Structural Materials Challenges for ESNII Reactors

L. Malerba, EERA-JPNM coordinator
CIEMAT, Spain
lorenzo.malerba@ciemat.es

www.eera-set.eu

EERA is an official part of the EU SET-Plan.

http://setis.ec.europa.eu/

EERA
European Energy Research Alliance
JPNM
JOINT PROGRAMME ON NUCLEAR MATERIALS

Part I Introduction




What can GenIV reactors do for you?

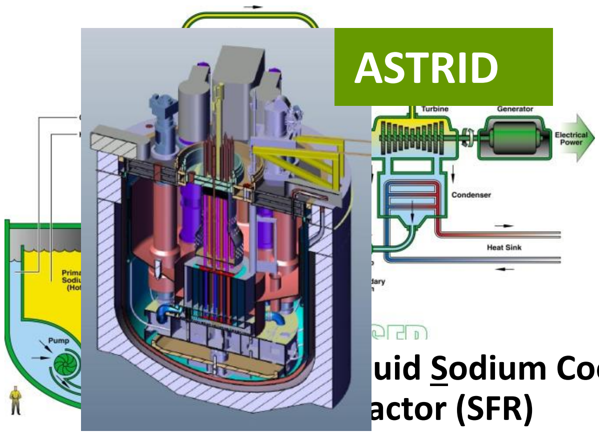
- **Produce more fuel than they use → recycling ensures energy for centuries (or millennia)**, recycling on-site increases proliferation resistance

- **Burn nuclear wastes while producing energy → minimisation of quantities, reduction of hazard**, wastes will still exist, but less, less dangerous and of much shorter life

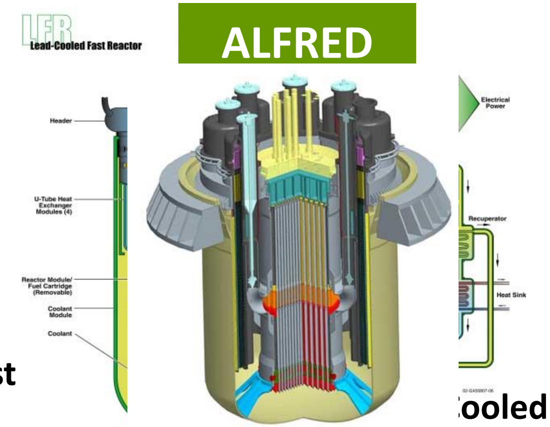
- **Work at higher temperature → be energetically more efficient**, allow use of gas turbines instead of steam turbines, produce industrial heat …

- **Use passive safety systems** *(based on physical laws rather than human/computer intervention)* → **be safer**

Sustainable nuclear energy:
GenIV reactors and fuel cycle facilities in Europe

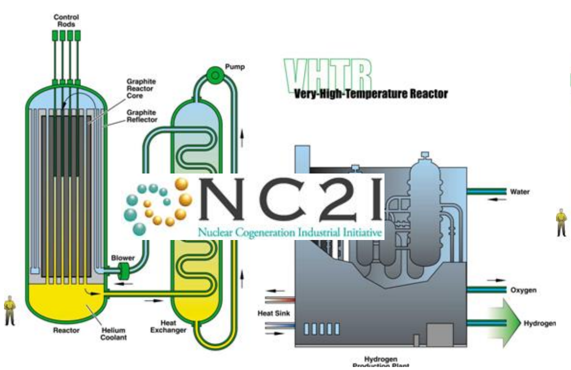


uid Sodium Cooled Fast
actor (SFR)

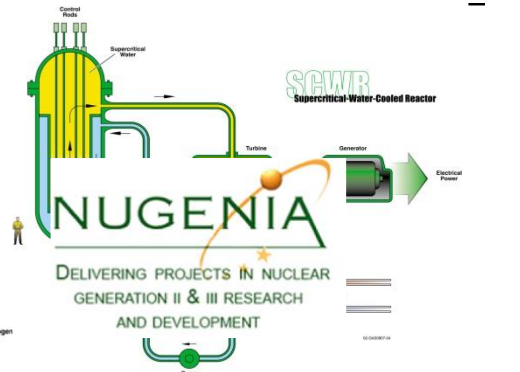


ooled
Fast Reactor (LFR)

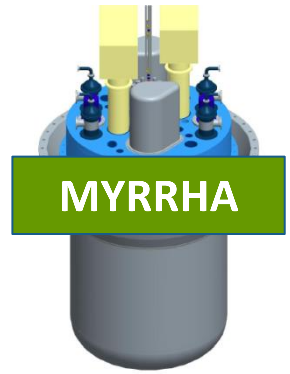
Fast
FR)



