



Wir schaffen Wissen – heute für morgen

# Laboratory for Nuclear Materials (LNM)

## - Overview, activities & selected highlights -

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Nuclear Energy and Safety

Paul Scherrer Institute

5232 Villigen PSI, Switzerland

**NES Info Event 2014, PSI, OSGA/EG06, March 27, 2014.**



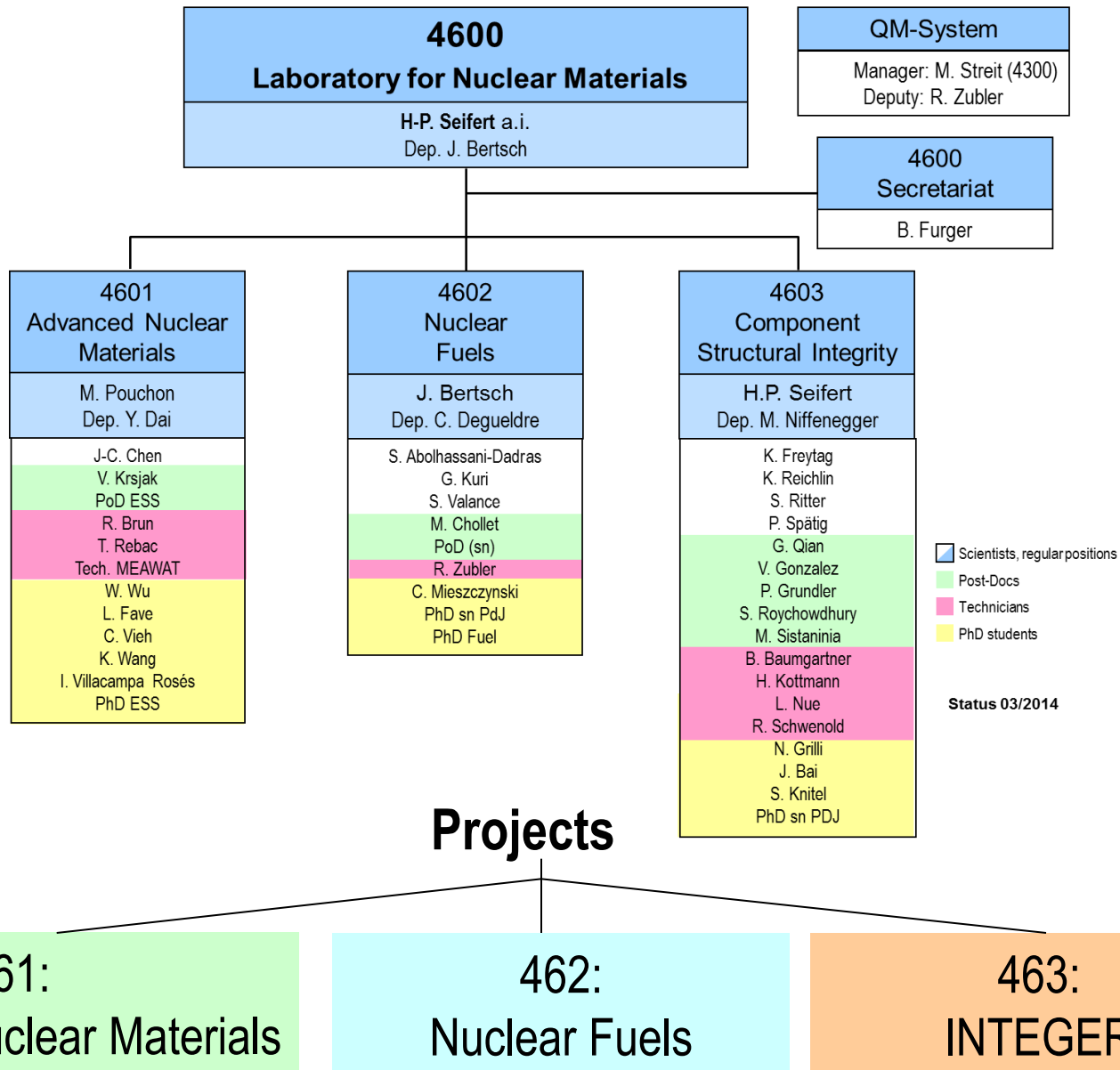
- **Overview on LNM**
  - Mission
  - Organisational structure
  - Research & project portfolio, core expertise's
  - Lab infrastructure & modeling tools
  - Education & teaching activities
  - Scientific services
- **Selected examples of current activities & highlights**
  - Advanced Nuclear Materials
  - Nuclear Fuels
  - INTEGER

# Overview on LNM

# Mission of LNM

- The LNM is the principal research unit and national centre of excellence in Switzerland in the domains of (radioactive) **materials behaviour** and **ageing** in **nuclear installations**.
- It provides material-related academic R&D contributions and scientific services to the **sustainability** of **current** and **future nuclear installations** for electricity & heat generation or waste reduction as well as to the performance of **nuclear research facilities**. A special emphasis is placed to the **safety & safe long-term operation** of the CH NPPs.
  - **Material ageing** in the primary circuit and its impact on **integrity, safety & lifetime**
  - **Performance** and **safety** of **LWR core materials** in **service** and storage
  - **Radiation damage** in structural and core/target materials of advanced nuclear and accelerator systems.
  - **Material irradiation program at SINQ** in co-operation with the Spallation Neutron Source Division.
  - **Post-irradiation examinations** and **failure analysis** in close cooperation with the Hotlab Division AHL.
- LNM is engaged in academic **teaching** and **education** as well as in **knowledge transfer** in its activity fields contributing to the education of the future nuclear specialists and preservation of expertise & excellence.
- Its **independent expertise** and **excellence** are always available to the **Swiss safety authority**, e.g. for expertise's and consulting (TSO), and for the **industry**, e.g., for material examinations and failure analysis.
- It operates a state-of-the-art **lab & computing infrastructure** and **modelling tools** for the **characterization** of **(radioactive) materials** (strongly benefiting from PSI's unique large scale facilities: hotlab, SLS, SINQ, SwissFEL) and for the **analysis & prediction** of the **material behaviour, integrity, safety & lifetime**.

# Current Organisation of LNM



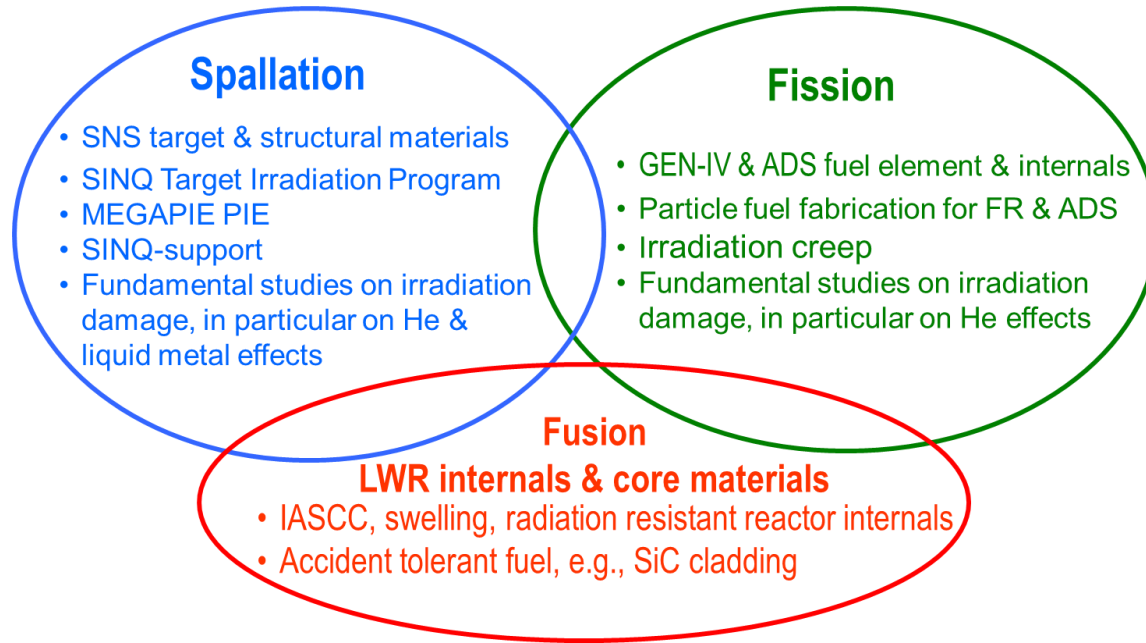
# LNM Research Portfolio

	Advanced Nuclear Materials	Nuclear Fuels	INTEGER
<b>Project leader</b>	<b>M. Pouchon</b>	<b>J. Bertsch</b>	<b>H.P. Seifert</b>
<b>Topic</b>	SNS/GEN-IV candidate materials radiation damage at high T & dose	Performance of fuel & integrity of cladding	Material ageing & structural integrity
<b>Components</b>	Target & structures (SNS) Fuel element (GEN-IV/ADS)	Core materials (fuel, cladding)	Pressure boundary comp. & reactor internals
<b>Key words</b>	Performance (SNS) Sustainability (GEN-IV/ADS)	Performance, safety	Safety, lifetime (extension), performance
<b>Systems</b>	Spallation Neutron Sources GEN-IV / ADS, LWR-III++	LWR (GEN-II & III)	LWR (GEN-II & III)
<b>Extern. funding Main source</b>	40 % EU, STIP, CCEM	50 % swissnuclear, industry	60 % ENSI, swissnuclear
<b>Indispensable for</b>	PSI (SINQ/accelerators) Education	Radioactive material know how	Independent expertise (TSO)

<b>R &amp; D share:</b>	65 % applied & 35 % basic 70 % GEN-II & 30 % GEN-IV/SNS	75 % experimental & 25 % modeling 55 % CH, 35 % international, 20 % PSI
<b>Lab capacity:</b>	10 % for teaching & education	10 - 15 % for expertise & service work

# Advanced Nuclear Materials

## Performance of SNS & sustainability of nuclear energy production (GEN-IV, ADS)



### Spallation Neutron Sources

↔ ADS ↔

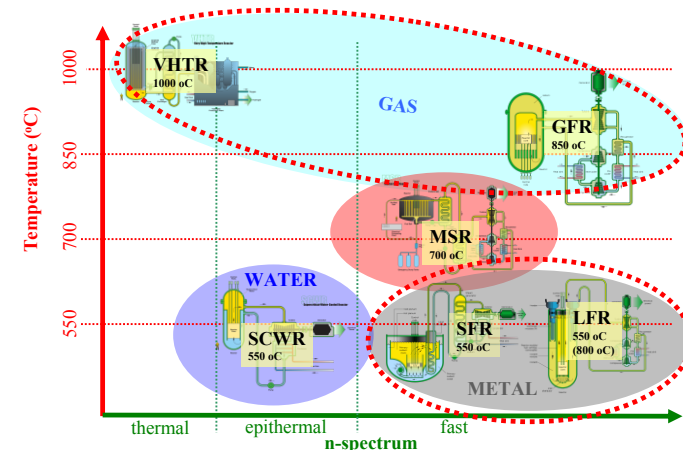
### GEN-IV Nuclear Systems

#### MW-Class Spallation Targets

- SINQ at the Paul Scherrer Institut (PSI)
- SNS at the Oak Ridge National Laboratory (ORNL)
- JSNS at the Japan Atomic Energy Agency (JAEA)



*same candidate materials  
(ODS, FM steels, ...)  
and similar problems  
(He, radiation creep, LME, ...)*



### + European Spallation Source (ESS)

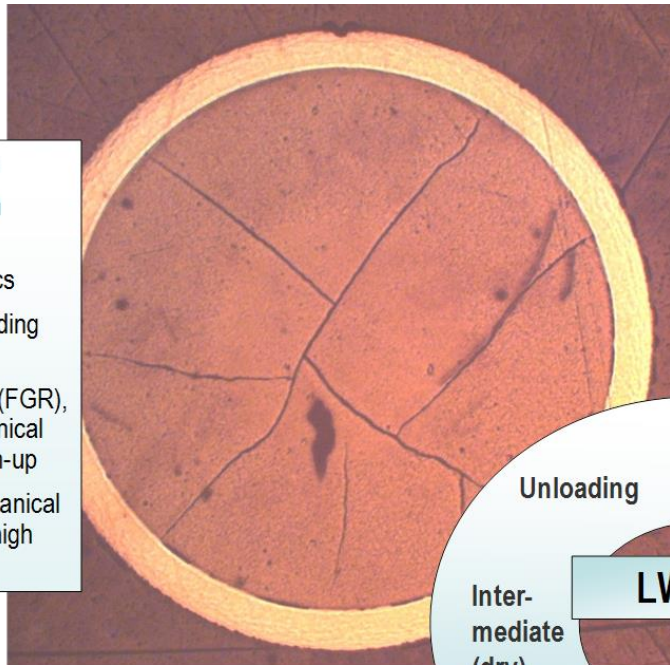


# Nuclear Fuels

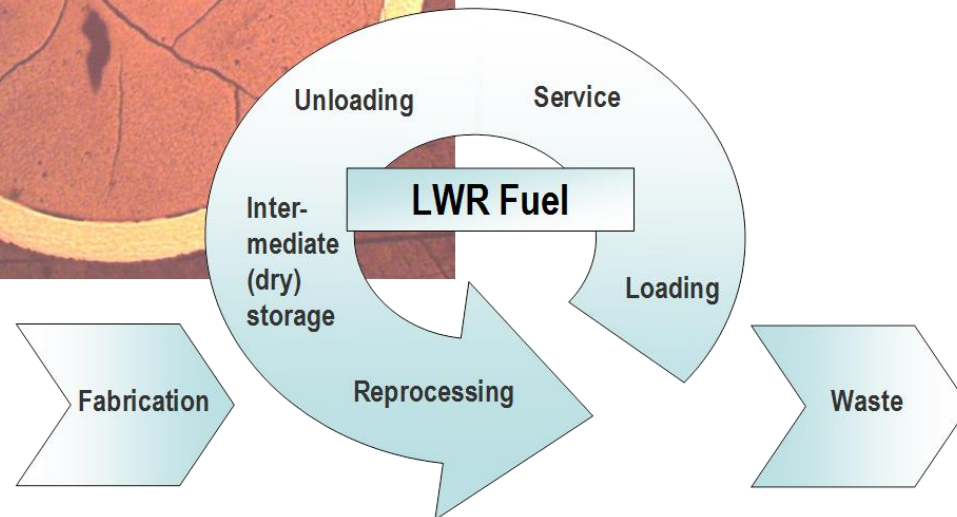
**Fuel life & integrity** → core materials issues with respect to **safety & performance**

