

DE LA RECHERCHE À L'INDUSTRIE



Some pending issues in nuclear fuel and in-core materials development for LWRs : experimental support for modeling and simulation

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January 9th , 2018

OECD/NEA Workshop
“Enhancing experimental support for
advancements in Nuclear fuels and materials”

Session 2 : “Advances in nuclear science : promising fuel
technologies, modelling and validation methods, and
associated experimental challenges”

**MANUFACTURING
LABORATORY PROCESSES**

**INTEGRAL, SEMI-INTEGRAL,
ANALYTICAL IRRADIATIONS**

**OUT OF PILE ANALYTICAL TESTS
ON NON IRRADIATED AND
IRRADIATED MATERIALS**

**PRE/POST IRRADIATIONS/TESTS
EXAMINATIONS**

EXPERIMENTS

**MODELLING
SIMULATION**

**MEASUREMENT
/CALCULATION
COMPARISON**

**Scientific
Understanding**

MULTISCALE

MULTI-D

MULTIPHYSICS

**IMPROVE CODE
PREDICTIVITY
IN NORMAL AND
OFF NORMAL
CONDITIONS**

- Discriminate/quantify mechanisms to improve models
- Get basic data as input for models/codes
- Validation of codes/models

- Select and prepare « smart » experiments
- Prediction of expected results
- Experiment interpretation

SUPPORT SAFETY ANALYSIS AND QUALIFICATION OF FUEL

**DESIGN OPTIMIZED or NEW MATERIALS/FUELS
life extension/maneuvrability, safety improvement**

Part 1 : Fuel, needs of dedicated in-pile, out of pile and PIE experiments (three example studies):

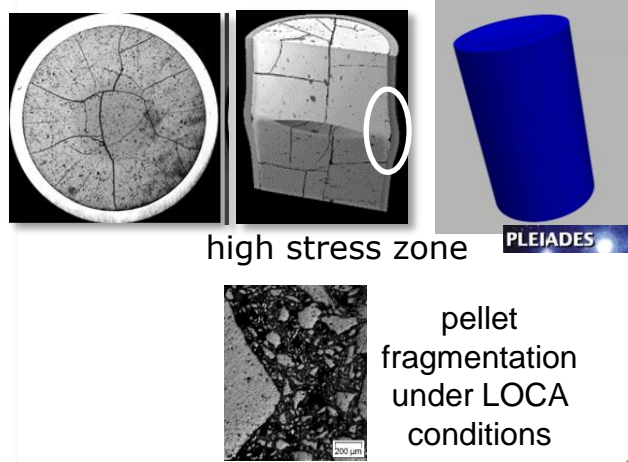
1- **Analytical irradiation** to quantify **thermomechanical clad load** mechanisms during LWR transients

2- **Out of pile analytical tests** on irradiated fuels to identify **fragmentation** mechanisms during LOCA transient

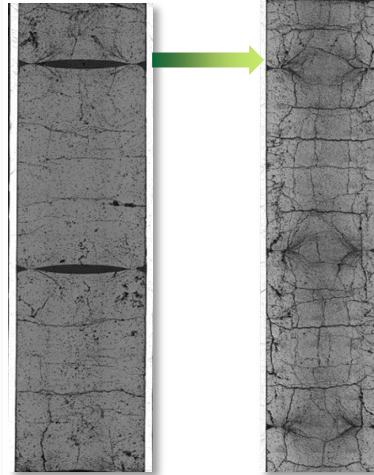
3- **Potential instrumented irradiations** with **tailored environment** : oxygen partial pressure

Fuel thermal expansion, cracking, fragmentation

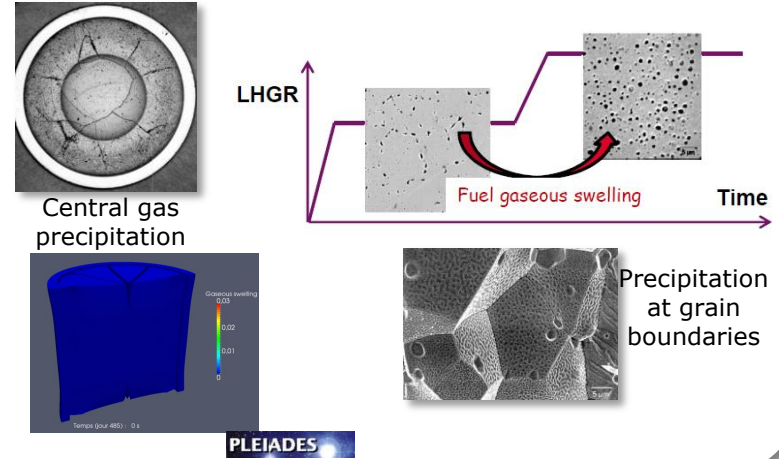
Power transient



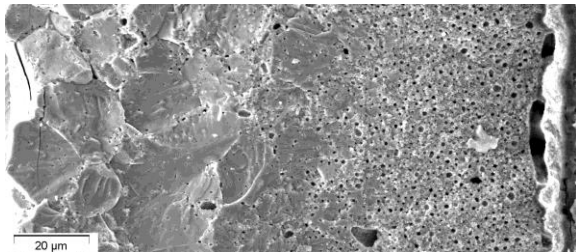
Fuel creep



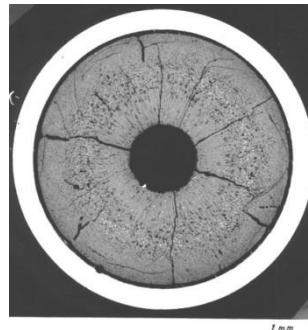
Fuel gaseous precipitation and swelling, fission gas release (FGR)



