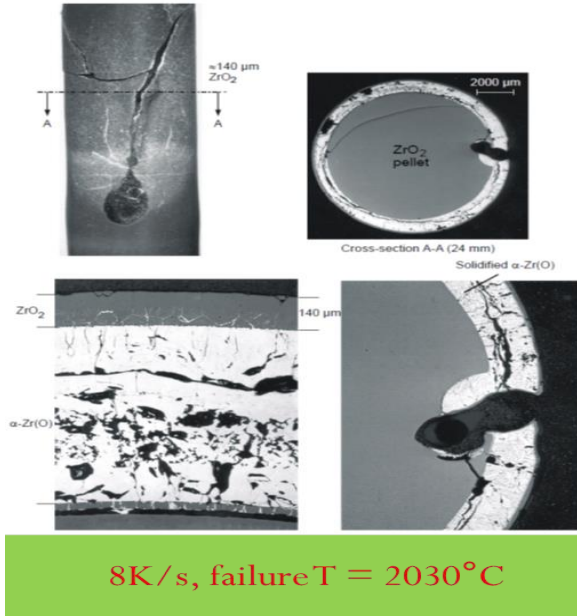
- Earlier failure, thin oxide layer (140 mm)
- Higher probability of local non-fully oxidised molten pools
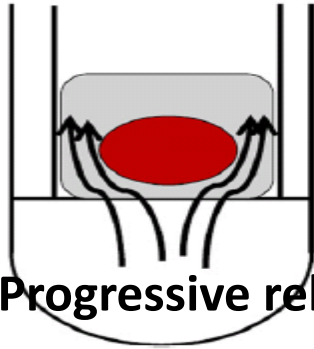
→ More probable progressive relocation

- Later failure, thick oxide layer (425 mm)
- Less probability of formation of local non-fully oxidised pools
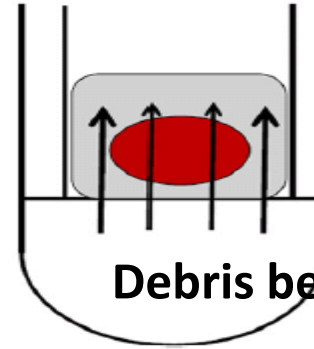
→ More probable debris bed slumping

Impact of relocation mode on the late phase (1/2) : 2 main situations

Progressive relocation

- Molten materials dominant in the lower plenum

- By-pass of steam,

- Less exchange and oxidation

- Higher temperatures reached in the pool

→ Relocation mode rather considered in ASTEC and MAAP

Debris bed formation

- Solid materials dominant

- Better exchange with steam through debris bed porosity

- Stronger debris oxidation

- Lower temperatures in the debris bed

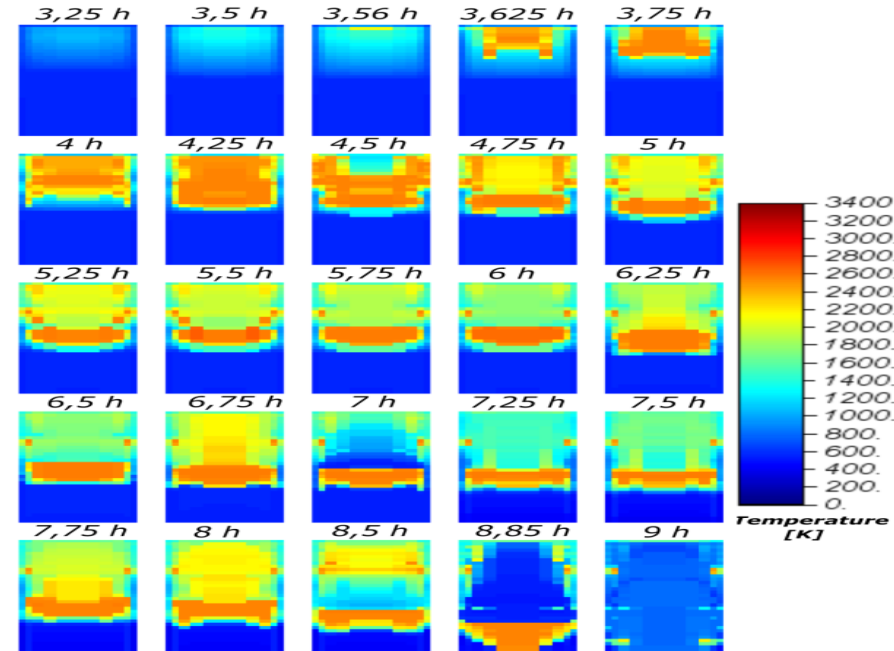→ Relocation mode rather considered in MELCOR