*close collaboration with Swiss utilities (KKL, KKG)  
& fuel vendors (AREVA, Westinghouse) or  
international programs (NFIR, SCIP, MUCIZ, ...)*

- **corrosion & mechanical behaviour** of **cladding** in service & storage
- **fuel element behaviour in service**
- **PIE of spent fuel & failure analysis** as scientific service work (with AHL)
- pre-studies on **accident tolerant fuel** and **Th fuel** for LWRs
- head end fuel reprocessing (in AHL)



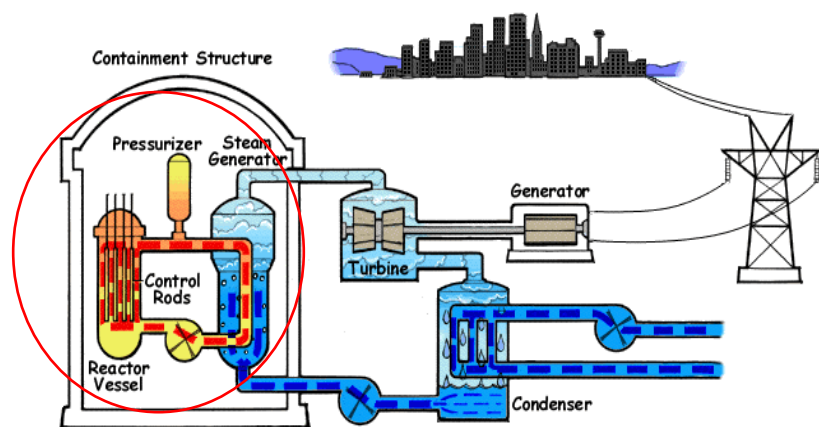
- CRUD: Magnetic properties
- Oxide: Corrosion, hydrogen uptake
- Cladding: Hydrogen, mechanics
- Gap: Isotopes, pellet cladding interaction (PCI)
- Rim: Fission gas release (FGR), physical and mechanical properties, high burn-up
- Pellet: FGR, physical/mechanical properties, doping, high burn-up





## Scientific contributions to & maintenance of independent expertise in the field of Safe & efficient LTO of Swiss NPPs in the context of material ageing

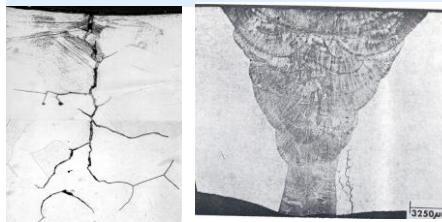
### Primary pressure boundary components & reactor internals



- Critical systems with regard to **safety** and **lifetime (extension)**
- Assurance of **structural integrity** in the context of material ageing a key task

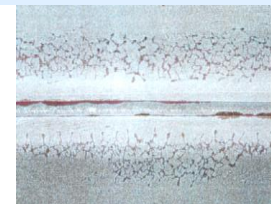
### Ageing & degradation mechanism's

Stress Corrosion Cracking



→ Formation and growth of cracks

Thermal Fatigue



Flow-Accelerated Corrosion



→ Wall thinning  
→ CS (and LAS) with less than 0.2 % Cr

Irradiation Embrittlement



→ Reduction of toughness & ductility  
→ Increase of DBTT & brittle fracture risk

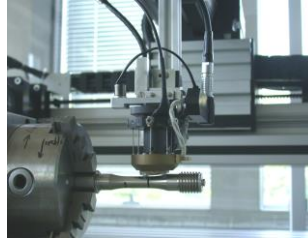
- **Ageing & degradation** → characterization & mechanism, mitigation
- **Integrity** → deterministic & probabilistic structural integrity & lifetime assessment
- **Diagnostic** → ND early detection of damage and monitoring of ageing conditions

## Microscopy



- FEG-SEM / EBSD& EDX, SEM/EDX
- TEM, FIB, *shielded FIB/SEM?*
- LM & SM. metallography

## ND Diagnostics



- Magnetic methods (EC, 3 MA, GMR, SQUID, Ferromaster)
- Electric & thermoelectric methods

## Beam Line Techniques



- SLS: EXAFS, XAS, XRD, in-situ testing with  $\mu$ -LD, ...
- SINQ: STIP, ND, residual stress,

**Broad spectrum of tools for chemical & microstructural or mechanical & physical properties characterization of in-active and highly radioactive specimens (bulk, surface, local)**

## Corrosion Testing



- 9 HT-water loops with autoclaves with loading systems. Static autoclaves.
- Crack initiation & growth monitoring
- HT electrochemistry (ECN, IS, RE).

## Mechanical Testing



- **Inactive:** TMF, HCF, LCF, impact, tensile, creep, hardness,  $\mu$ -hardness, furnaces, hydrogenation facility, DIC.
- **Active:** Tensile, LCF, n-intender, small punch, drop tower, in-situ irradiation creep.

## Hot Laboratory (AHL)



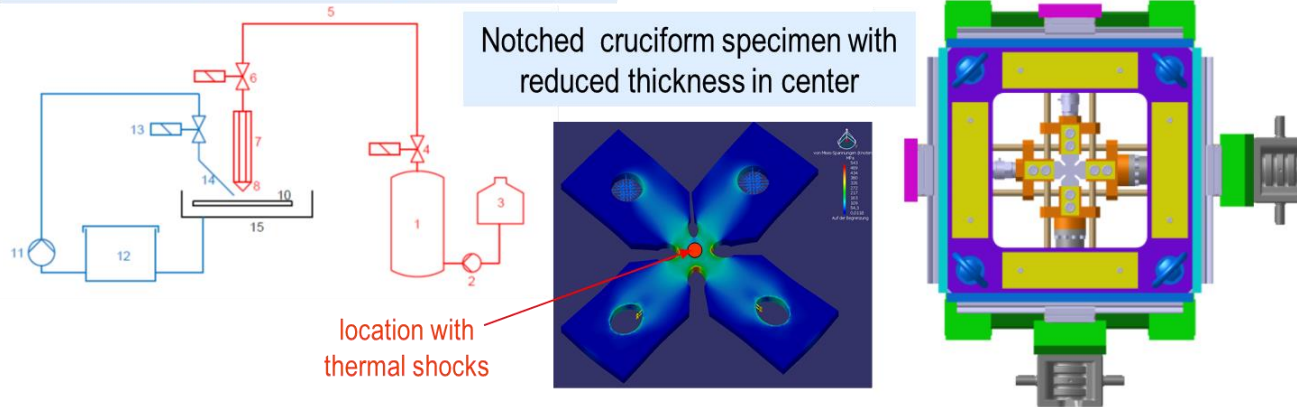
- LA ICP-MS, EPMA, SIMS
- Active metallography & sample preparation
- $\gamma$ -spectrometry, fission gas analyzer

# Examples of Unique Facilities

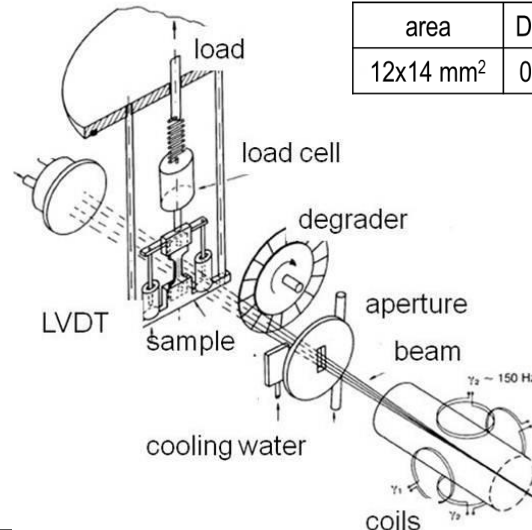
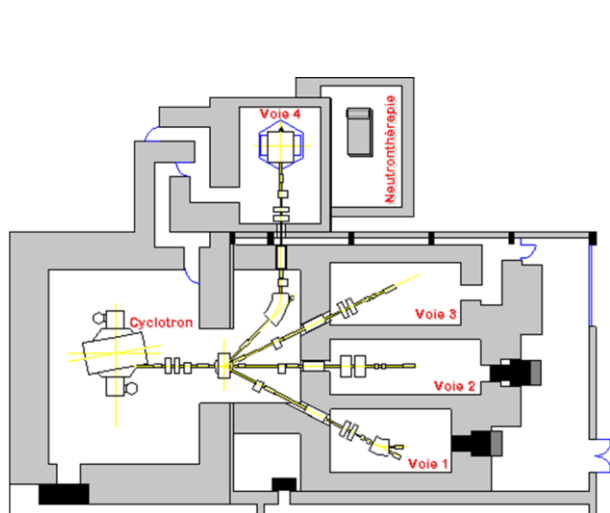
## Cyclic TMF Facility with Biaxial Pre-Loading (INTEGER)

Cyclic thermal shocks by hot steam & cold water with specimen heating option

Controlled independent biaxial pre-loading



## Facility for In-Situ Creep under Irradiation at CRNS / CEMHTI (ANM)



area	Dispal. rate	Impl. rate	T (° C)
12x14 mm <sup>2</sup>	0.03 dpa/h	150 appm-He/h	150-1000

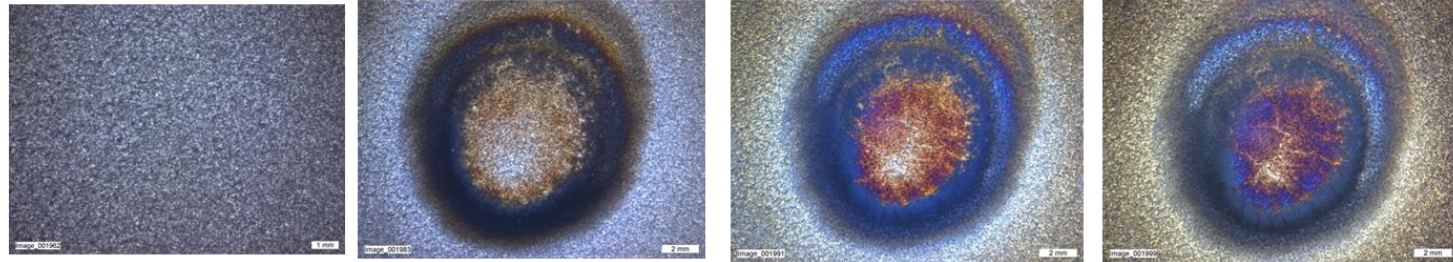
Ions	Energy (MeV)	Intensity (μA)	Range (μm)
p	5--38	15	2800
H <sub>2</sub> <sup>+</sup>	5--25	13	380
d	5--25	40	760
α	10--50	15	380
<sup>3</sup> He	10--50	15	470

- $\Delta\varepsilon \sim 10^{-5}$
- $\Delta T = \pm 2^\circ \text{C}$



# Crack Network Formation due to Cyclic Thermal Shocks

Crack network formation due to cyclic thermal shocks ( $\Delta T=160\text{ }^\circ\text{C}$ , 1Hz, H<sub>2</sub>O)

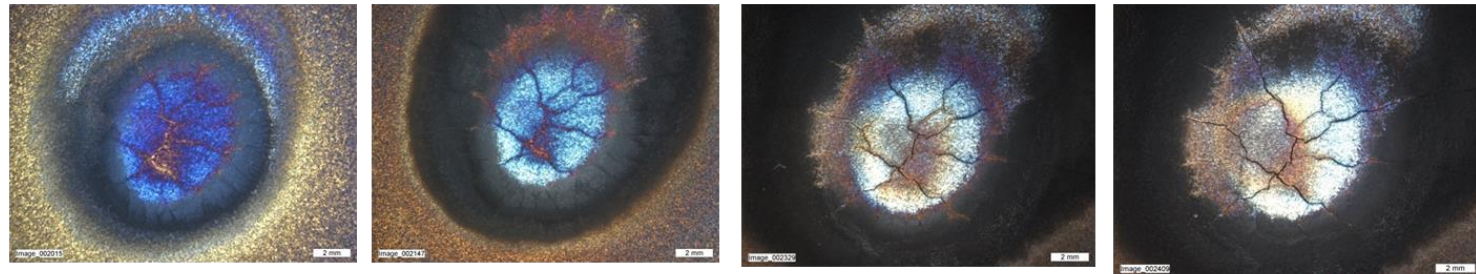


0 cycles

150 kc

400 kc

1050 kc

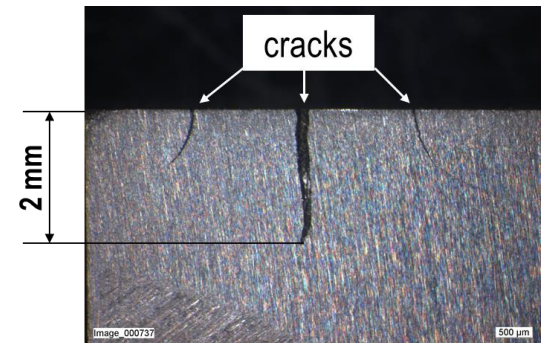
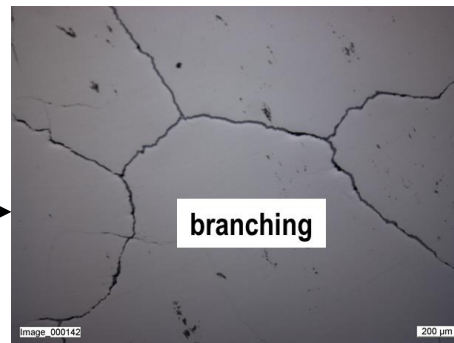
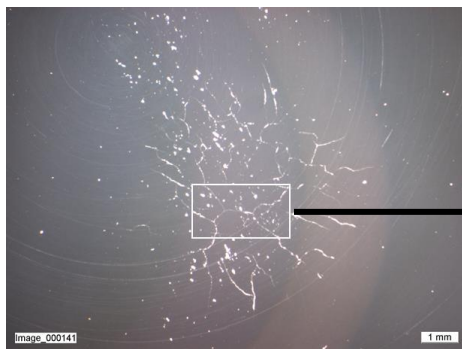