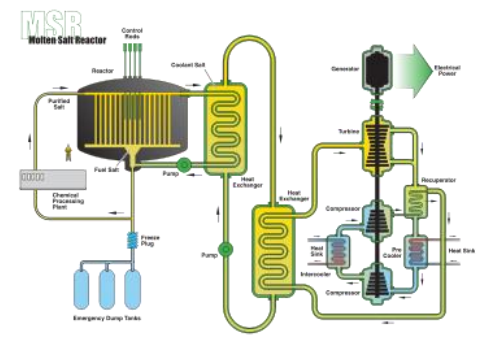
(Very) High Temperature Reactor (HTR)



SuperCritical Water Cooled Reactor (SCWR)



Accelerator Driven System (ADS)

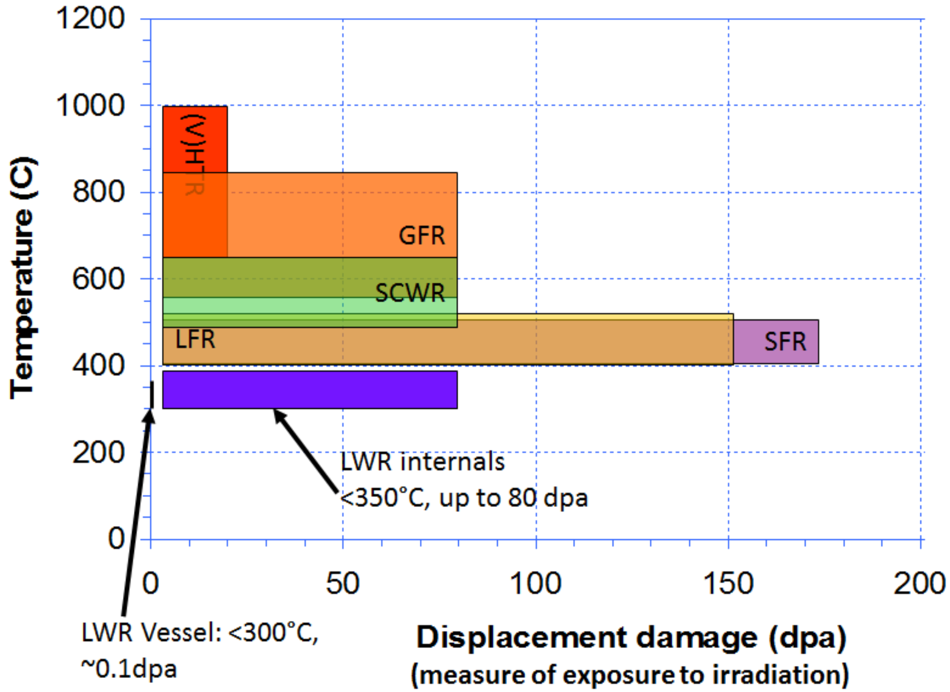


Molten Salt Reactor (MSR)

European Sustainable Nuclear Industrial Initiative ESNII concepts

But materials pay the price …

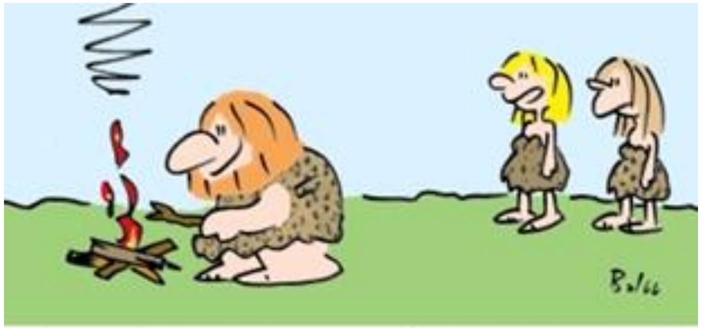
- GenIV reactors need to
  - **Push the burnup of fuel as high as possible → very high irradiation dose (0.1 → >100 dpa)**
  - **Push the operating temperature as high as possible → from ~300°C to 500-1000°C**
  - **Use coolants different from water**: liquid metals (Na, Pb, PbBi, …), gases (He), *molten salts, supercritical water* → **problems of corrosion, erosion, dissolution …**



Materials are one of the main bottlenecks for sustainable nuclear energy

GenIV nuclear materials are a tough challenge

- Radiation resistance
  - Swelling / Irradiation creep
  - Low temperature embrittlement
- High temperature-resistance
  - Creep strength
- Compatibility with
  - (Heavy) liquid metals (Na_{liq}, Pb_{liq}, PbBi=LBE)
  - Gas (He)
  - Super-critical water
  - Molten salts



"Well, I guess we'd better get started inventing fire-resistant fabrics."