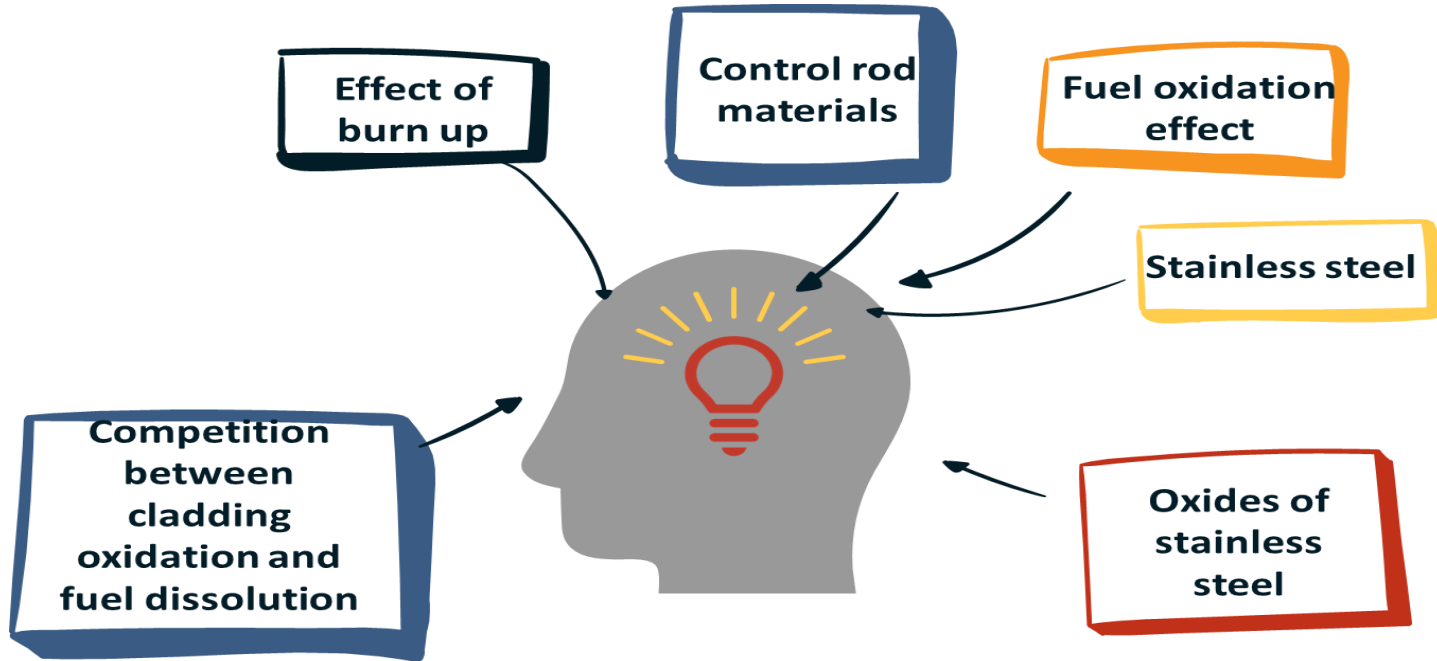# Impact of relocation mode on the late phase (2/2) : Fukushima Daichi U1