1950 kc

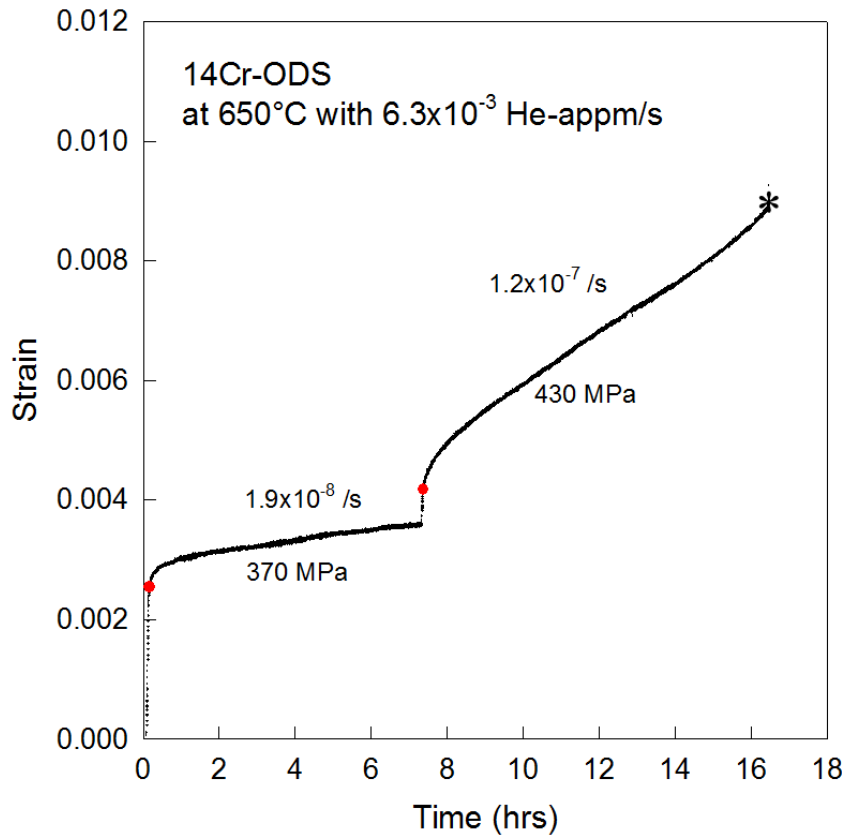
2450

3000 kc

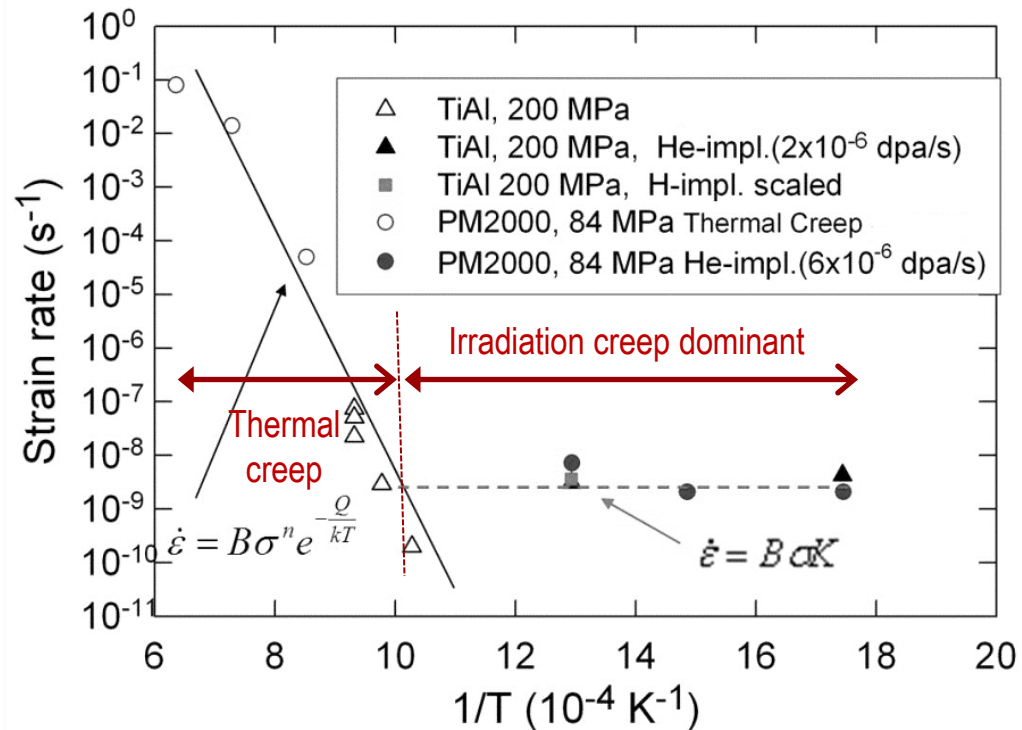
3300 kc



## Creep strain of 14Cr ODS during He-implantation at 650°C

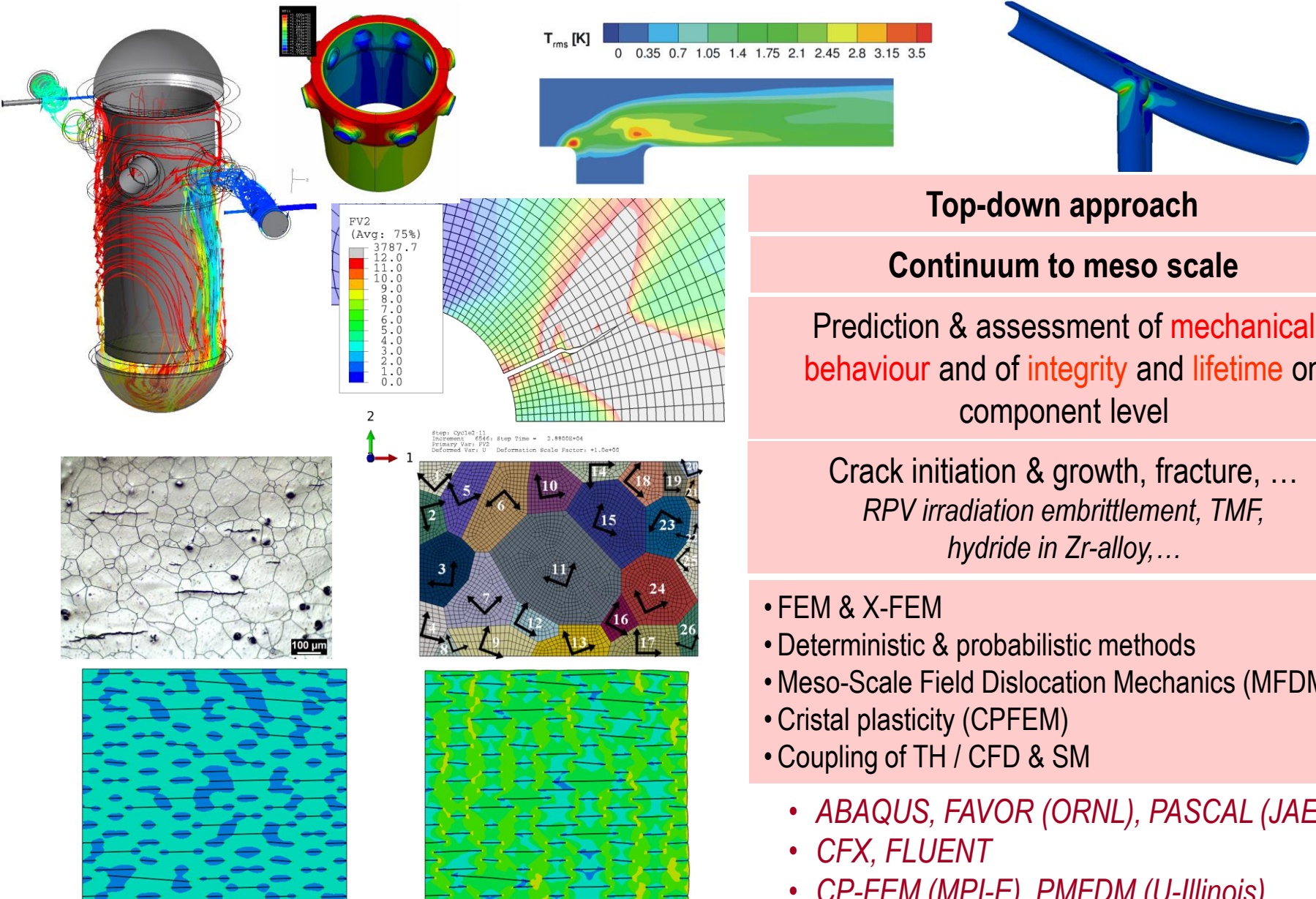


## Thermal & irradiation Creep in TiAl and ODS





# Modeling Activities and Tools in LNM



Top-down approach

Continuum to meso scale

Prediction & assessment of **mechanical behaviour** and of **integrity** and **lifetime** on component level

Crack initiation & growth, fracture, ...  
*RPV irradiation embrittlement, TMF, hydride in Zr-alloy, ...*

- FEM & X-FEM
- Deterministic & probabilistic methods
- Meso-Scale Field Dislocation Mechanics (MFDM)
- Cristal plasticity (CPFEM)
- Coupling of TH / CFD & SM

- *ABAQUS, FAVOR (ORNL), PASCAL (JAERI)*
- *CFX, FLUENT*
- *CP-FEM (MPI-E), PMFDM (U-Illinois)*



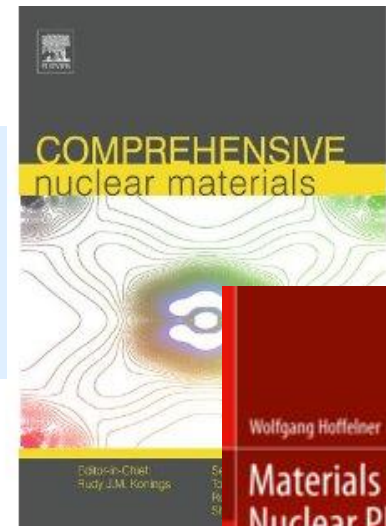
- **10 PhD** and **8 PoD** running projects in LNM
- Contribution to **ETHZ/EPFL NE Master** by lectures & master projects
- **EPFL Doctoral School in MS & E and Physics** by lectures
- Contribution to **swissnuclear/PSI education course Kerntechnik**
- Other university teaching (Uni Geneva, ...), summer schools, tutorials
- Textbooks and reference books on nuclear materials

## Nuclear Materials Workshop



ÉCOLE POLYTECHNIQUE  
FÉDÉRALE DE LAUSANNE

EPFL Doctoral School in MS & E,  
Course MSE-600, Effects of  
Radiation on Materials **by LNM**



**Materials for  
Nuclear Plants**

From Safe Design to Residual Life Assessments

Springer

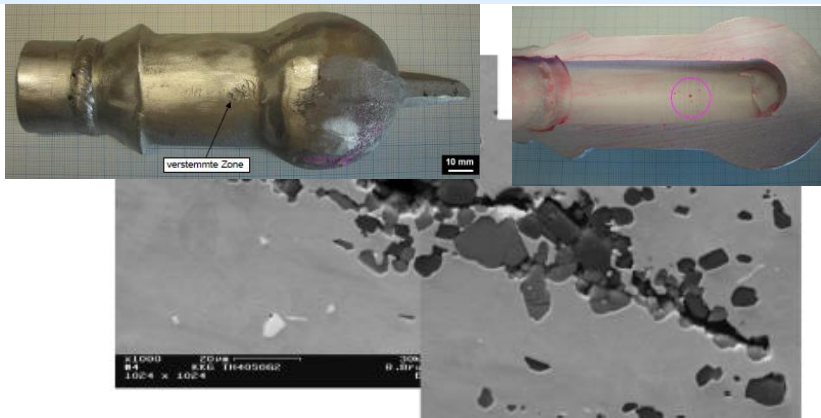
Staudinger-Durrer Medaille 2012 of MS Department of  
ETHZ for **W. Hoffelner**



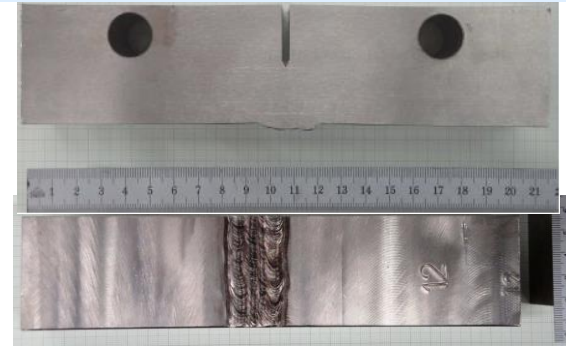
# Examples of External Services

- Expertise on KATAM (basic document of ageing management) for ENSI
- Expertise on SCC of SS for ENSI
- Consulting for CH NPPs (e.g., Overlay repair welding of SCC in feedwater nozzle in KKL)