Microstructural Restructuration

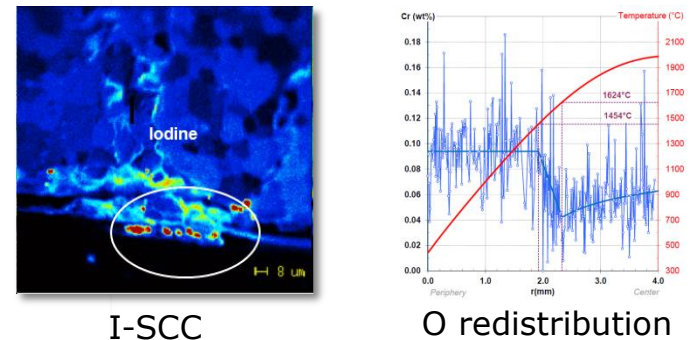


High Burn-up Structure in PWR



Columnar grains in SFR

Thermo-chemistry and matter redistribution

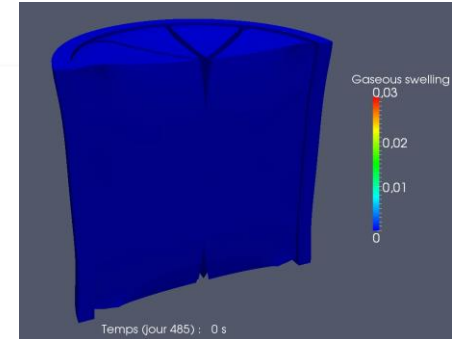


I-SCC

O redistribution

Discriminate / quantify mechanisms

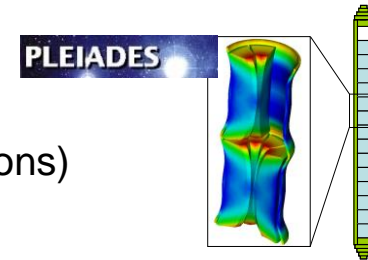
- During any type of power transients, with focus on phenomena inducing a moderate to high load on the clad:
 - ↳ Fuel thermal expansion
 - ↳ Fuel gaseous swelling
 - ↳ Fission gas release
 - ↳ Fuel volume change at melting



Modeling/Simulation

Enlarge current databases for code validation (including codes benchmark and transposition to reactor conditions)

- Pre-calculation of the rod behaviour (test conditions / instrumentation specifications)
- Prediction of expected results:
 - ↳ Fuel central T vs. LHGR, Margin to fuel central melting, Melted volume fraction at maximum LHR
 - ↳ FG distribution, migration and release
 - ↳ Expected clad deformation in hot/cold conditions
- Post-test comparison calculation vs. experimental results



Application to industrial needs

- Results transposition for addressing issues facilitating NPP flexible operation, fuel manufacturing and procurement processes, preserving safety and clad reliability
- Improve **quantification of available margins on current fuel management constraints**

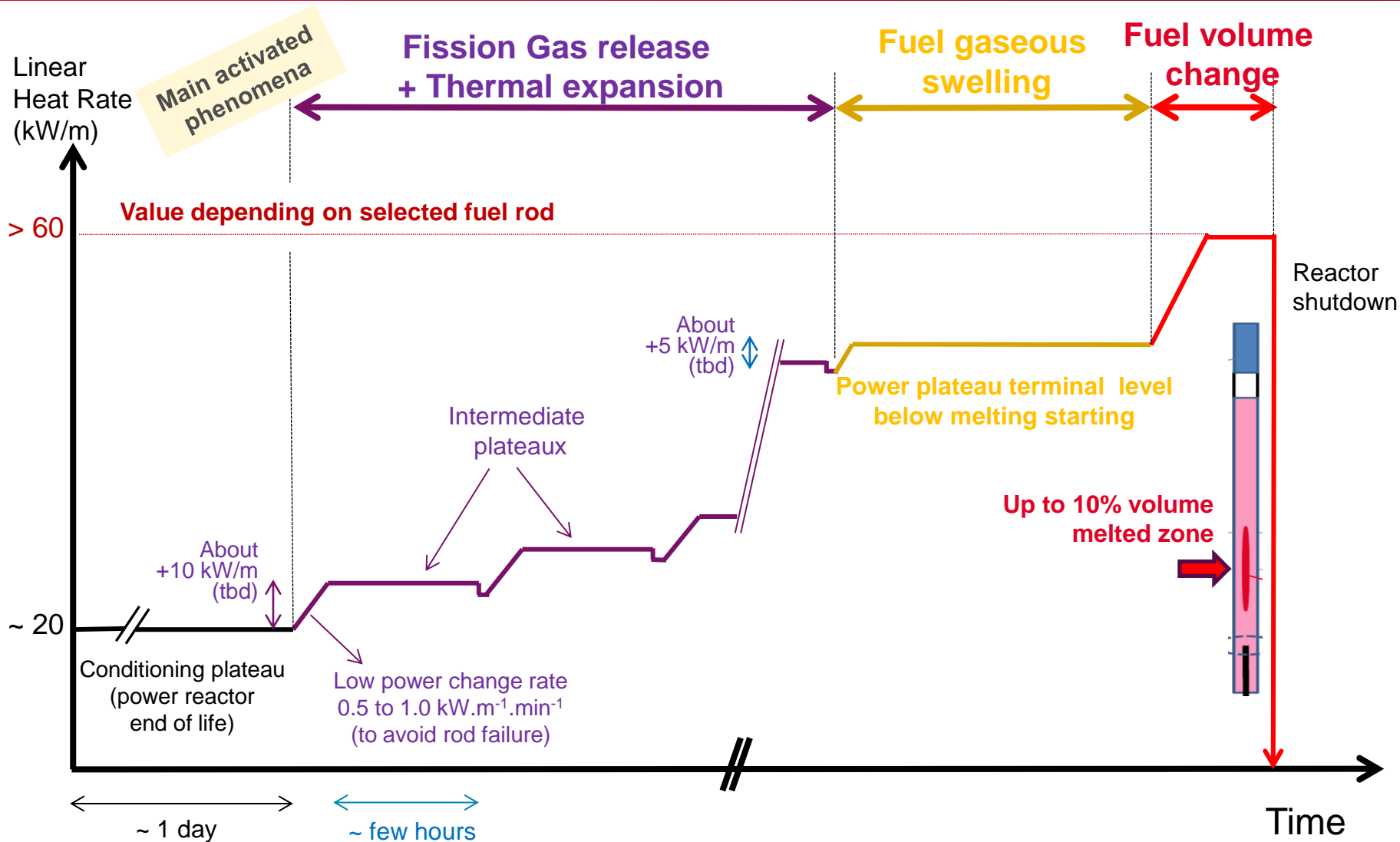
Main experimental specification of such an irradiation