MELCOR
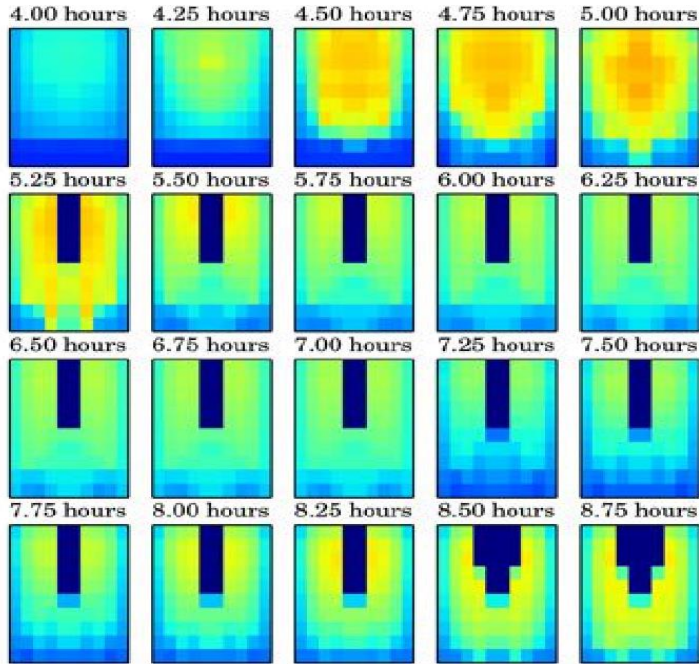
ASTEC

Faster relocation in MELCOR

Higher temperature in ASTEC

Temperature of melt relocation 2500 K- acceptable but why ?

Temperature of melt relocation 2500 K- OK but why ? (1/2)

- Impact of **metallic** control rod materials **(evoked in CORA tests, Hofmann et al., 1994)**

  - Phebus FP : no significant action of Ag-Cd-In and SS on fuel degradation, agrees with TMI-2 observations

  - No thermodynamic reason that metallic melts reduce UO2 and ZrO2

- Impact of **oxide components** of steel (mentioned in TMI-2, Strain et al., 1989)

  - Can reduce very significantly the UO2/ZrO2 melting points

  - PWR second order effect (as shown in Phébus FP)

  - May be different in BWR where steel amount is higher

- Impact of **burn-up** (mentioned for VERCORS, Pontillon et al, 2005)

  - No difference between Phebus FPT0 (fresh fuel) and FPT1 (32 GWj/tU)

  - No real change of chemistry with FP regarding the degradation

MYTH

REALITY MYTH

MYTH

- Impact of **oxygen potential** (**evoked in VERCORS**)

  - No structural materials (as stainless steel or CR materials) in the VERCORS tests

  - No metallic zircaloy since the cladding is fully oxidized before reaching high T

  - Melting temperature of UO2 can be strongly reduced as function of the oxygen potential

- Impact of **metallic melt produced after the cladding failure**

  - Amount of sub-oxidised melts increased in FPT0 vs FPT2

  - Lower temperature for fuel rod relocation in FPT0 (2570 K) vs FPT2 (2650 K)

- **So what to do ?**

  - No reason to use this temperature of 2500 K as an input parameter in the reactor calculations

  - Cladding failure mechanism to be better modelled

  - Thermochemistry of O-U-Zr system (NUCLEA database) must be taken into account in the degradation models

Application to TMI-2 accident (Drai et al. 2018)-ASTEC calculation-Sensitivity study to the melting temperature of UO2 on the H2 production

$T^m(UO_2)$=2500 K
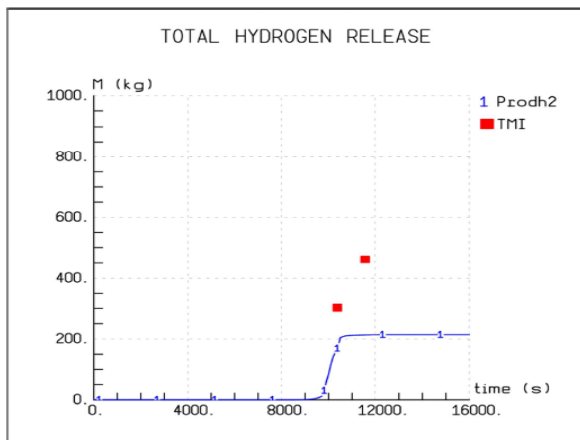
$T^m(UO_2)$ standard
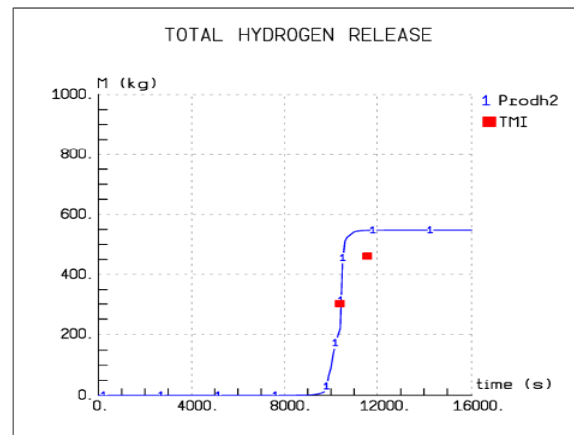
- Significant impact of melting temperature on the H2 production and the degradation (not shown)