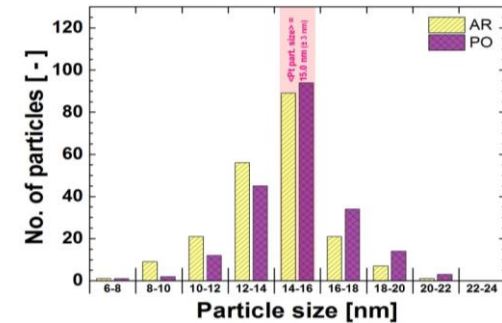
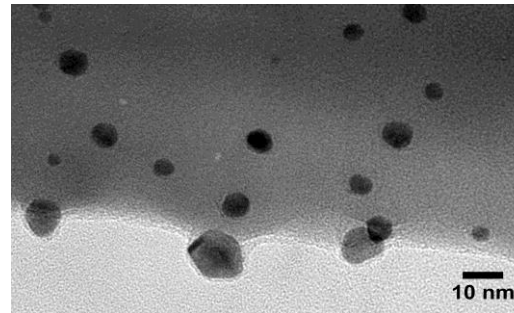
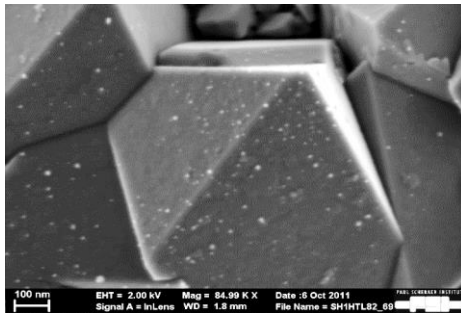
## Failure analysis of leaking valve for KKG



## NDT test bodies with fatigue cracks in SS welds for KKG



## Pt particle analysis for KKL, KKM, Cofrentes & EPRI, YUMOD



## Scientific support of PIE & failure analysis in hotlab

# **Selected examples of current activities & highlights**

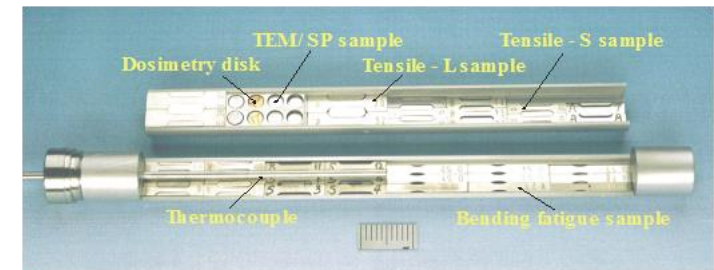
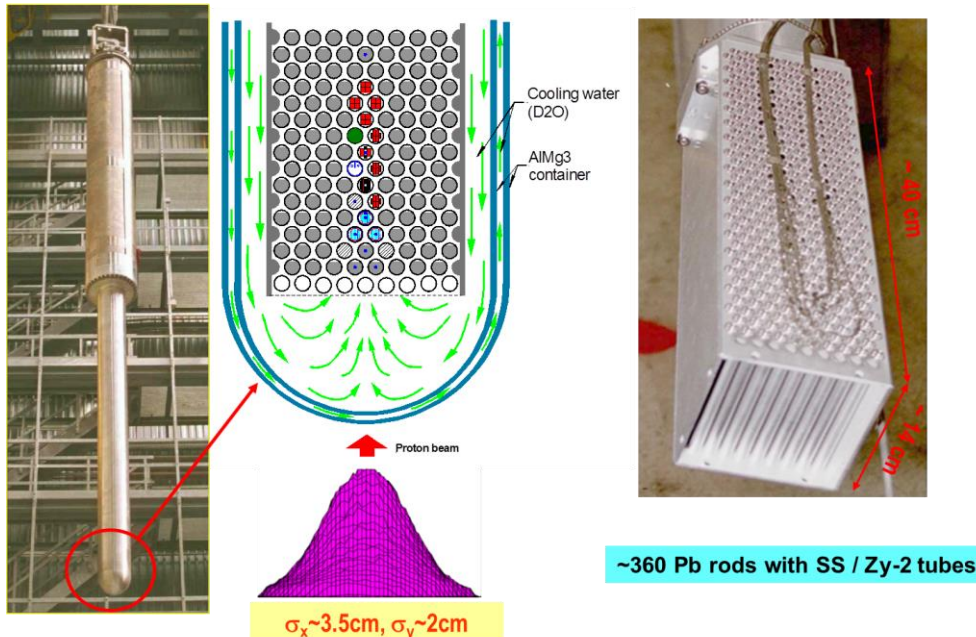
# Advanced Nuclear Materials

## Main purposes:

- 1) to provide necessary materials data for developing advanced spallation targets;
- 2) to understand radiation, He and H effects in different structural materials;
- 3) to study liquid metal effects on structural materials in intensive irradiation environments.

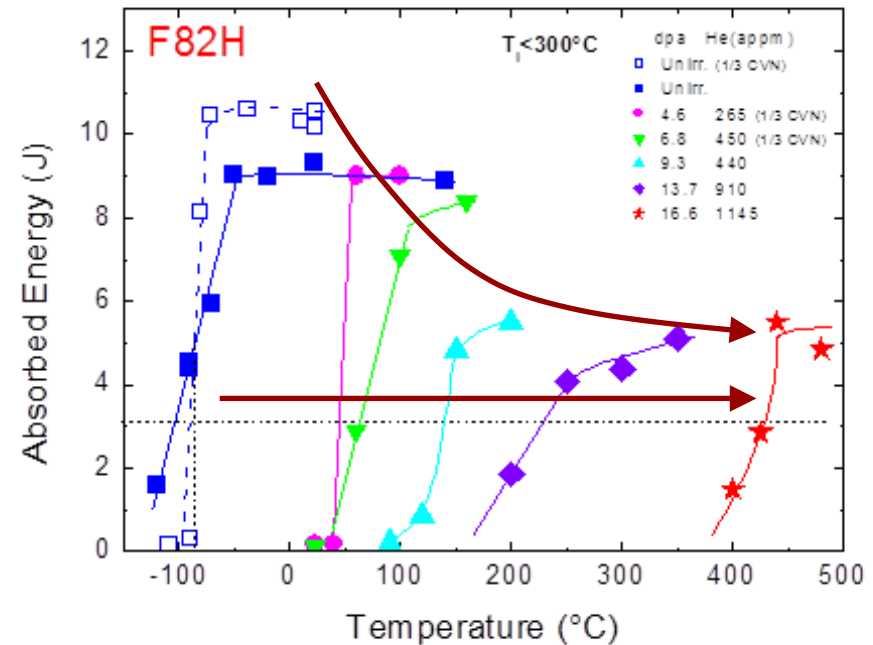
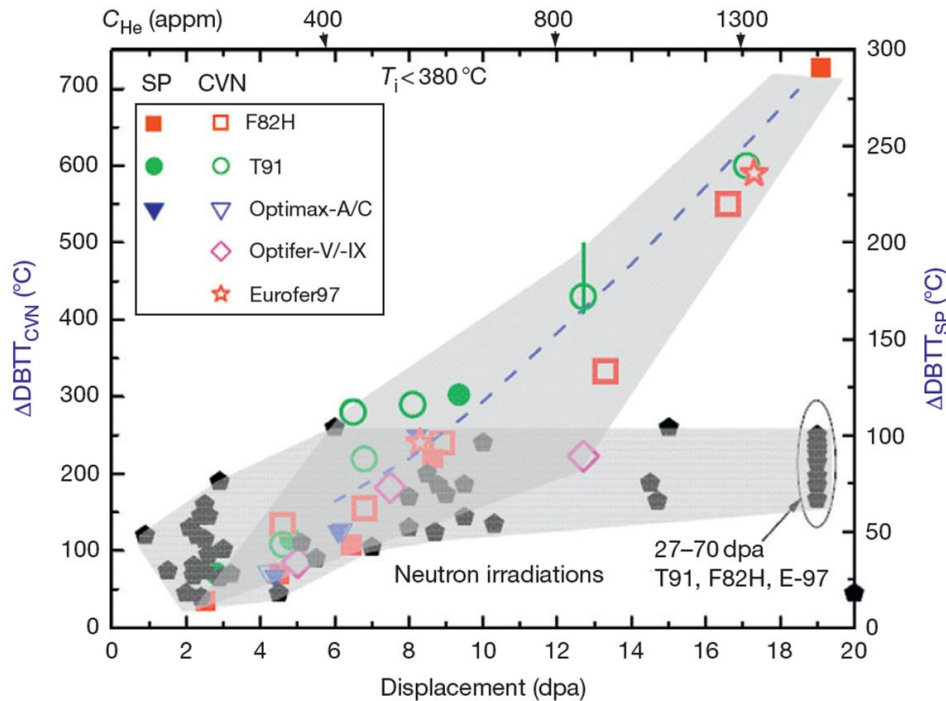
The main STIP partners are from spallation, fusion and ADS communities in Europe, Asia and USA (e.g. CEA, FZJ, CIAE, IMP, JAEA, LANL, ORNL, UCSB)

More than 7000 samples from 60+ different materials of Fe-, Al-, Ti- Ni-, Mo- W-alloys, ceramics (C/SiC, SiC/SiC...) were irradiated in first six experiments (STIP-1 to -6) up to **28 dpa / 2000 appm** He (in steels) at temperatures **up to ~800° C**.



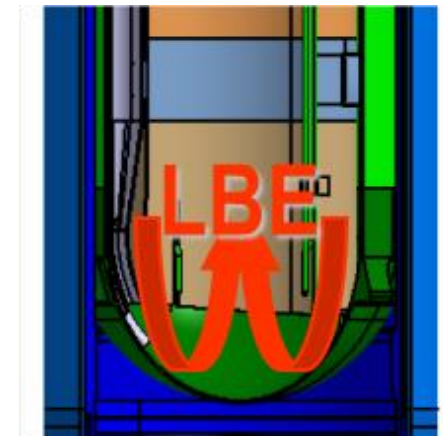
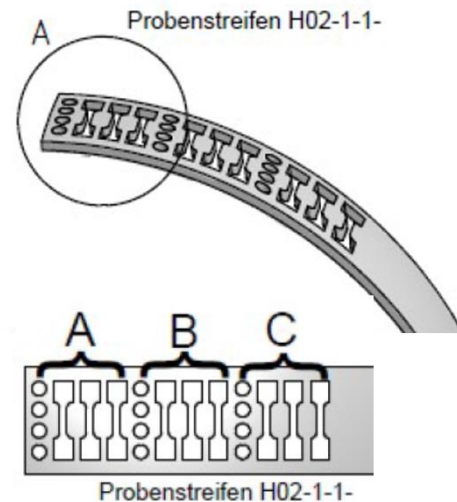
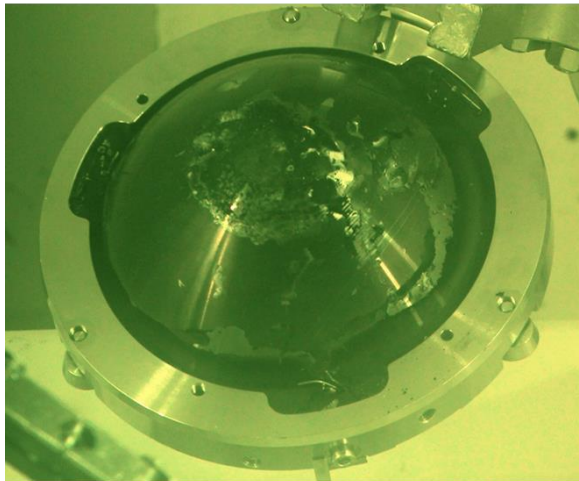


## Embrittlement of FM Steels





- MEGAPIE is a joint initiative by six European research institutions, the EU, JAEA (Japan), DOE (USA), and KAERI (Korea). Essential Element of EU-/ GERMAT project.
- Design, operate & explore a liquid PbBi (LBE) spallation target for 1 MW of beam power.
- The irradiation of the target was done at **PSI in 2006**.
- About **800 samples** were extracted from the lower part of the target for PIE:
  - changes in mechanical properties and microstructure of structural materials
  - radionuclide inventory produced in LBE and their precipitation behavior