- To work with **high burn-up fuel** (40-60 GWd/tu)
 - ❖ Large available quantity of fission gases → **Significant clad deformation**

- To go **step by step to high values** of LHGR (up to > 60 kW/m)
 - ❖ Discrimination of successive phenomena
 - ❖ Prohibit clad failure occurrence (“slow ramp”)
 - ❖ Reach **incipient fuel melting** in the central part of the fuel pellet at max T flux plane

- **On-line measurement** of interest parameters
 - ❖ Internal gas pressure (FGR kinetics)
 - ❖ Fuel central temperature
 - ❖ Clad outer diameter (versus device possibility and state-of-the-art instrumentation)

- **Full pre- and post-test rod status comparison** (hot cell)
 - ❖ Metrology, Gamma scanning, Metallography, Micro-analyses...

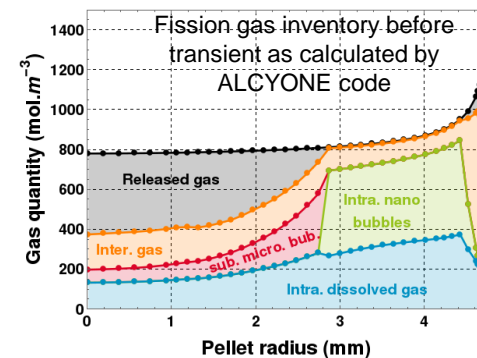


Discriminate / quantify mechanisms

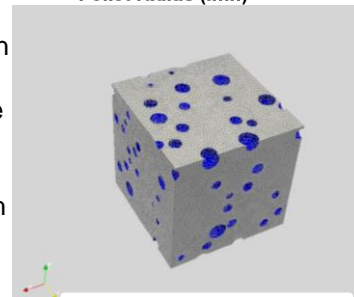
- Responsible for **Fragmentation, Relocation & Dispersal** of fuel during a LOCA transient
- Fuel initial state : precipitation bubbles, inter/intra granular distribution, high burn up structure...
- Rod environment : temperature, external pressure, transient rate, ...

Modeling/Simulation

- CEA **Fuel performance code ALCYONE** used for LOCA simulations
- Taking into account an **fuel inter-granular failure criteria**
- Code validated on both separated effects test and integral experiments : failure time, cladding strain and fission gas release are satisfactorily assessed
- Some pending issues : **Fragmentation modelling** (initiation, fragment size and localization) in relation to fission gas distribution and fuel microstructure; 3D modelling of **fragments ejection** following first burst; gas axial flow model



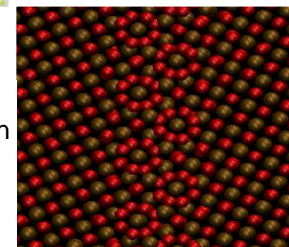
Fuel fragmentation in HBS at microscopic scale simulated with a full 3D Finite Element approach (EDF-CEA PhD thesis)



Application to industrial needs

- Justify that **safety requirements** are respected
- Propose fuel solutions** to limit fragmentation during LOCA

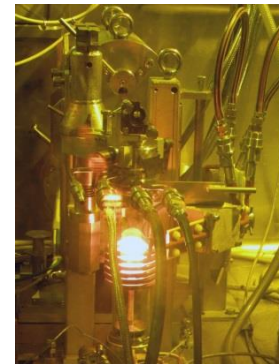
Atomistic simulation of grain boundaries mechanical properties



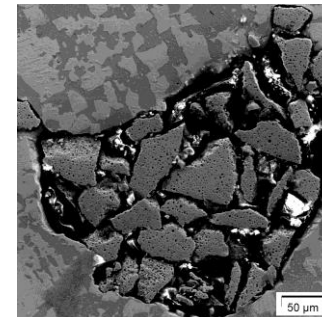
High Temperature /high pressure annealing test devices

such as *MERARG* and *MEXIICO* in the *LECA/STAR* facility

- Temperature range for LOCA : 600-1200°C
- Fragmentation versus fission gas precipitation and release
- **MERARG** : atm pressure, T up to 2600 °C, controlled pO_2 , on-line fission gas release measurements ; **MEXIICO**: controlled Temp. up to 1600°C & pressure up to 1600 bar, impact of pressure/de-pressurization on fragmentation
- Combined with fuel **micro/nano characterizations** (SEM FIB, TEM, EPMA, SIMS...) to connect microstructure, release and fragmentation ; On pellet size samples, fuels discs, core samples to probe various radial positions