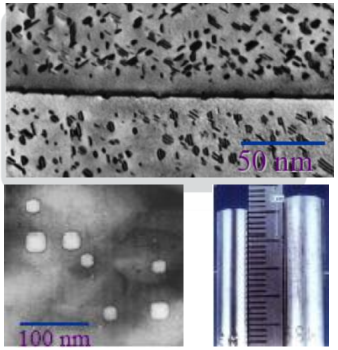
Qualify existing materials
Understand processes
Improve materials properties

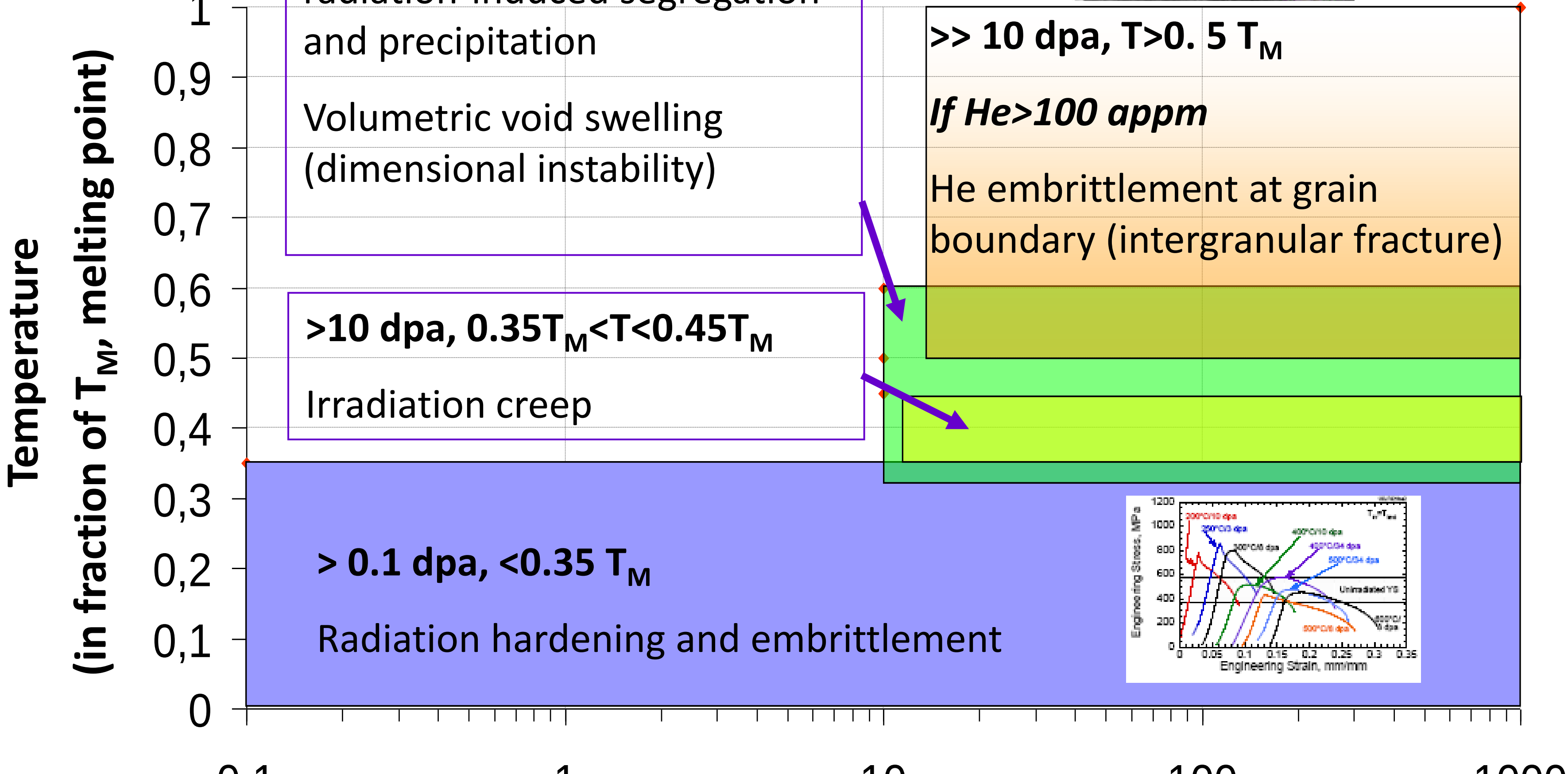
Part II

Degradation mechanisms



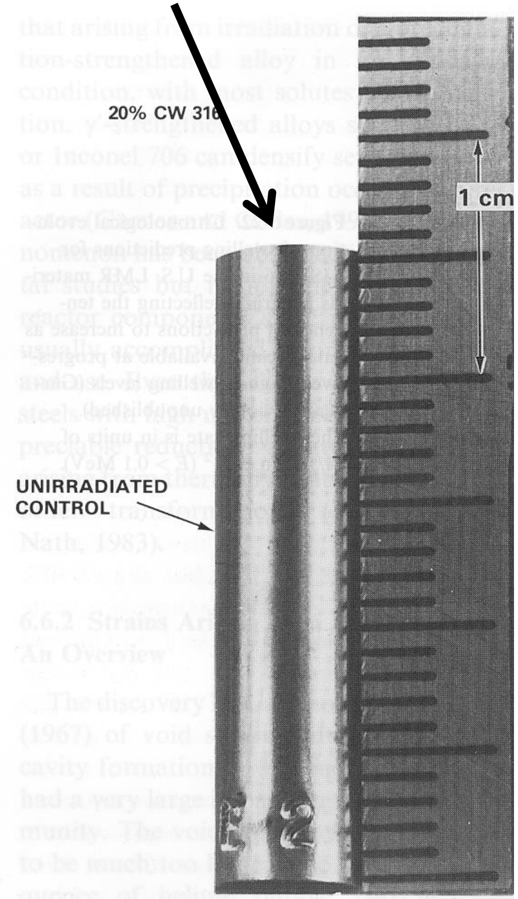

Radiation effects in structural materials depend on temperature and dose





What is swelling?

Before irradiation

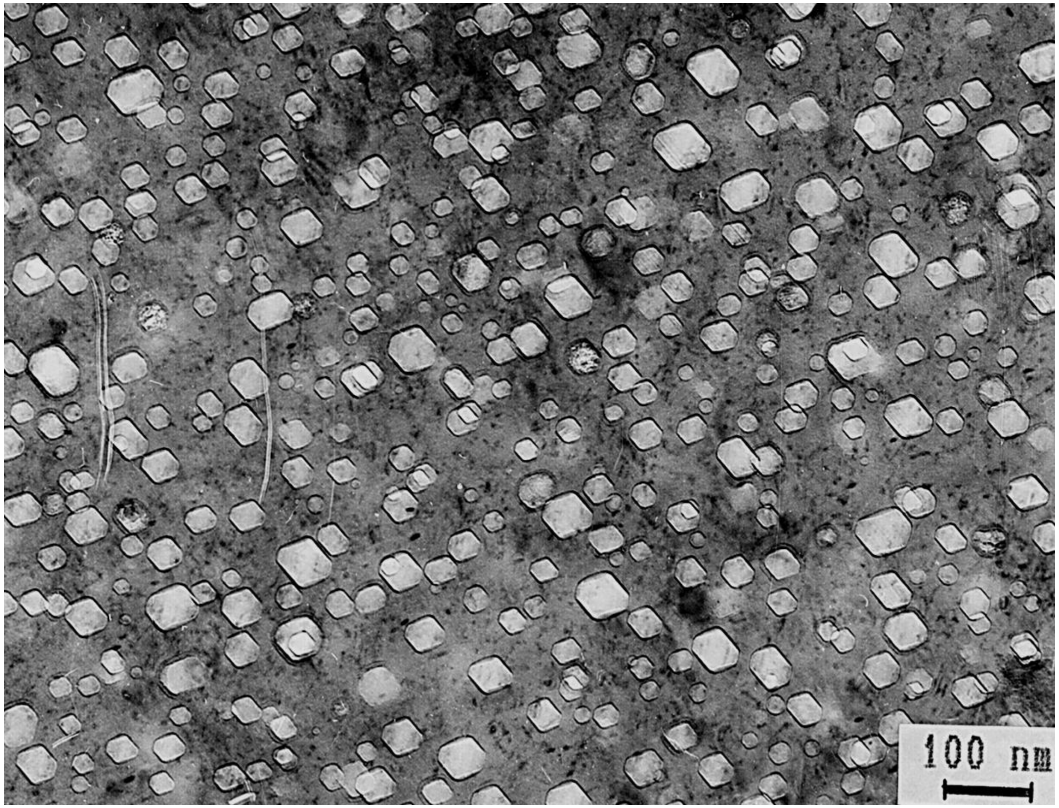


- Hardly any swelling is expected in current light water reactors (LWRs)
  - ‘low’ temperatures
- Effect will be important in GenIV reactors
  - temperatures of 400°C to 700°C
  - flux ≈ 100 flux LWR

Swelling is the consequence of radiation-induced removal of atoms from bulk & creation of voids

Voids observed in stainless steel after 73 dpa at 335ºC in BN-350 sodium fast reactor (Kazakhstan)