- With higher T^m, fuel rods and claddings keep their 'intact' geometry longer, staying available for oxidation,

PWR/BWR

- Most of results obtained in two last decades were for PWR geometry

- For BWR, interactions during the early phase can promote the degradation, not the case for PWR

- B_2O_3 formed from B_4C oxidation can spread towards surrounding fuels rods

- Eutectic reactions with UO_2 and ZrO_2 at low temperatures (EC ENTHALPY)

- Observed in CORA W2 and Phebus FPT3
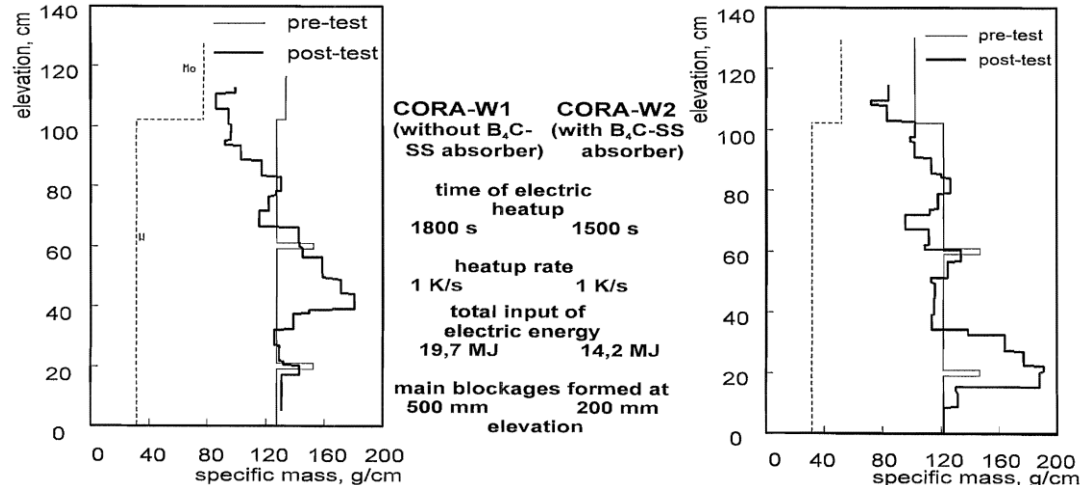
Conclusions –Remaining open questions (1/2)

- Difference in corium melt relocation highly impacts final condition of debris and structure materials, which includes redistribution/mechanical properties of debris, redistribution of major constituents in debris, degree of oxidation, degree of failure of structure material, etc.

- 2 schematic core degradation schemes.
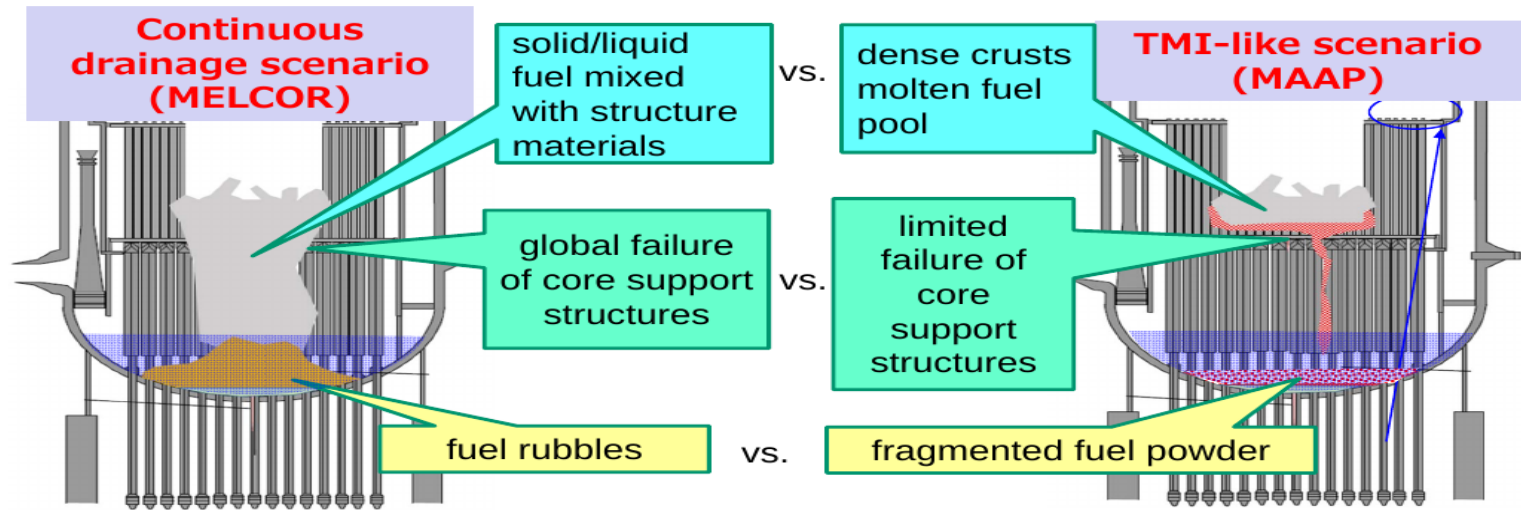
Figure provided by M. Kurata (JAEA)