MERARG annealing device



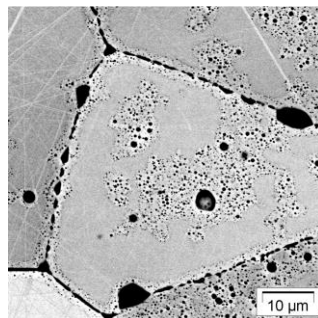
Heating of NFIR high burn-up fuel discs

Towards EATF fuels

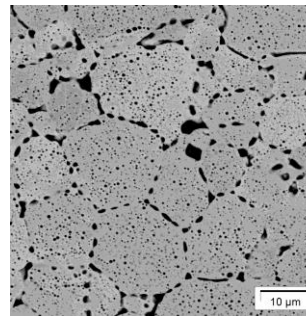
Potential way to reduce fragmentation in case of LOCA:

Large grain fuels which seems to have a local high resistance to intra-granular bubble formation (NFIR program)

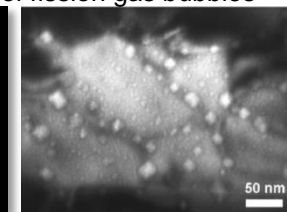
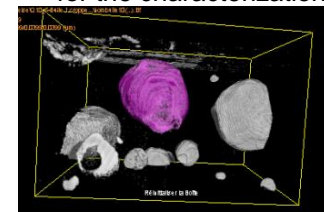
Specific fabrication
Large Grain fuel
42 GWd/tHM



Standard Grain fuel
44 GWd/tHM

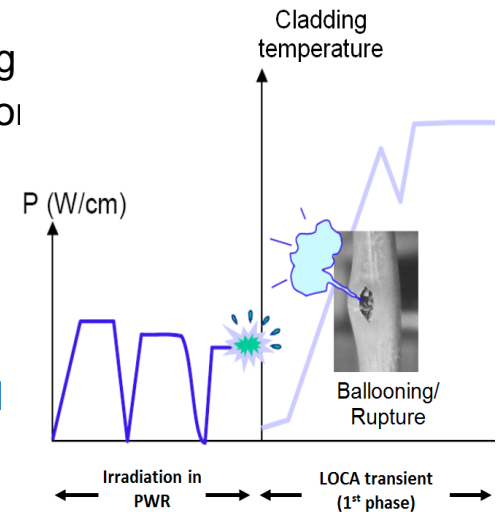


SEM FIB and TEM powerful tools
for the characterization of fission gas bubbles



VERDON-LOCA a promising tool

- CEA, with the support of EDF and AREVA, is currently developing the VERDON-LOCA test device (STAR facility)
- To get a **detailed characterization** of all the events of interest regarding “**Fuel Fragmentation, Relocation & Dispersal**” phenomena resulting from cladding ballooning and rupture during the first phase of a LOCA transient
- Originality : mini-rods, **gamma measurement capacities** to get access on-line to the **kinetics of fission gas release** and in-situ to the **potential (axial) relocation of fuel fragments**
- Objective : improve the understanding and validate the associated modeling for LOCA simulations (for CEA in the ALCYONE code)
- Complementarity with future in-pile irradiation LOCA-Test using LORELEI loop in JHR

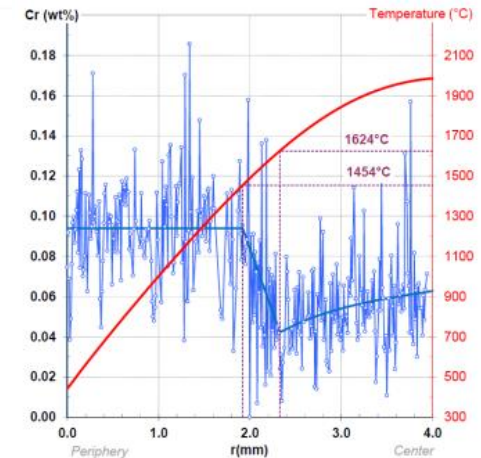


Oxygen potential in fuels

- Most of fuel properties are controlled by oxygen potential (or oxygen partial pressure) which controls point defect concentration
- E.g. Sintering, mechanical (creep), thermal (conductivity), chemical (corrosion, interactions) properties, microstructure evolution under irradiation, FP diffusion and release,...

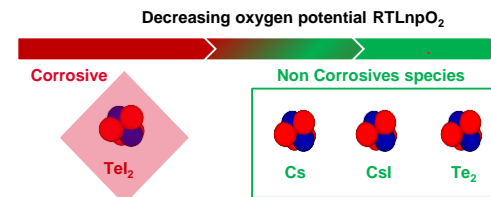
Key-role of Oxygen potential during power ramps

- Large temperature gradient implies radial oxygen redistribution in fuel
- This results in strong oxido-reductive perturbations inside the pellet -> impacts speciation of volatile FP involved in clad SCC
- Such redistribution well evidenced in Cr-doped fuel, Cr acting as oxido-reduction tracer (Cr solubility <-> Oxygen potential)
- Thermochemistry of irradiated fuel and oxygen thermal-migration to be taken into account in fuel codes (ALCYONE)



Next step: Mastering oxygen potential during irradiation

- Innovative fuel concepts for a better resistance to PCI
- Redox buffer additives to control oxygen partial pressure and speciation of volatile FP (inert form limiting clad corrosion)
- Additives increasing the oxygen redistribution kinetics