Porollo, Konobeev, Garner, 2000

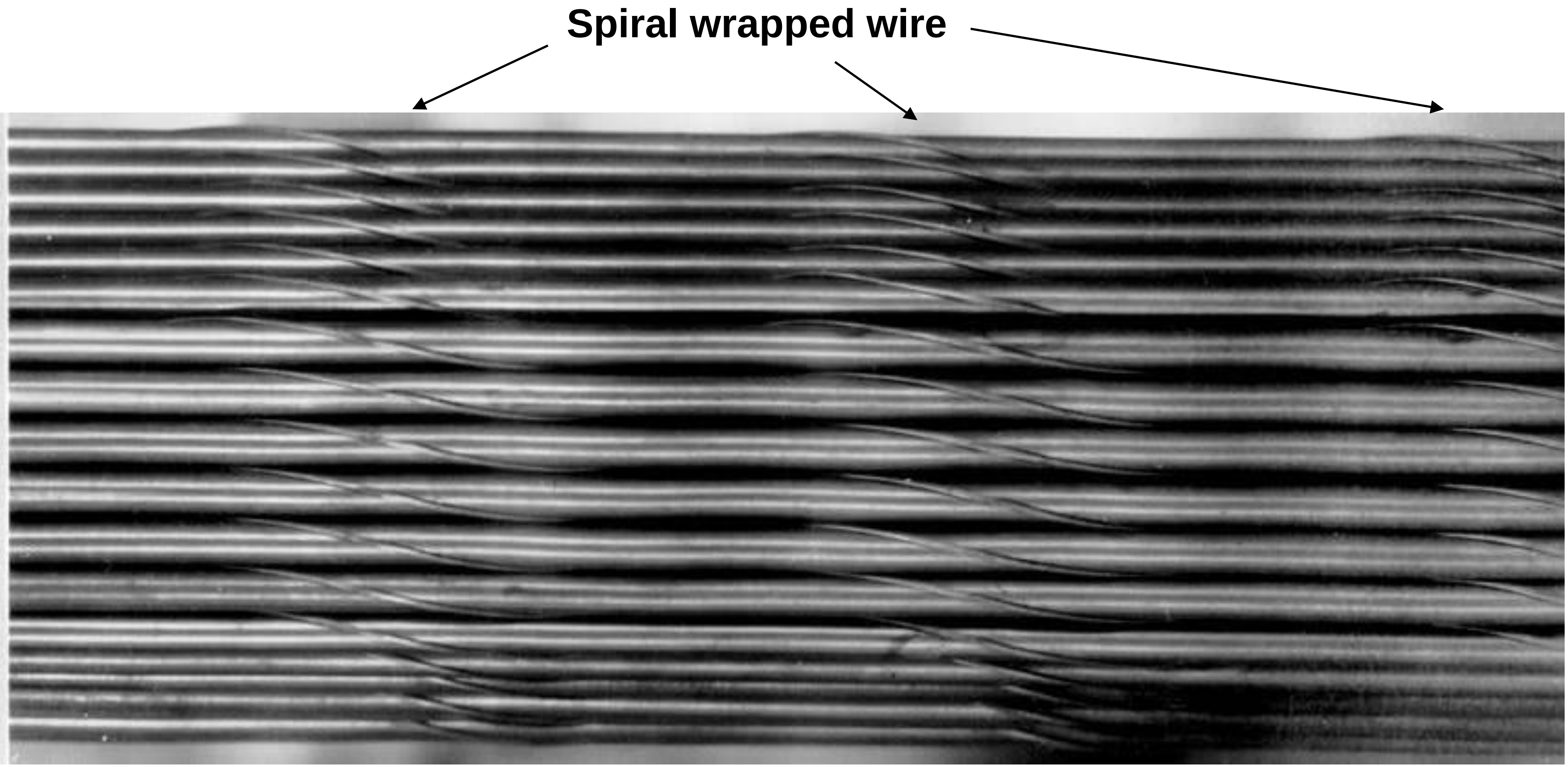


1.4×10^{-6} dpa/sec

6.2% swelling

Consequences of swelling can be serious

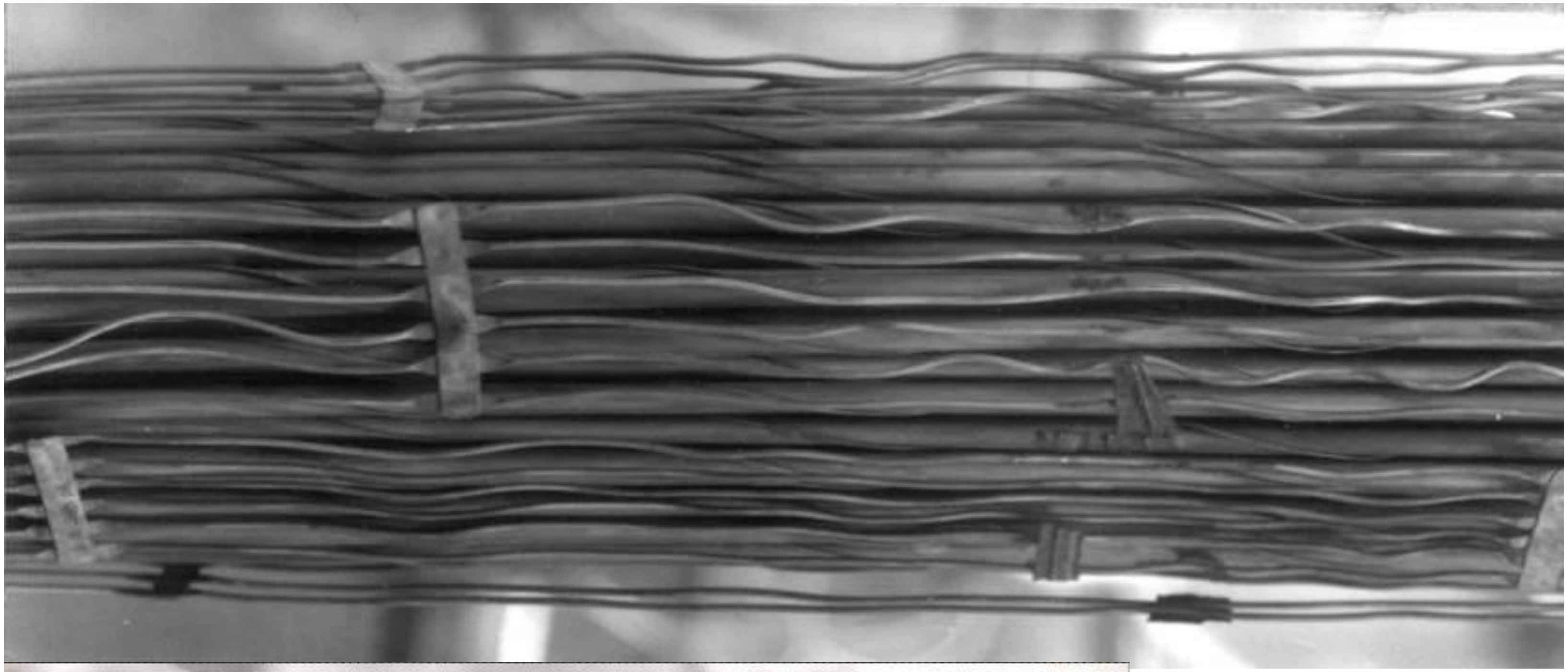
Annealed wire wrap spacer used on fuel pins assemblies in BN-600 (Russian sodium-cooled fast breeder reactor**) – *before irradiation***