Conclusions –Remaining open questions (2/2)

- What are the relocation processes of degraded materials **before fuel melting** ?

- What is the gas **permeability** through the degraded materials ?

- What the mechanical/thermal resistance of the crust supporting the molten pool ?

- What is the failure mode of the core **support structure** (strongly impacts the amount of stainless steel inside the molten pool ?

- What is the role of the **grids** in the relocation process ?

Relevant & unique phenomena for near term ATF under severe accident conditions & available experimental and analytic studies

▌ **Cladding** : FeCrAl , SiC and Cr-coated zirconium alloys

▌ **Fuel** : Cr_2O_3-doped UO_2 fuel

▌ **Evaluation of the impact of these new materials on**

   – Oxidation   **SiC, FeCrAl, Cr-coated Zr**

   – Fission product release and transport   **Cr_2O_3-doped UO_2 fuel**

   – Interaction cladding/fuel   **SiC, FeCrAl, Cr-coated Zr**

   – Molten pool behavior (in- and ex-vessel)   **Cr-coated Zr**

ATF claddings (1/3)

ZR-ALLOYS REFERENCE

▌ **Oxidation kinetics**

 – Parabolic oxidation

 – Faster kinetics when ZrO_2 is cubic, however ZrO_2 melts only at very high temperature

▌ **Physico-chemical interaction with fuel**

 – Small reduction of the melting temperature of the UO_2-ZrO_2 mixtures in comparison with fuel

COATED ZR-ALLOYS

▌ Improvement of oxidation resistance, hydrogen uptake, mechanical properties **up to 1300°C (in comparison with Zry-4)**

▌ Oxidation behaviour (*Steinbruck et al., 2019*) apparently **worse** than for non-coated cladding at 1450°C

▌ Interaction with UO_2 fuel : **no expected difference** with reference cladding material (numerous data from Hofmann, Olander, Hayward …)

▌ Cr coating vaporization under $CrO_2(OH)_2$ or CrO_2OH

FECRAL CLADDING

▌ Very few data above the melting point of the cladding → expert judgement based on Fe

▌ Oxidation kinetics of **Fe** above 1600°C

   – FeOx is **molten**

   – **Catastrophic oxidation** (molten oxides) (*P. Kosfstad 1988*) only limited by steam diffusion in gas (steam starvation)

   – Heat of reaction by cladding length lower than for Zry-4

▌ Cr and Al vaporization under $CrO_2(OH)_2$ or CrO_2OH and $Al(OH)_3$ (resp.)

▌ Physico-chemical interaction with fuel

   – **Strong reduction** of the melting temperature of UO_2 fuel by interaction with FeO (*Bechta et al., 2007*)

   – Possible intergranular attack leading to wet sand relocation (*TMI-2, Strain et al. 1989*)

   – Preliminary tests from *Sakamoto et al. 2019*, in agreement with this assumption

FeCrAl-ODS cladding   UO₂ pellet   Oxidized cladding   UO₂ pellet

Before heating test   After heating test

IRSN

ATF claddings (3/3)
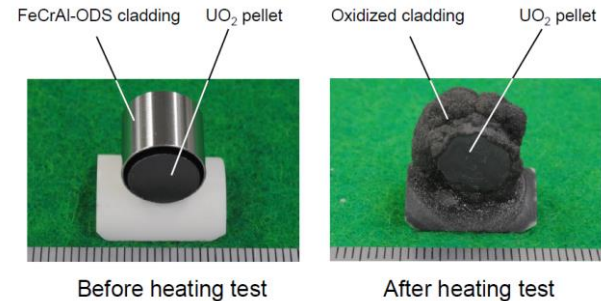
SIC CLADDING

▌ Oxidation kinetics of SiC below 1700°C

  – Oxidation by steam, more than 2.5 times more explosive gases ($CO+H_2$) *produced per cladding length unit at thermodynamic equilibrium, assuming same thickness of Zry and SiC*