Part 2 : Fuel cladding and In-core materials, two examples studies:

1- Behavior of **in-core internal** baffles materials of LWRs under neutron irradiation (304-SA stainless steel) : neutron spectrum/flux effects

2- Enhanced **Accident Tolerant Fuel claddings** R&D and some pending issues needing further dedicated MTRs experiments & PIE

- short / mid term R&D : Cr coated Zr based claddings

- breakthrough / long term R&D: SiC-SiC sandwich claddings



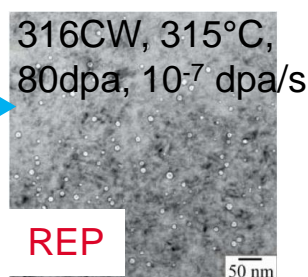
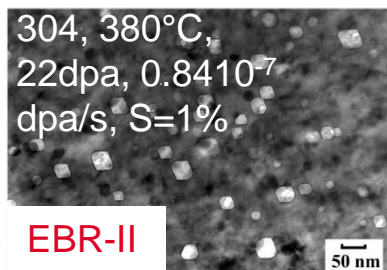
Patents

- LWR internal structures support the core, fix fuel assemblies in position, distribute coolant flow, support and protect control rods, shield reactor pressure vessel
- Ageing issue: Structural integrity (SI) degrades under lifetime doses of 10 dpa (core barrel) – 100 dpa (baffle-former bolts)
- No surveillance programs - Ageing monitoring programs for internals are based on PIE and MTR irradiations

Identified Gaps and needs

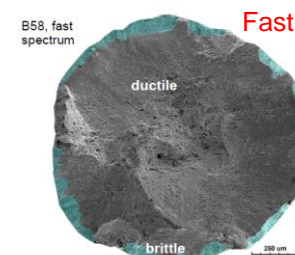
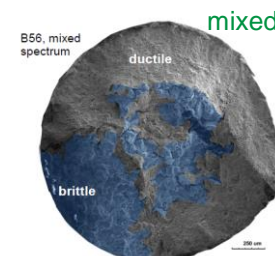
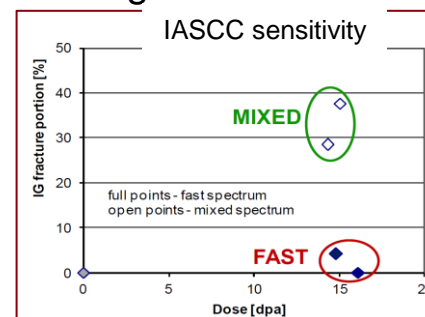
- There is no simple transferability of swelling and IASCC data (dose, T_{irr}) from FRs or MTRs to LWRs

- Need **well-characterised experimental data** to understand the effects of irradiation conditions
- Need **modelling tools** to describe microstructure evolution as function of irradiation conditions

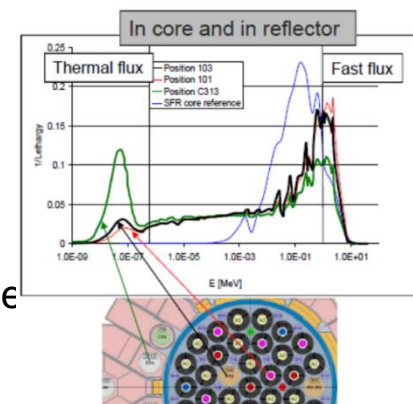


- Key requirement in predicting / ensuring SI is to understand the transferability of data from MTR to operational conditions

- Microstructural properties
- Tensile properties
- Stress-corrosion cracking
- Swelling

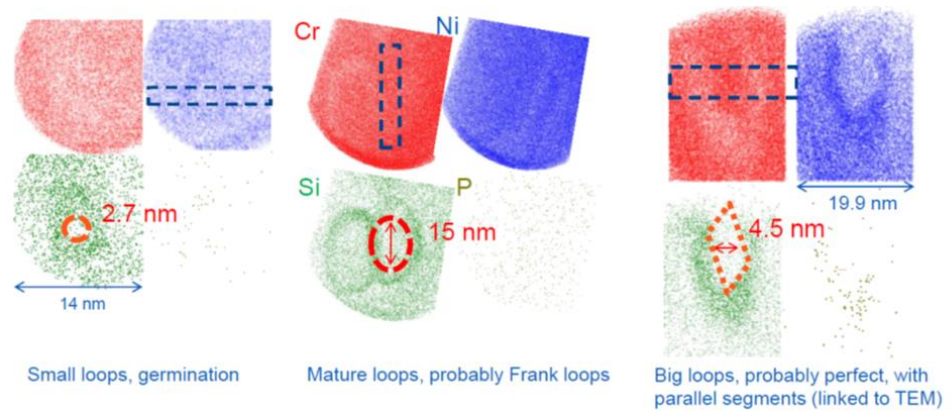


- **Needs:** Irradiation of austenitic steel in MTRs with **tailored neutron spectra**
- Using 2 or 3 MTRs, with portions of capsules shielded from thermal neutrons, will provide a range of neutron spectra
- Aiming for
 - $T_{irr} \sim 370-380^{\circ}\text{C}$
 - neutron flux $> 10^{14} \text{n.cm}^{-2} \text{s}^{-1}$; $E > 1 \text{MeV}$
 - $\text{dpa} > 5-10$
 - a very well known irradiation location in terms of neutronic performance (flux levels, spectrum, nuclear heating)
 - State-of-the-art **real time neutron flux measurement**