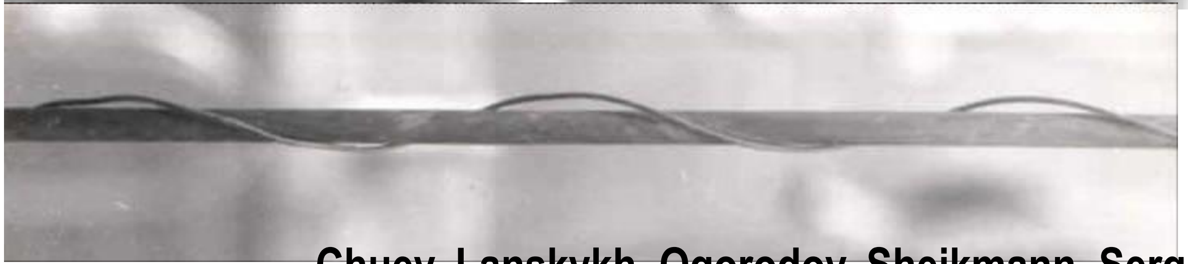


Chuev, Lanskykh, Ogorodov, Sheikmann, Sergeev, 2004

Consequences of swelling can be serious

Annealed wire wrap spacer used on fuel pins assemblies in BN-600 (Russian sodium-cooled fast breeder reactor**) – *after irradiation***