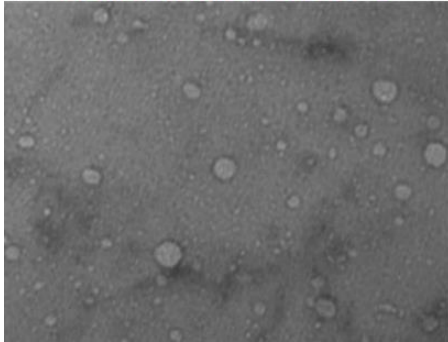


**highly activated and  $\alpha$ -contaminated samples**

## He bubble formation by TEM

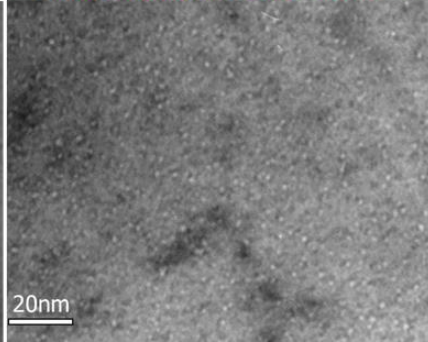
F82H

20.3 dpa, 1800 appm He,  $400 \pm 50^\circ \text{C}$



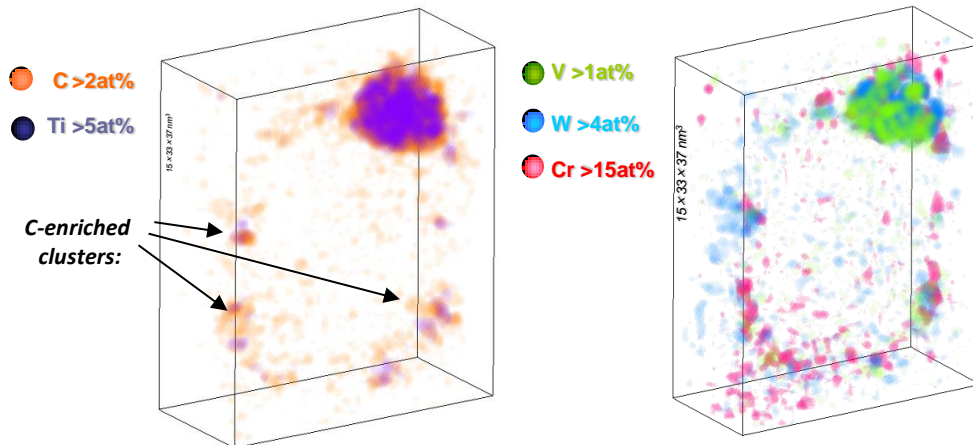
SS 316LN

19.6 dpa, 1800 appm He,  $420 \pm 55^\circ \text{C}$

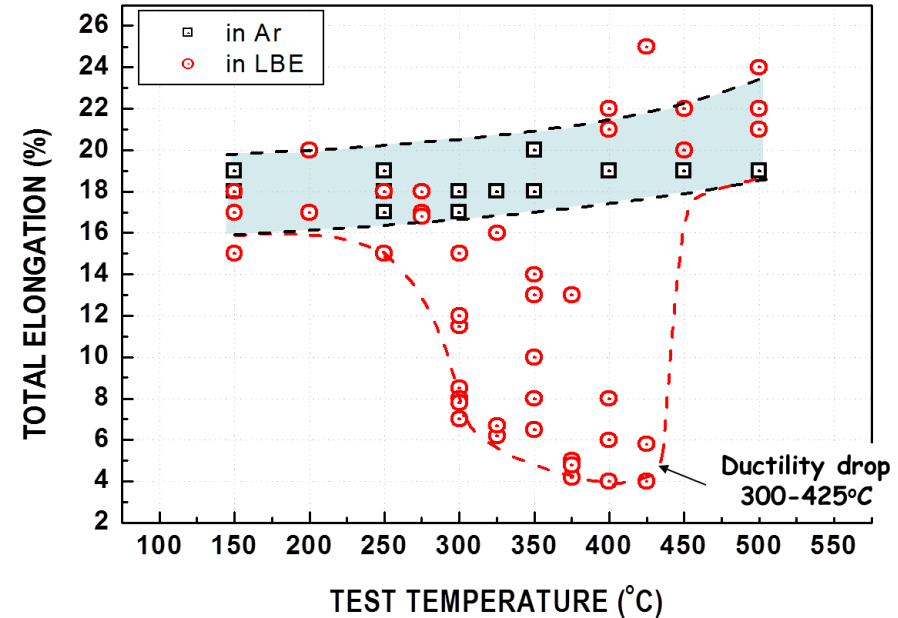


## Atom Probe Tomography (APT)

First time observation of the behaviour of solid **spallation products** (e.g., Ti) in steels



## Lead-Bismuth Eutectic (LBE) Embrittlement of FM Steel

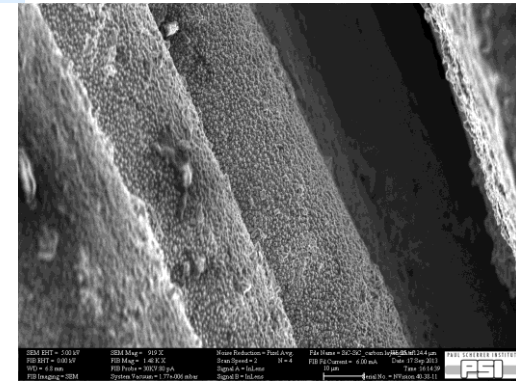


Piece of potential cladding tube  
(SiC/SiC with Ta)

PyC layer on SiC fibers

Investigation of SiC based composite tubes as potential cladding materials (e.g., in ATF):

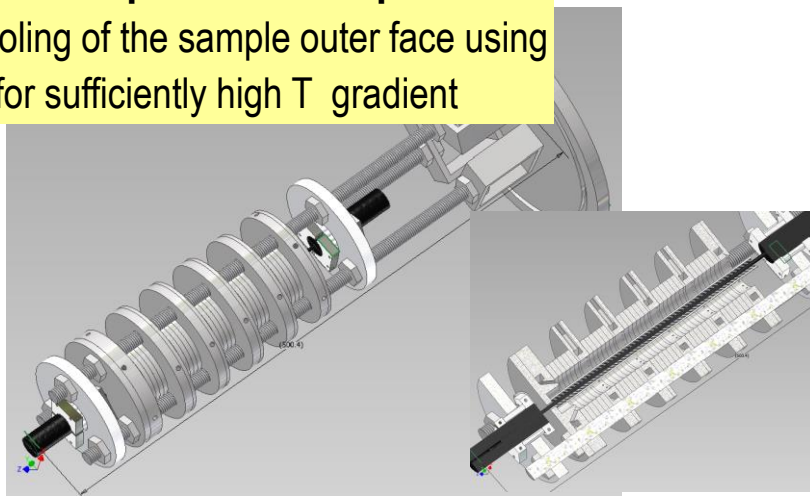
- Microstructure & irradiation-induced changes
- Thermal conductivity  $\lambda$
- Interaction between  $\mu$ -structure &  $\lambda$
- Design of  $\lambda$  measurement facility



## Thermal conductivity measurement using radial heat flow

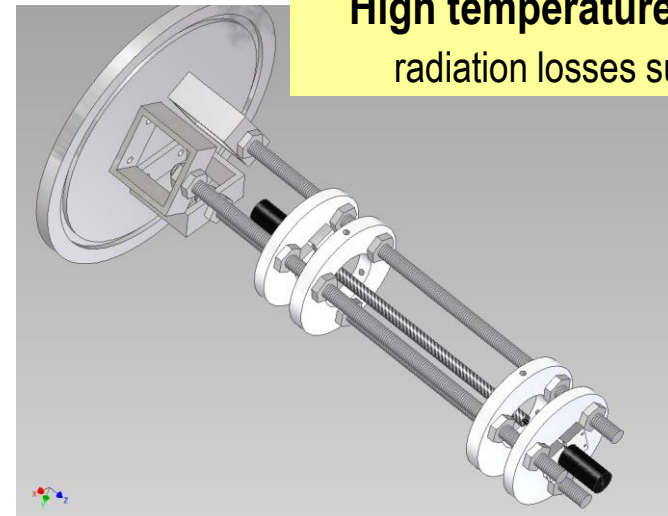
### Low temperature setup

forced cooling of the sample outer face using  $N_2$  for sufficiently high T gradient



### High temperature setup

radiation losses sufficient

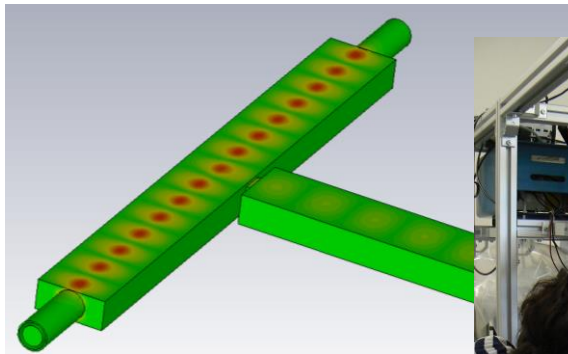




## CCEM-Project MeAWaT: Methods for Advanced Waste Treatment

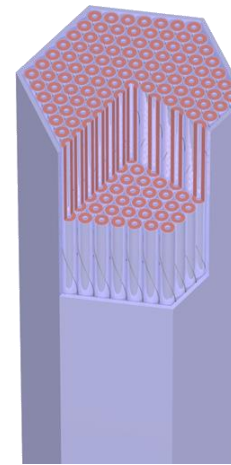
- **Particle fuel** (containing MA) **production** feasibility studies by **internal  $\mu$ -wave gelation technology** for fast reactor (FR) & acceleration-driven systems (ADS)
- Collaboration with **Chinese ADS project** (IMP/CAS) and EMPA, EPFL, LNM, AHL, LTH & LRS

### Microwave internal gelation for remote nuclear fuel production $\rightarrow$ transmutation targets

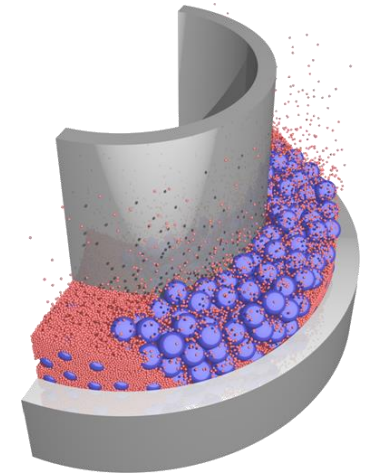


### Sphere-pac in annular pin for better heat transfer

Neutronic & thermal-hydraulic analysis by LRS & LTH



FR bundle with annular pins

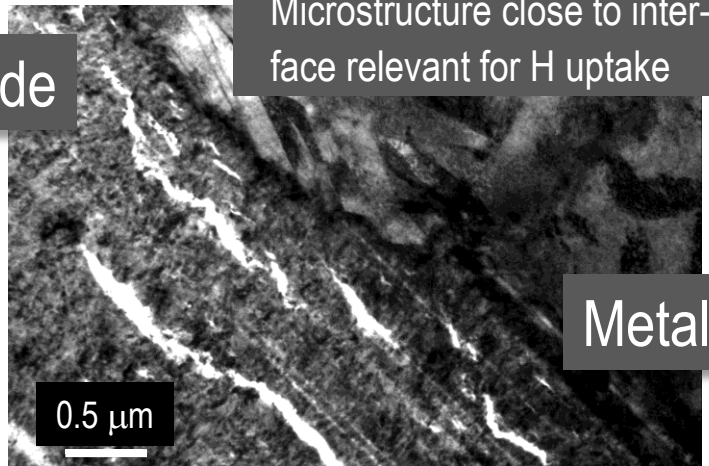


Annular pins with inner and outer cooling

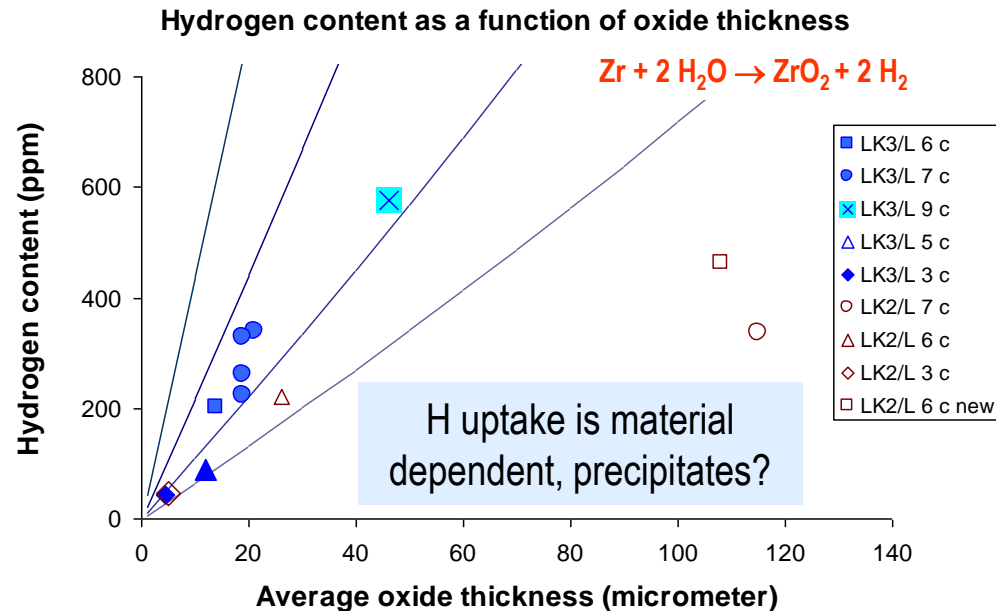
# Nuclear Fuels

# Fuel Cladding Projects – Hydrogen Uptake

- **Corrosion & mechanical behaviour** of **cladding** in service, (dry) storage and transport
- Hydrogen uptake, hydride formation, mechanical behaviour, DHC, SPP,.....
- Nuclear Fuels project investigates H uptake in **irradiated** cladding.
- H affects the **mechanical behaviour** and integrity during transients & transport & in storage
- H uptake as a function of alloy, elevation, burn-up, number of cycle, oxide thickness, ...
- Methods: hotgas extraction, EPMA, SIMS, TEM, metallography, SLS
- Collaboration with **swissnuclear, KKL, Westinghouse**

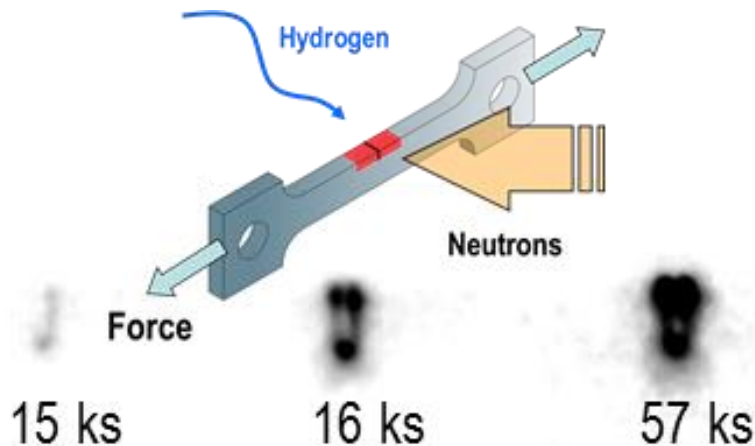


TEM bright field contrast of metal-oxide interface, LK3/L cladding, 7 cycles (BWR / Leibstadt)