▌ Oxidation kinetics of SiC above 1700°C

  – Molten SiO_2 looses its protective effect

  – **Catastrophic oxidation** (molten oxides) (*P. Kosfstad 1988*), only limited by steam diffusion in gas (steam starvation)

  – Vaporisation of SiO_2 into $Si(OH_4)$ or $SiO(OH_2)$

▌ Physico-chemical interaction with fuel

  – **Strong reduction** of the melting temperature of UO_2 fuel by interaction with SiO_2 (*Ball et al. 1993*)

IRSN

Appendix:

Impact of ATF on FP release :
- Cr$_2$O$_3$-doped UO$_2$
- Effect of aerosols from cladding

Cr$_2$O$_3$-doped UO$_2$ (1/2)

▍ **Expected effects on microstructure and impact on FP release**
- grain already "big" at t$_0$, grain growth rather expected as a second-order effect.

   → greater ease for percolating gases since the "grain joint" surface of percolation is reduced.

- formation of the RIM zone: expected decrease in the concentrations of interstitial uranium in the irradiation cascade

(*Guo et al. 2017*)

   → should delay the appearance of the RIM zone compared to undoped UO$_2$

▍ **Expected effects on FP diffusion coefficients**
- Uranium vacancies = stable positions in UO$_2$ for most of the FP (Cs, Kr, Xe, Ba, Ce, Ru, Rb, Zr, Y) (*Grimes et al. 1991, Crocombette 2002, Busker et al. 2000, Petit et al. 1999, Brillant et al. 2008*)

   → substitutional mechanism for FP diffusion usually assumed

- **Cr doping** eases the formation of U vacancies (Guo et al., 2017)

   → could contribute to an increase of the FP diffusion coefficient (**still speculative,** uranium vacancy migration energies

remain to be determined)

Cr$_2$O$_3$-doped UO$_2$ (2/2)

ANTICIPATED EFFECTS ON CR$_2$O$_3$-UO$_2$ DOPED FUEL

▎ **Expected effects on fuel and FP chemistry**

  – Cs may form **stable chromates** in doped fuel

  – Competition with **chromium uranate** (in hyperstoichiometric fuel UO$_{2+x}$) formed from Cr^{3+} and U^{5+} (*Cooper et al. 2013*)

$$UO_2 + 1/2\ Cr_2O_3 + 1/4\ O_2 = UCrO_4$$
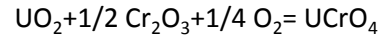
  – **FP partition** between UO$_{2+x}$ and chromium uranate estimated by ab-initio calculations (*Cooper et al. 2013*)

  ▪ Small tetravalent FPs (Mo^{4+}, Zr^{4+}, Ru^{4+}) preferentially in CrUO$_4$// bigger ones (Ce^{4+}) and actinides (Am^{4+}, Pu^{4+}, Np^{4+}) in UO$_{2+x}$

  ▪ Trivalent FPs (lanthanides) are calculated to remain in UO$_{2+x}$

  ▪ Divalent FPs (Sr^{2+}, Ba^{2+}) are calculated to remain in UO$_{2+x}$

  → impact on the Cs release through the modification of the Mo chemistry

▎ **Key points for SA codes to be developed**

  – Mo and Cs chemistry modified by chromium presence and consequence on iodine partition (aerosol/gas) and transport

FP transport in the RCS

CR- COATED ZR

❚ Cr vaporization under $CrO_2(OH)_2$ or CrO_2OH

❚ Impact on cesium chemistry (Cs chromates) and then on iodine one

SIC

❚ SiO_2 vaporization under $Si(OH_4)$ or $SiO(OH_2)$ (high steam flow rate)

❚ Impact on cesium chemistry (Cs silicates) and then on iodine one

FECRAL

❚ Cr and Al vaporization under $CrO_2(OH)_2$ or CrO_2OH and $Al(OH_3)$ (resp.)

❚ Impact on cesium chemistry (Cs chromates) and then on iodine one