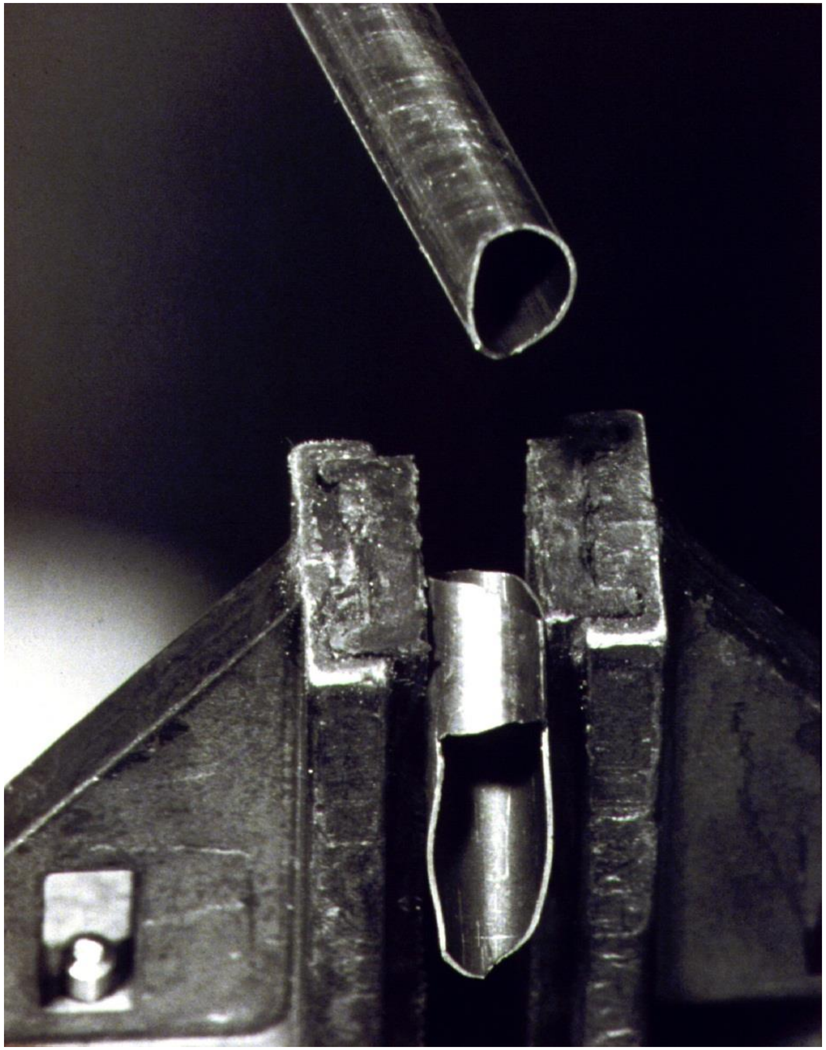




Wire swelled more than cladding

Chuev, Lanskykh, Ogorodov, Sheikmann, Sergeev, 2004

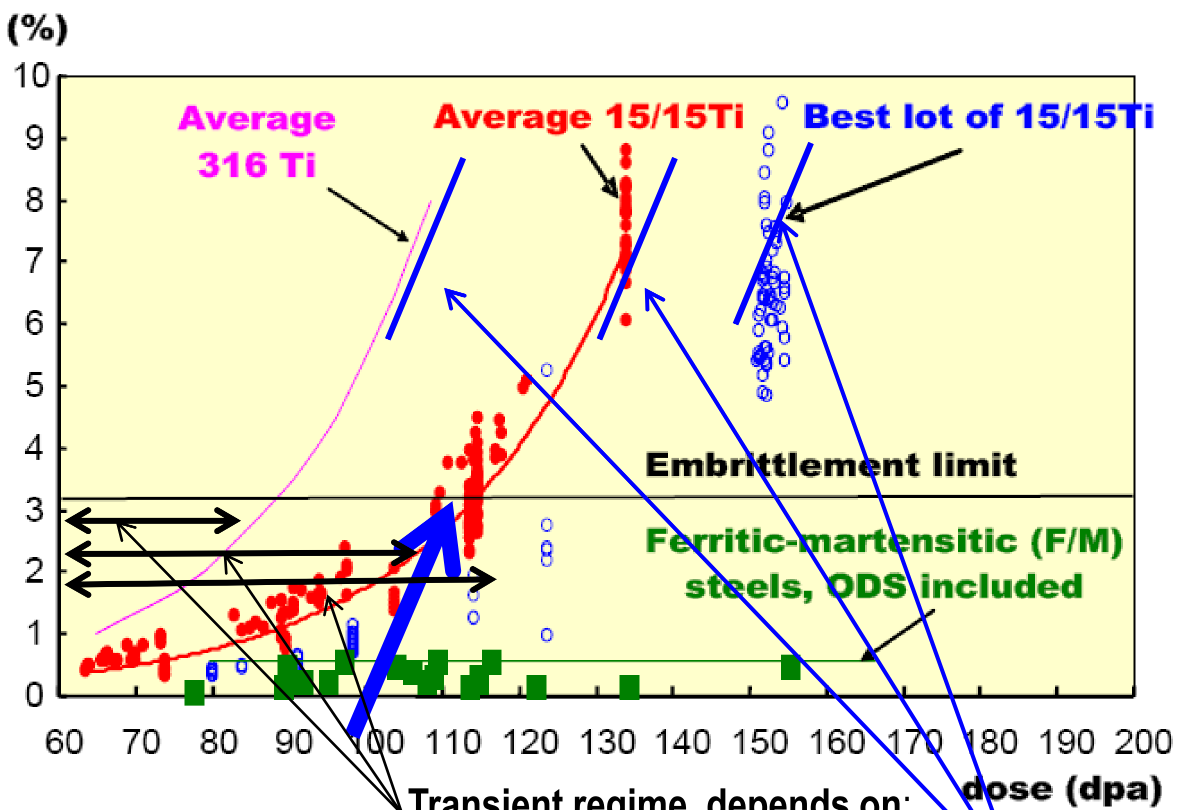
Another serious consequence of swelling: Void-induced embrittlement



Porter and Garner, 1988

- 14% swelling in 316 austenitic stainless steel irradiated at ~400ºC
- Failure occurred during clamping at <u>room temperature.</u>