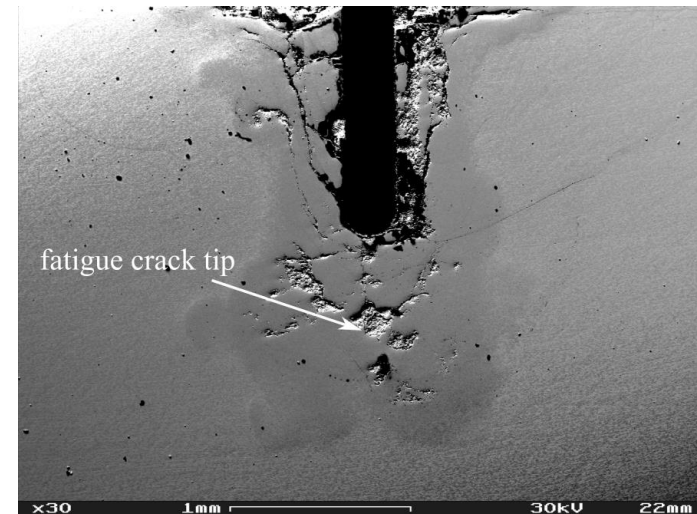




## Neutron Tomo- & Radiography at SINQ / NEUTRA

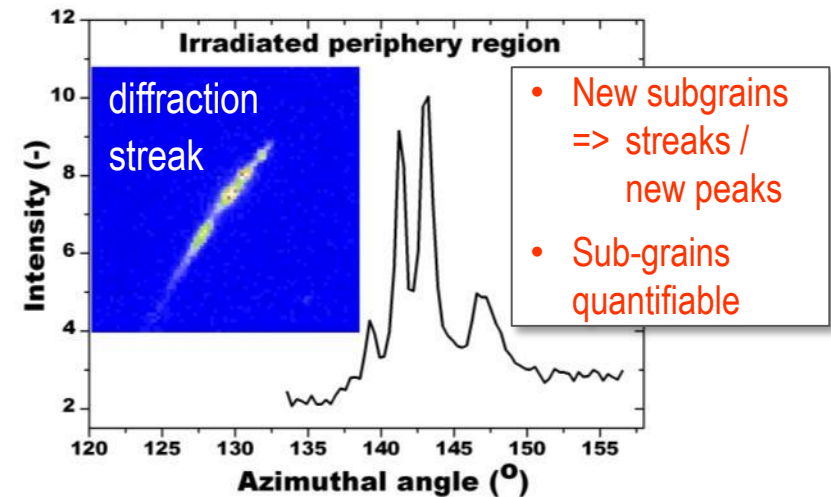
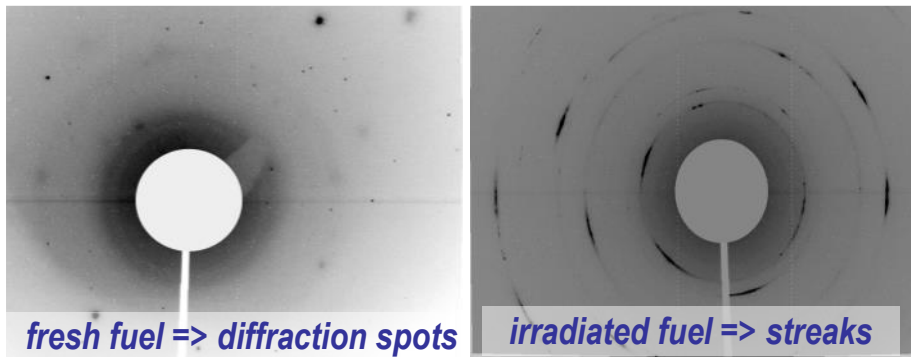


Heavy hydriding at notch root  
BSE SEM image



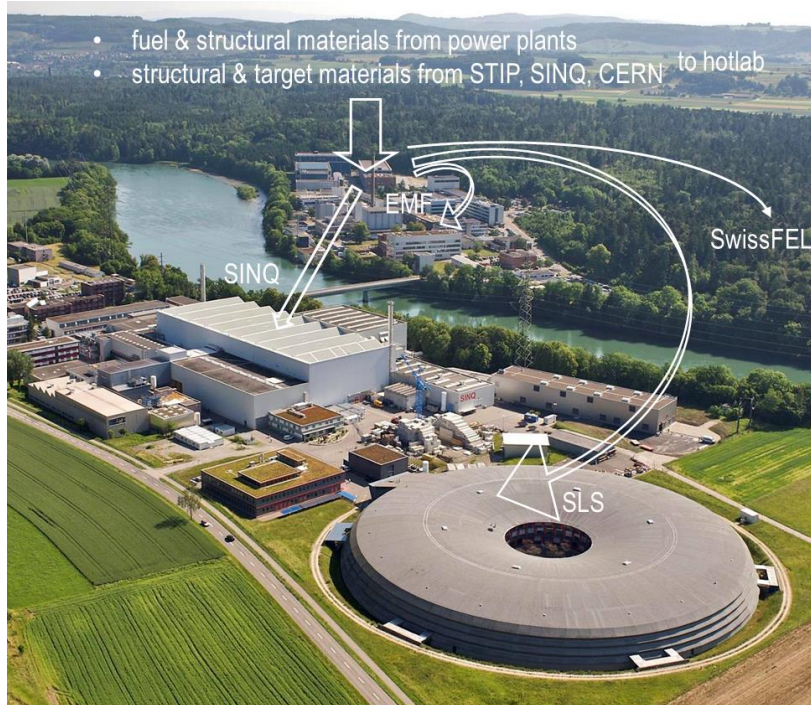
- **Hydrogen uptake & distribution** in **cladding** under **mechanical stress**
- Collaboration with ASQ & KIT,
- Cofund / PSI Fellow project proposal in 2014

- Characterization by PIE and modelling of fuel element behaviour in service
  - HBU, doped fuel, crud, PCI, fission gas retention, transport & release, ...
  - Pre-studies on accident tolerant fuel/cladding and Th fuel for LWRs
- Performance & safety of nuclear fuel depends on microstructure & crystallography.
  - During irradiation fuel changes its structure. Break up of fuel crystallites with increasing burn-up => impact on thermal, physical, mechanical properties.
  - For very tiny fuel particles: **crystallographic changes observed at SLS and quantified.**
  - Collaboration with hotlab (AHL/NES),  $\mu$ XAS beam line (SLS), industry (swissnuclear, Areva), Université Paris-Sud

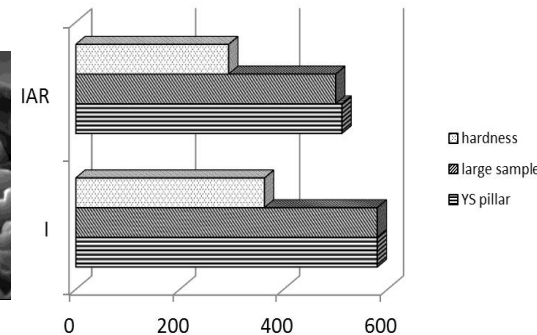
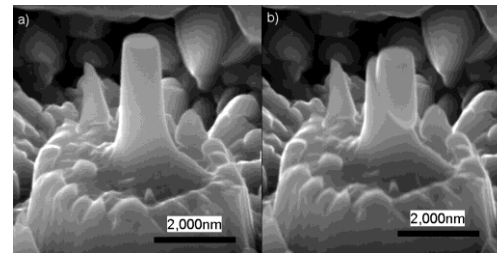
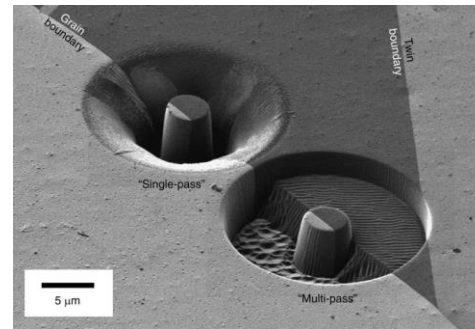


- Practically relevant nuclear material questions with PSI's large-scale facilities
- Leading position with highly radioactive fuel/cladding materials

# Shielded FIB/SEM – A Key Investment of LNM & AHL



$\mu$ -chemical &  $\mu$ -mechanical characterization  
(e.g., GB segregation & strength)

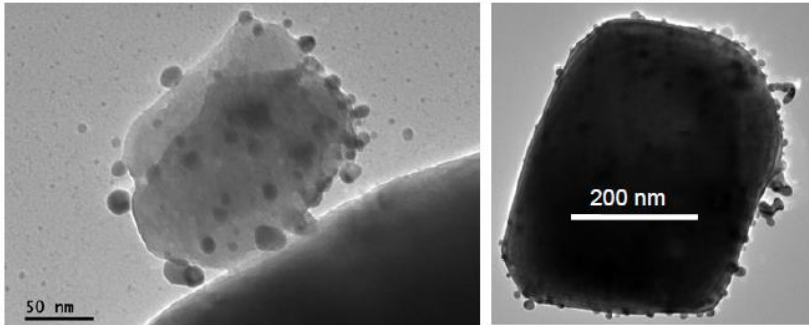


- Joint investment of LNM & AHL of  $\sim 2$  MCHF, SNF-R'equip proposal in 5/2014
- Replacement of 20 y old SEM. Indispensable **workhorse for service work**, e.g., failure analysis
- Targeted & efficient sub-size sample preparation of highly radioactive materials (LWR fuel & reactor internals, spallation & accelerators) for  $\mu$ -chemical, -microstructural, & -mechanical characterizations
- Cost reduction, significant **efficiency, productivity & capacity increase**
- Crucial for the **optimal exploitation of PSI's unique research infrastructure** (hotlab, SLS, SINO, EMF), the **acquisition of additional competitive second part** (e.g., SNF) and **international funding** (e.g., industry), but also for the **attraction of young talents and foreign scientists** and for **visibility**.



## Scientific support of PIE of spent fuel & failure analysis in hotlab (AHL)

### Analysis of fuel crud particles



TEM bright field contrast of CRUD particles decorated with small Pt particles

⇒ Analysis of possible impact of Pt addition on the fuel rods (e.g., on accelerated H uptake or corrosion, increased Crud formation, ... )

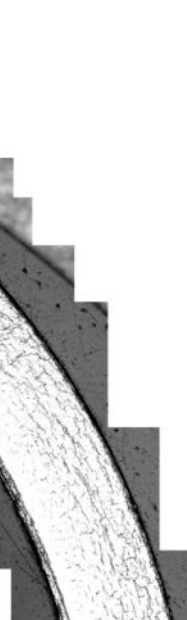
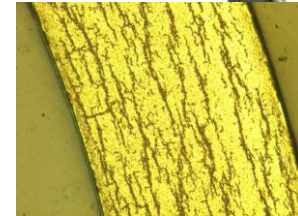
### Quantitative hydride analysis

#### Hydrides analyses

- Complete circumference
- Radial and angular position
- Length
- Orientation

#### Allows investigation of

- hydrides reorientation
- Influence of uneven stress distributions (of interest in case of, e.g., soft versus standard pellets, pellet cracks or oxide layer damage)
- Influence of temperature and fluence (axial position, rod position in bundle)

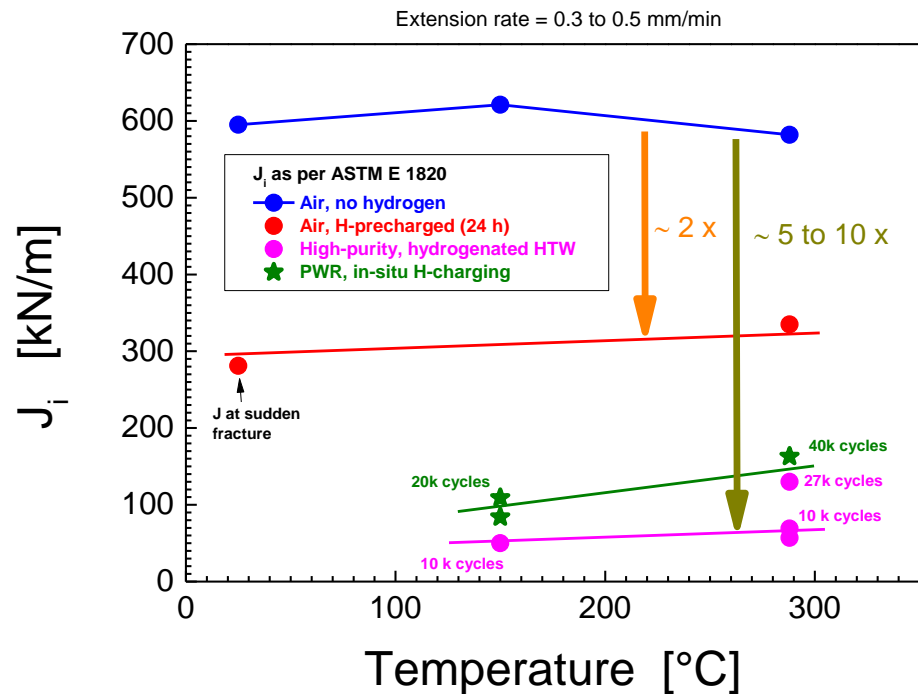
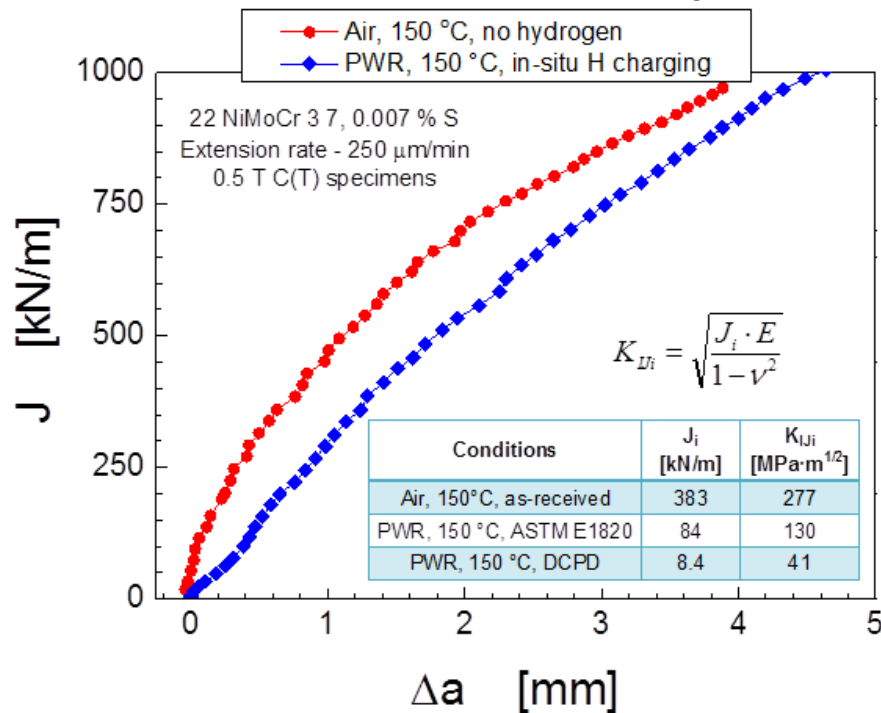