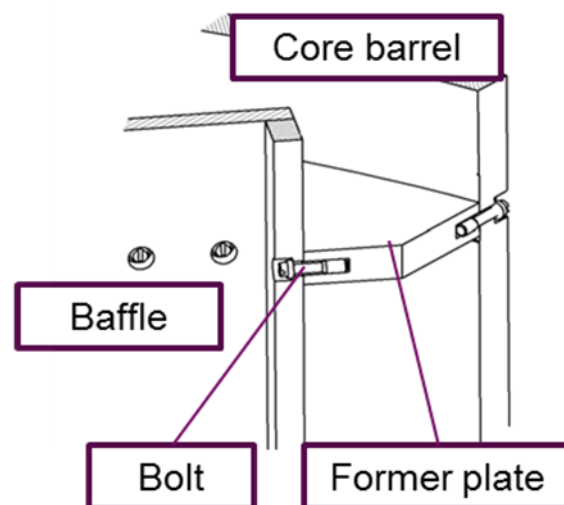
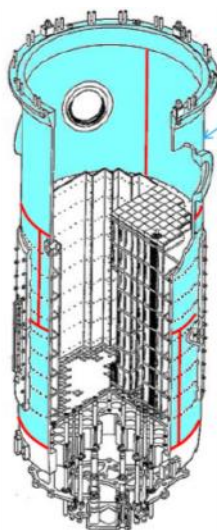
- Characterise property changes
- Dedicated benchmark, **protocol development**
- Mechanical properties
 - Tensile tests at RT and T_{irr} ,
 - Fracture toughness
- Microstructure (SEM, TEM and APT)
- Simulations with **multi-scale modelling tools**

Characterization of irradiated nanoscale microstructure based on APT



Expected outcomes:

- Utilisation and (further) development of **best practice guideline** in Internals irradiations
- Harmonisation in characterisation/evaluation of exposure conditions and PIE
- **Improved understanding** of roles of flux and neutron spectrum in the degradation of internals
- Guidance on transferability of FR / MTR data to LWR operational conditions
- Production of data in support of **predictive model** development for Internals degradation and plant condition assessment



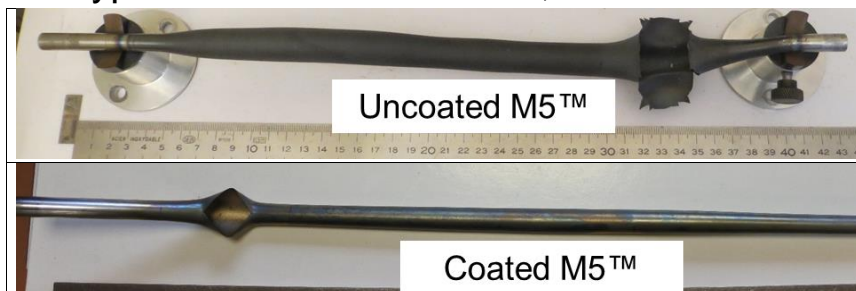
Some SOA results of CEA-R&D / Out-of-pile demonstrated benefits:

- Negligible in-service clad corrosion (& fretting resistance ↗) => no significant in-service H uptake => positive effects also expected on post-service issues (transportation and storage)
- Improved LOCA & beyond prop.: higher HT steam oxidation resistance => less H₂ production and additional copying time before reaching full clad embrittlement (quenching & PQ strength/ductility ↗), Peak clad T ↘, creep resistance ↗, balloon & burst opening sizes ↘ (fuel relocation/dispersal mitigation)

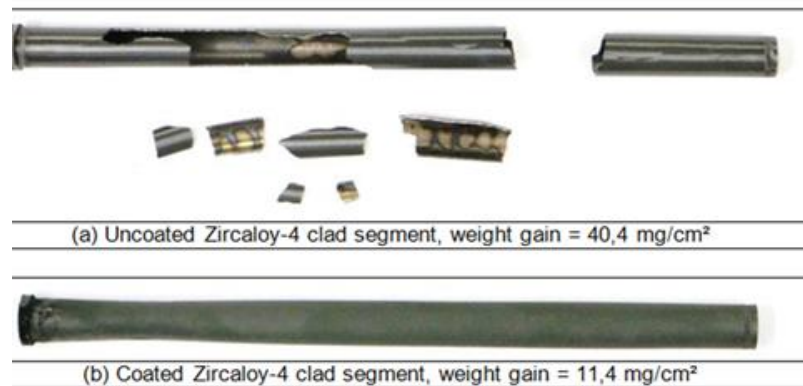
Steam oxidation for 300s at 1200°C (DBA conditions):



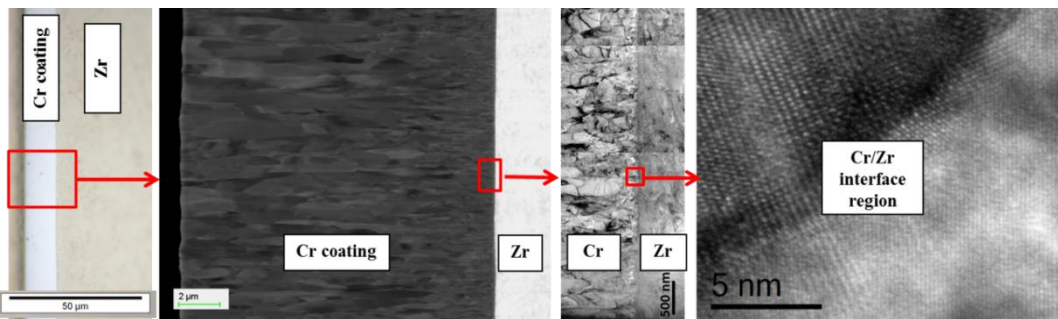
Internal pressure “EDGAR” creep/rupture tests (prototypical of LOCA transients, steam environment):



Steam oxidation for 6000s at 1200°C + direct water quenching (Design Extension Conditions):



On-going: Very high temperature (>1300°C) steam oxidation tests + quenching to address “Zr-Cr Eutectic” issue for more severe hypothetical accidental scenarii



(a) Optical Microscopy

(b) Electron Backscattered Mode SEM

(c) Conventional TEM

(d) High Resolution TEM

Ion irradiation already performed

- Zr-Cr interface crystallinity-bonding maintained after ion irradiation up to >20dpa at 400°C
=> very good residual irradiated Cr coating adhesion, no delamination...