# INTEGER

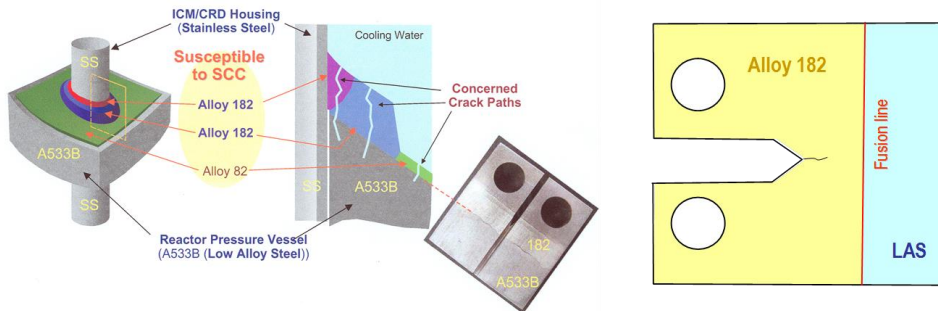
## SAFE project: SCC initiation & growth and environmental effects on fatigue & fracture

Results of first screening EPFM tests in hydrogenated high-temperature water

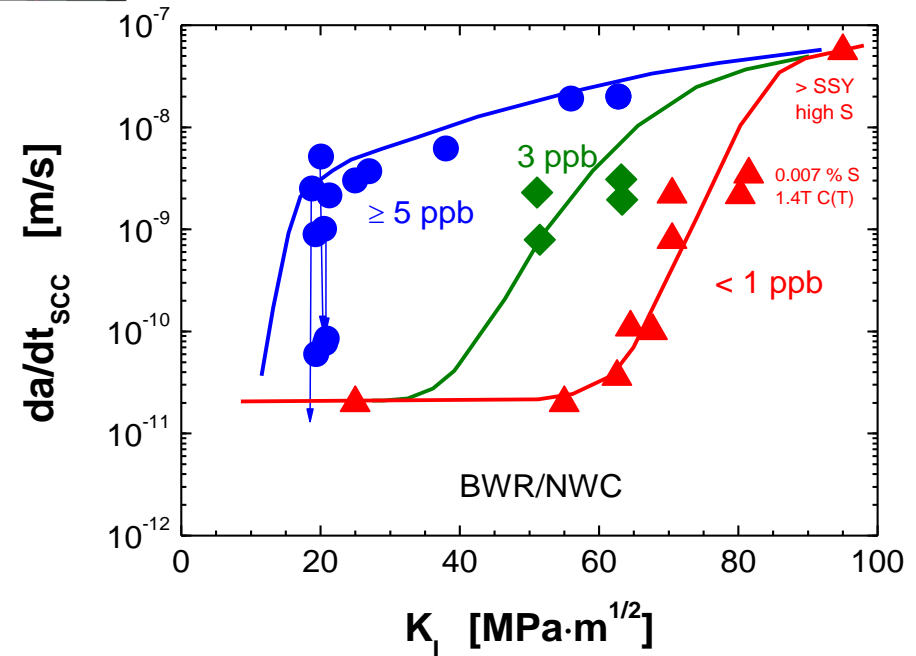
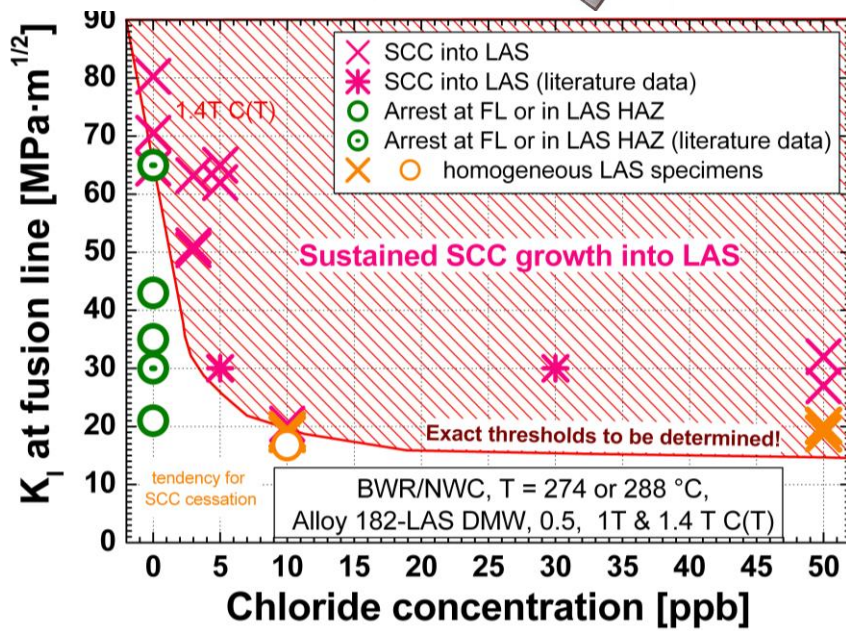


- Significant reduction in initiation toughness (& tearing resistance) in RPV base metal in BWR/HWC & PWR environment compared to fracture toughness tests in air without H
- $J_i$  by DCPD is a factor of ~ 3 lower than  $J_{ic}$  by ASTM E1820
- Systematic tests with variation of T, loading rate, environment, pre-exposure time, etc. including simulated peak hardness coarse grain RPV HAZ materials.

# SAFE: SCC in Alloy 182-RPV Steel DMWs in LWRs



recent SCC incidents in feedwater nozzle DMW in KKL (93 % of wall thickness) & core shroud support DMW of Swedish BWR



- SCC into the LAS cannot be excluded in high-purity BWR/NWC water at  $> 60-70 \text{ MPa}\cdot\text{m}^{1/2}$
- For 3, 5 & 10 ppb of Cl<sup>-</sup>, fast SCC into LAS is possible down to at least 50, 30 & 20 MPa·m<sup>1/2</sup>, respectively.
- Accurate prediction of residual stress profile in DMW (& resulting  $K_I$  at fusion boundary) is crucial!
- Termination of JNES project due to Fukushima → no weld residual stress simulations & measurements

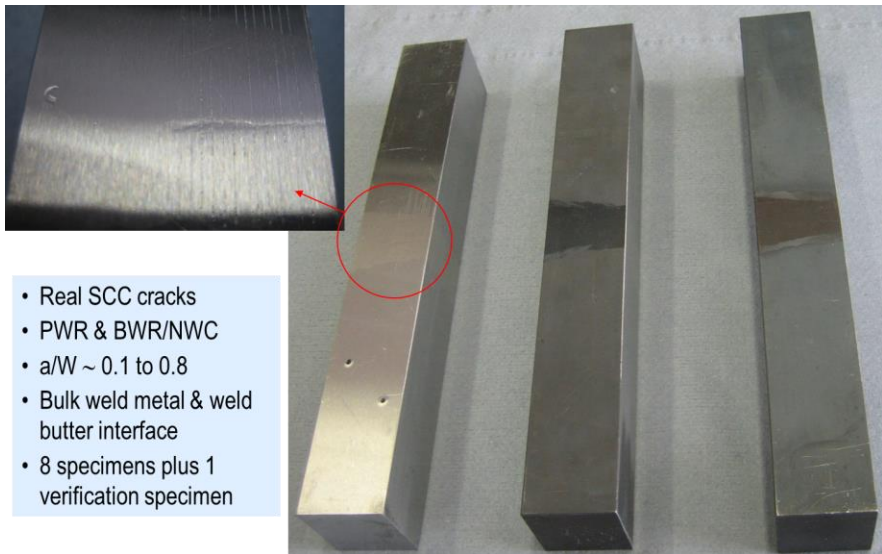
Detection and, in particular, **sizing** of **SCC defects in DMWs** represents a **challenge** and is related to **relevant uncertainties**. Crack depth is often significantly underestimated by NDT!

**PARENT: Program to Assess the Reliability of Emerging Nondestructive Techniques**

follow-on project to **PINC: Program for the Inspection of Nickel Alloy Components**

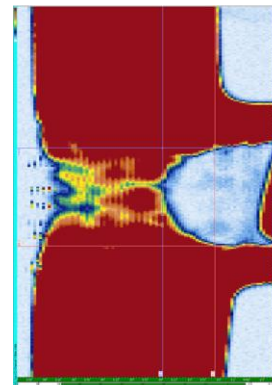
Participation of **Swiss consortium** (ENSI, PSI, ALSTOM, SVTI, EMPA) in **PARENT-Project**

- International program including regulators, industrial groups and research institutions
- Assessment & quantification of established & new promising NDE techniques
- NDT tests bodies with well characterized SCC cracks for open round robin as PSI contribution
- Participation in open and closed round robin programs (ALSTOM, SVTI)



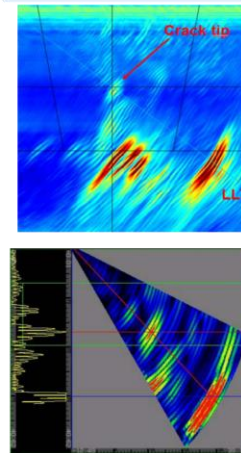
- Real SCC cracks
- PWR & BWR/NWC
- $a/W \sim 0.1$  to  $0.8$
- Bulk weld metal & weld butter interface
- 8 specimens plus 1 verification specimen

UT immersion  
Alstom

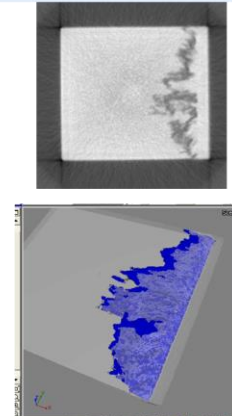


BWR/NWC SCC Crack

UT phased array  
SVTI, Alstom



X-ray radiography & tomography  
EMPA

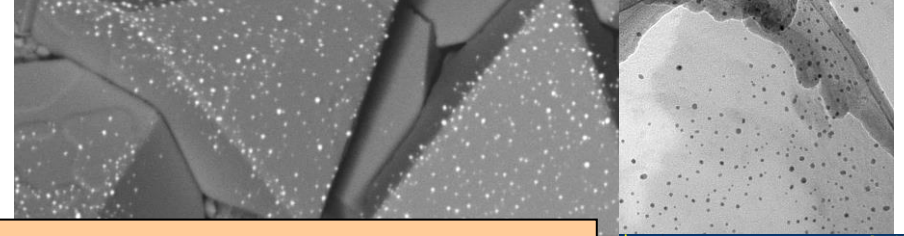
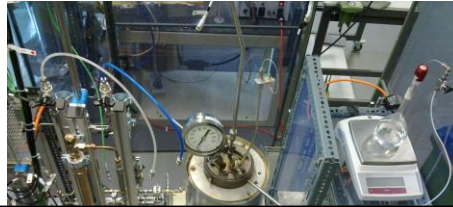




# NORA-I & -II - On-line Noblechem in BWRs

Systematic Pt deposition tests in loops & autoclaves with flat & pre-cracked specimens

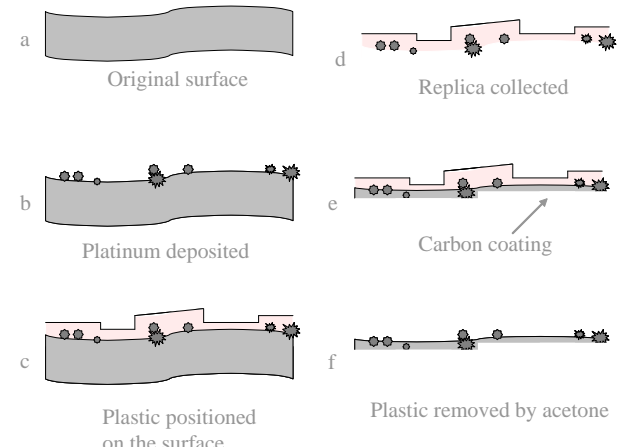
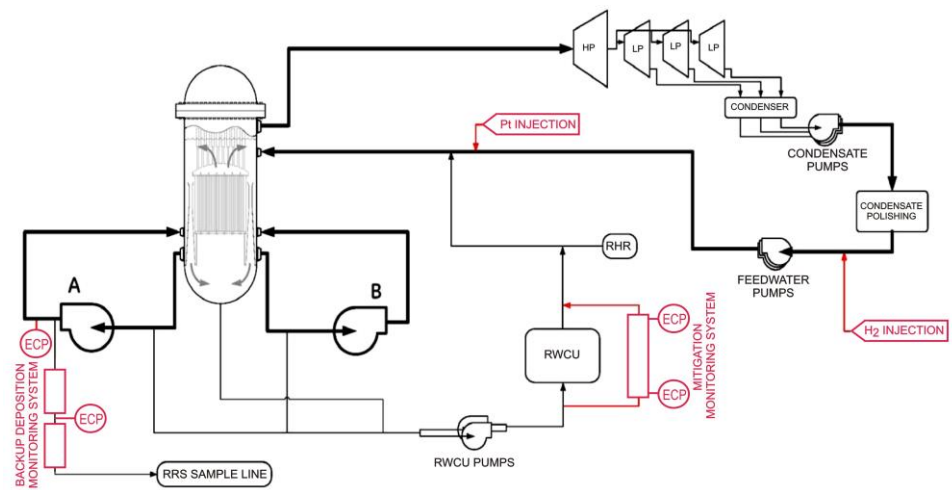
Characterization (size, distribution, conc.) by LA-ICP-MS, FEG-SEM/EDX & TEM/EDX/EELS



**PSI, ENSI, KKL, KKM & collaboration with EPRI BWRVIP**  
**Results of direct practical use (optimal OLNLC application)**

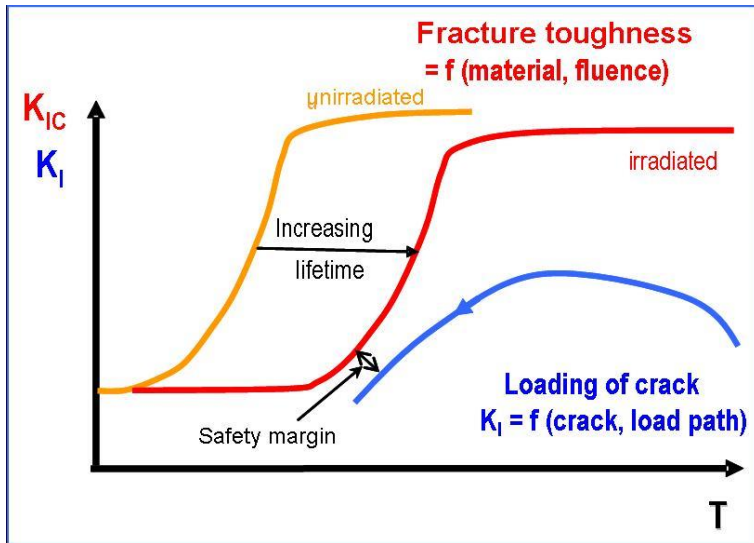
Exposure of specimens in Mitigation Monitoring System & Reactor Water Sample Line at KKL

Development of a replica-based ND technique for radioactive components



Several additional side projects: YUMOD (KKL), optimization of OLNLC injection system (KKL), Pt-analysis on dry tubes (EPRI/KKL), crud (KKL), monitors (Spanish BWR), crevice monitors (KKM), EPRI/PSI OLNLC projects, ...