On-going neutron irradiation at Halden

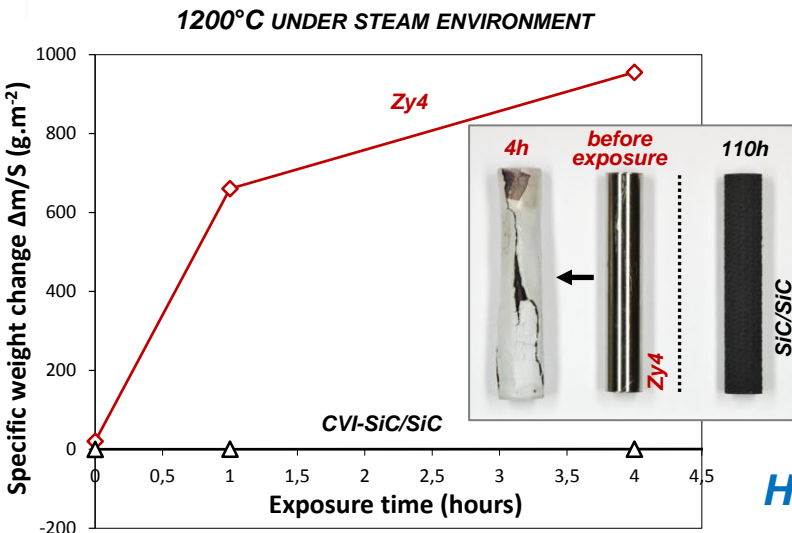
- IFA-796, PWR environment)
=> will need dedicated/specific PIEs

Some pending issues (and further work needed)

- Confirm the Cr-coated claddings corrosion resistance under irradiation in representative PWR/BWR environments;
- PCMI issues (confirm the good coating adhesion, fretting resistance... of neutron irradiated Cr coatings...) => needs of dedicated/specific in-pile & PIEs experiments, including power ramp testing;
- Perform post-irradiation semi-integral and integral LOCA tests to confirm/assess:
 - the enhanced creep/burst properties and the potential benefits on irradiated fuel relocation / dispersal issues at high BU;
 - the enhanced HT steam oxidation resistance and quenching/PQ mechanical properties;
- Evaluate the Cr coated claddings benefits for post-service situations (transportation and storage, behavior upon hypothetical spent-fuel-storage-pool accidents...).

Application temperature range and dimensional stability => CVI-SiC/SiC composites have a very high potential for EATF

SOA results of CEA-R&D / Out-of-pile demonstrated benefits at very High Temp.



- Extremely slower oxidation kinetics for SiC/SiC vs Zr alloys !

Competition between simultaneous growth and volatilization of a silica oxide

$$\Delta m/S = \Delta m_{\text{SiO}_2(\text{s})}/S + \Delta m_{\text{SiO}_2(\text{vol})}/S$$

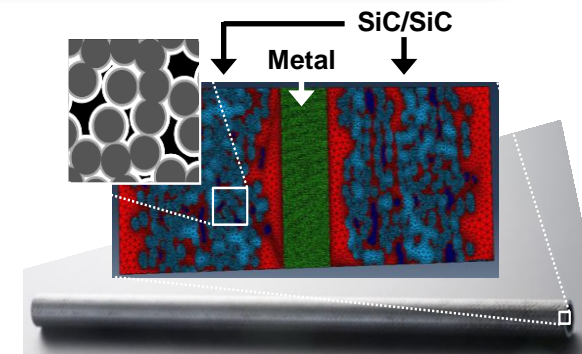
- Mechanical properties unchanged up to at least 1000°C
- Outstanding ability to preserve a coolable geometry after water quench from 1500°C to RT
- Significant reduced hydrogen production and energy release

High safety margin expected (LOCA event and beyond)

Increase coping time / delay fuel melting / ...

On-going : continue evaluation experiments to consolidate viability of the “sandwich” concept for LWR (normal conditions / out-of-pile)

- Autoclave testing – SiC recession needs to be mitigated from reaction with water to prevent silica release in the coolant
- Transposition of sandwich clad to LWR requirements (design, suitable liner)
- Manufacturing of elongated and closed clad (welding closure)



SiC Gaps – Recommended evaluation and testing under representative PWR conditions

On-going and planned neutron irradiation on CVI-SiC/SiC

- **IMAGO** – (AREVA-NP proprietary) **Irradiation** in Gösgen **commercial reactor**

Corrosion kinetics and evolution of microstructure under irradiation to help for the justification of future fuel rod irradiations



- **IL TROVATORE** – **Irradiation of tubes in BR2 Belgium reactor**

Fundamental material data induced by neutron:
Swelling and irradiated properties on nuclear grade SiC/SiC composite



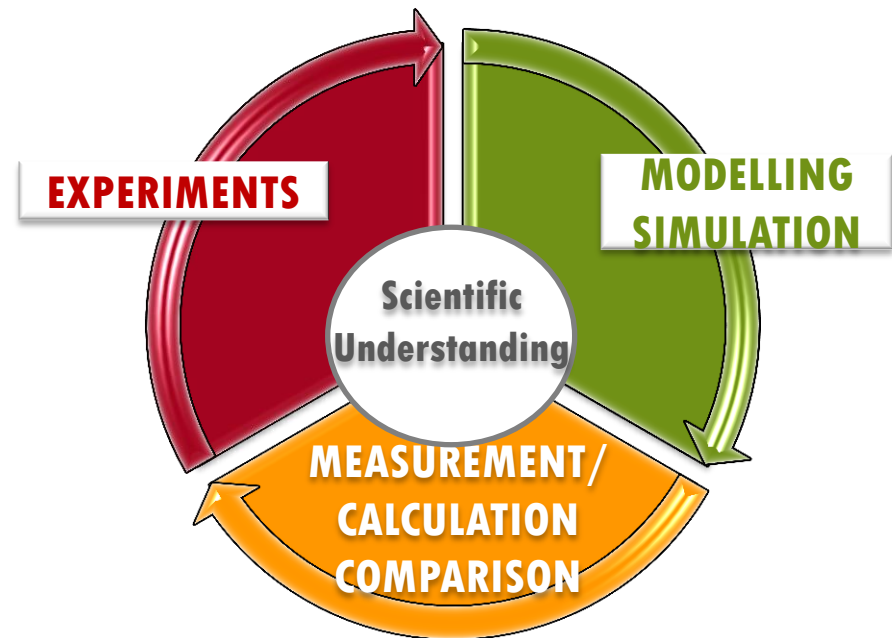
Pending issues and further work required

- Compatibility of the investigated solutions to mitigate the hydrothermal corrosion of SiC need to be addressed in pile
- The decrease in thermal properties under neutron must be known, as well as the consequences on a multilayered cladding concept
- PCMI (and SiC/SiC swelling) issues at LWR relevant temperature and neutron spectrum required, including Power ramps testing

Irradiation testing on complete assemblies needed to demonstrate the viability of the « sandwich » design

- Ability of the metal / SiC interface to retain integrity during normal and transient conditions
- Characterization of strength and hermeticity of the closure by liner welding

This presentation has shown some examples of pending scientific issues on R&D on Fuel & Material: enhanced experiments performed through international joint projects is of primary importance to successfully address these issues.



Many people at CEA took an important part in the presented work. We want to sincerely thank them all !

EDF and AREVA NP are acknowledged for their technical and financial support

Thank you for your attention



FROM RESEARCH TO INDUSTRY



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Back-Up Slides

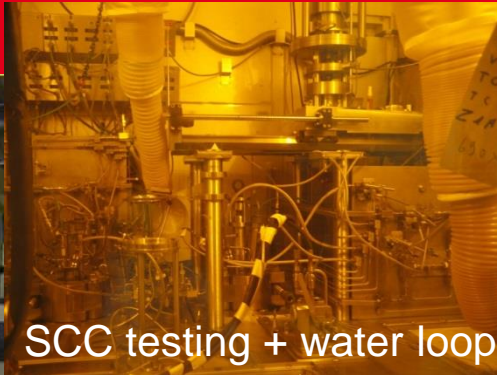
A FEW REFERENCES FOR MORE INFORMATION

- C. Riglet-Martial et al., J. Nucl. Mater. 447 (2014)
- C. Riglet-Martial et al., J. Nucl. Mater. 480 (2016)
- B. Baurens et al, J. Nucl. Mater. 452 (2014)
- J. Noirot et al, J. Nucl. Mater. 446 (2014)
- J. Noirot et al, J. Nucl. Mater. 462 (2015)
- C. Esnoul et al, Top Fuel proc. (2015)
- B. Michel et al, Key Engineering Materials (2016)
- B. Michel et al, Engineering Fracture Mechanics 75 (2008)

State of the art equipments



Ligne M of LECI lab (CEA Saclay)



SCC testing + water loop



Tensile testing + furnace

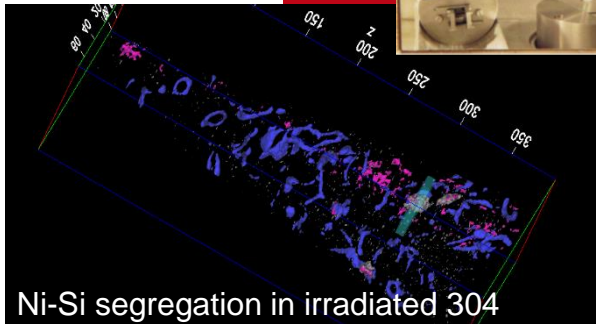
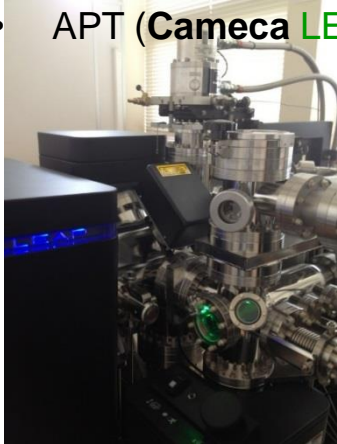


EPMA



ICP - AES

- APT (Cameca LEAP 4000X HR)



Ni-Si segregation in irradiated 304



TEM
(FEI 300 kV)

Sample preparation by nuclearized FIB or electropolishing