## Deterministic



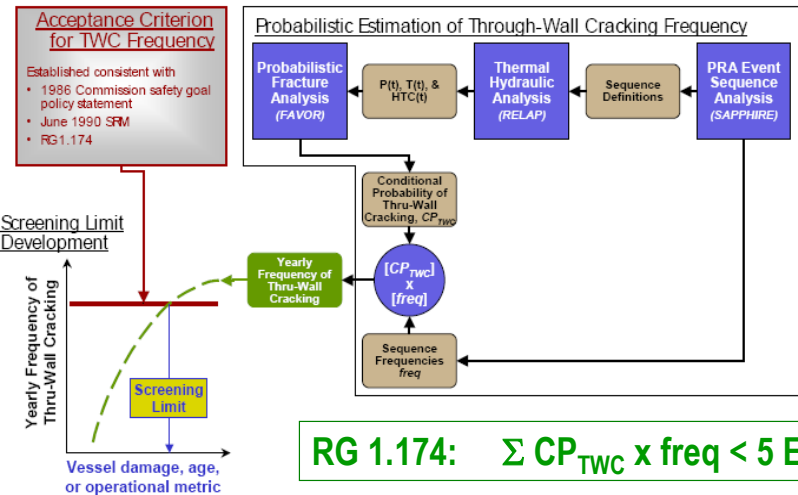
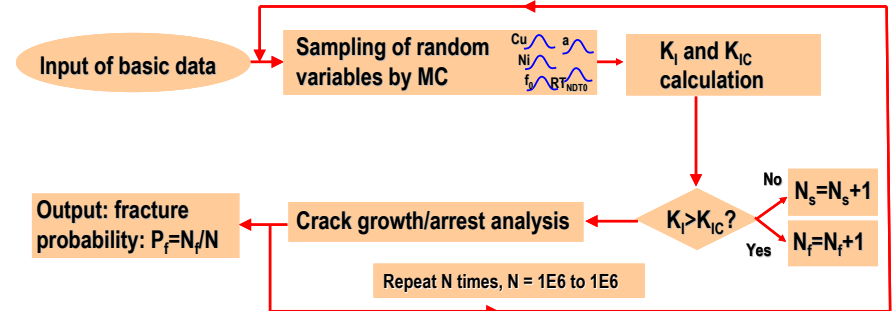
For postulated cracks & all operating & selected bounding accident (e.g., PTS) conditions

$$K_I = f(a, \text{load}) < K_{IC} \text{ or } K_{Ia}$$

Operating conditions  $\rightarrow a = 1/4$  of wall thickness

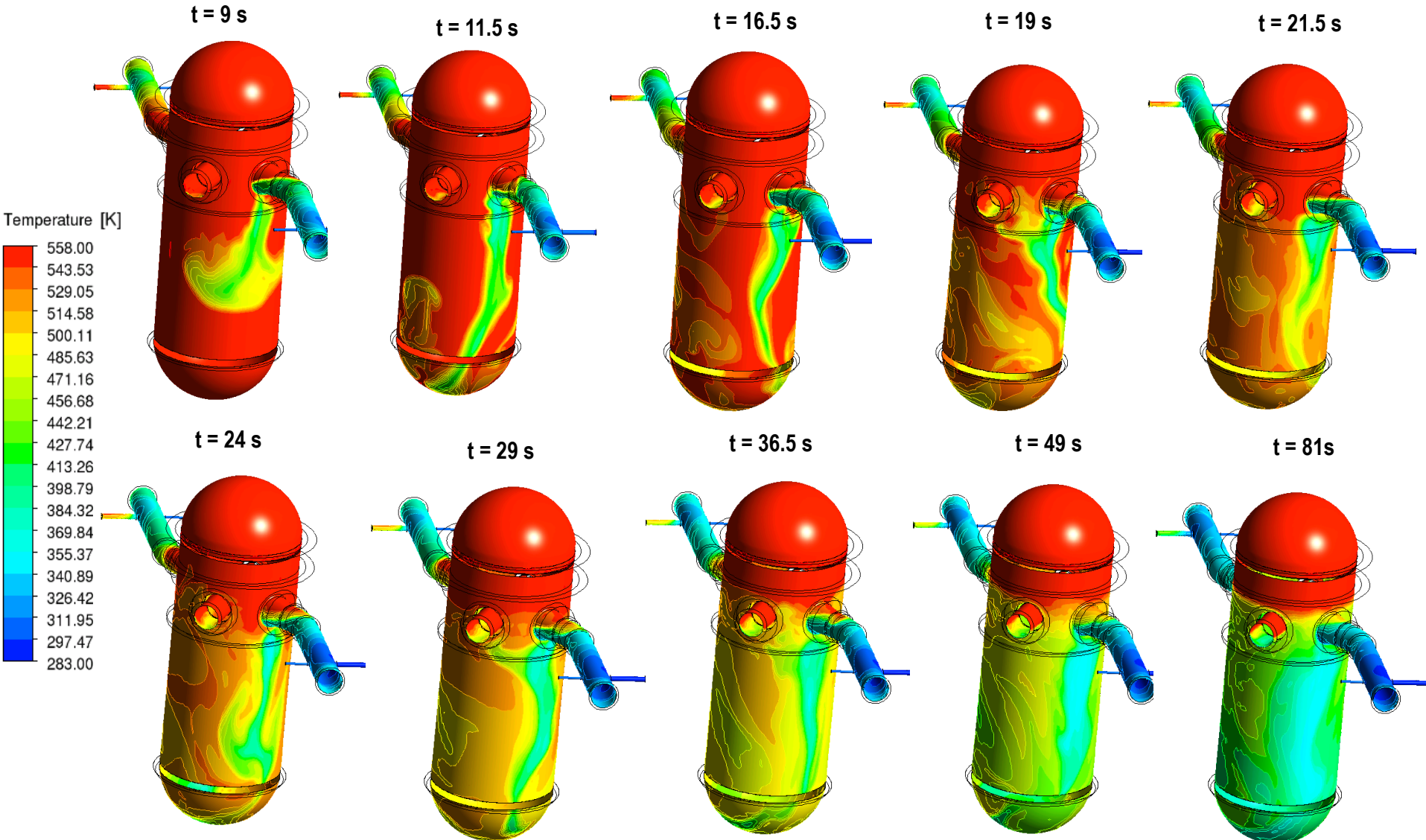
Accident conditions  $\rightarrow a = 2 \times$  NDT resolution limit

## Probabilistic

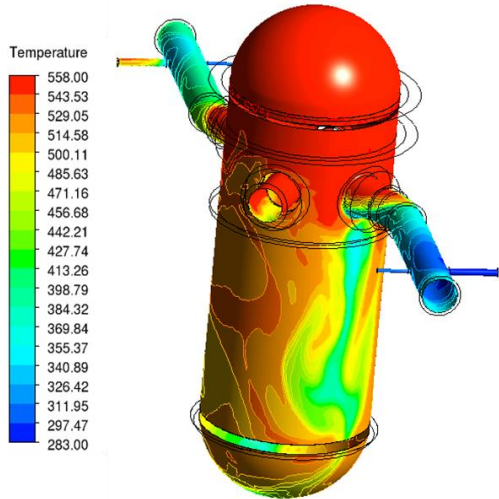


- Identify accident scenarios & estimate their frequencies
- Estimate RPV failure probability for different transients
- Estimate total RPV failure frequency
- Consideration of random & lack of knowledge uncertainties
- Better estimation of safety margins

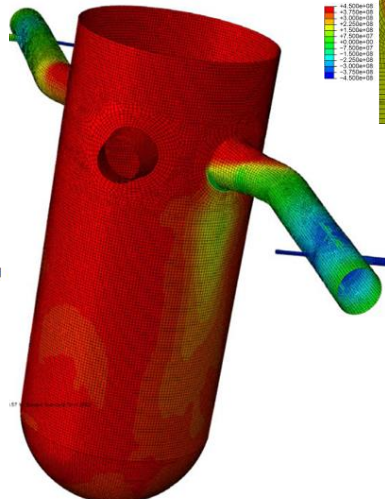
## 70 cm<sup>2</sup> MLOCA transient with cold water injection in one loop only



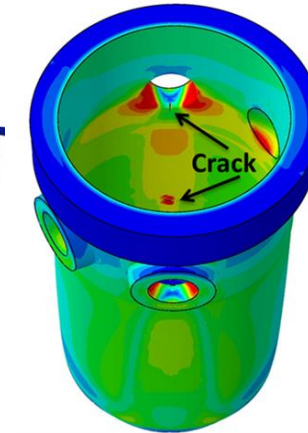
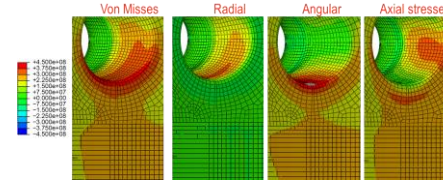
## Fluid T



## Wall T



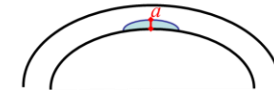
## Stress $\sigma$



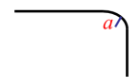
## Crack configurations

- axial & circumferential cracks
- at nozzle corner & cylindrical shell
- inside & outside the plume region
- surface & underclad cracks
- base metal or weld

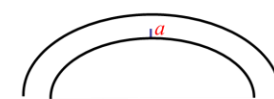
Wall circumferential crack



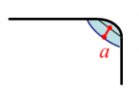
Inlet nozzle circumferential crack



Wall axial crack

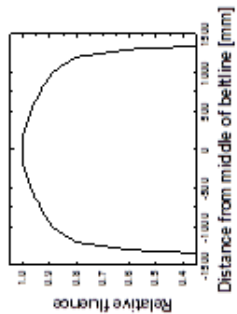


Inlet nozzle axial crack



## Fluence

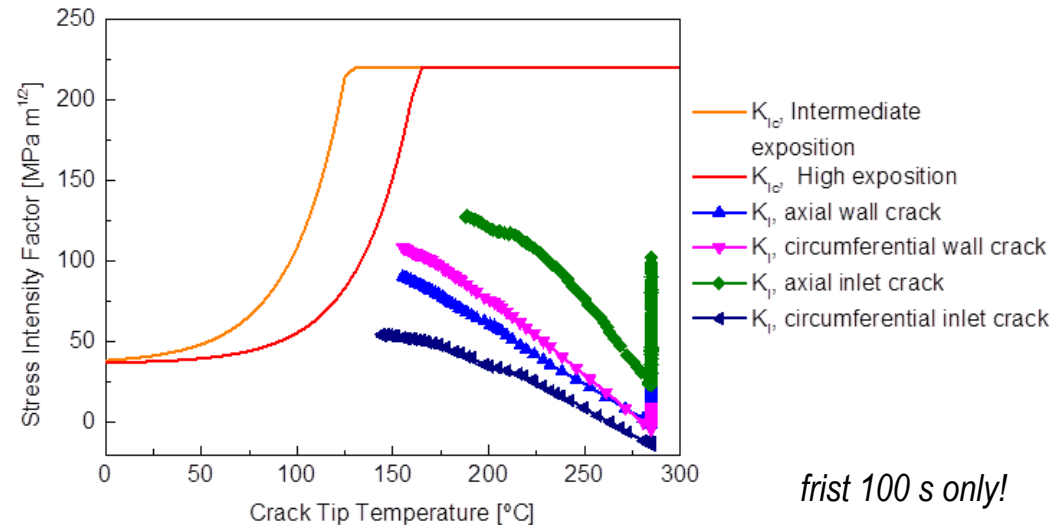
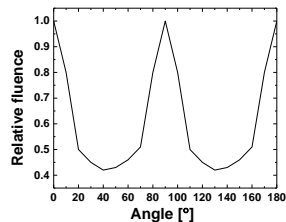
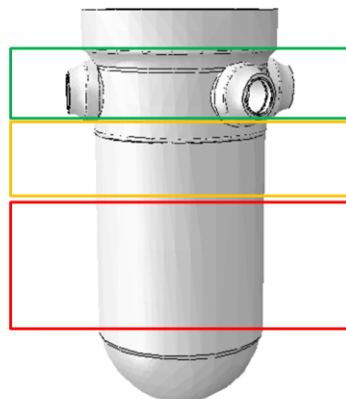
= f(location, operation time)



Low fluence

Intermediate fluence

High fluence



*frist 100 s only!*