Austenitic steels are especially prone to swelling, ferritic/martensitic steels resist much better



Incubation dose

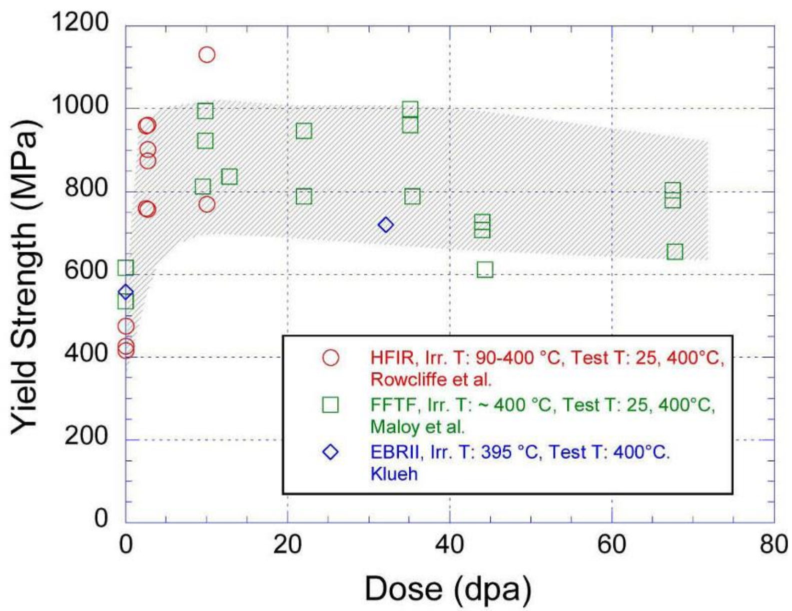
Transient regime, depends on:
Material and composition
Phase distribution
Temperature
Dpa-rate
Stresses

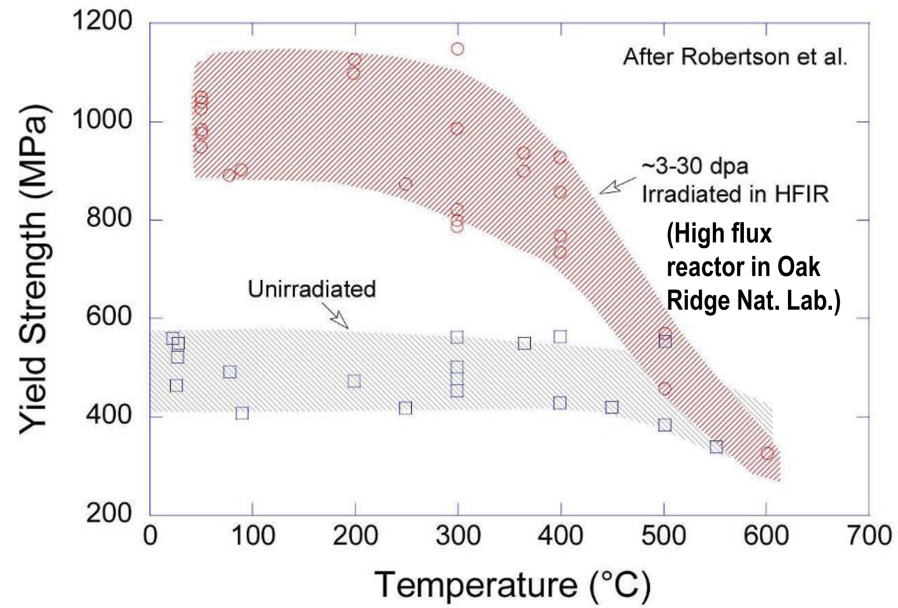
Steady-state swelling rate:

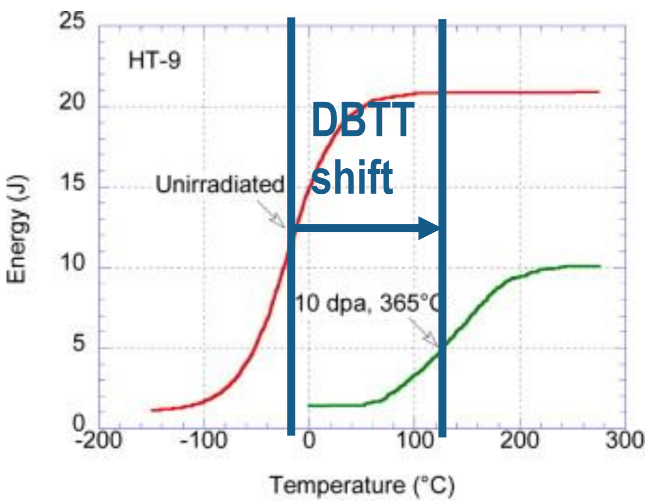
Almost material's constant eventually always reached in steels: 1%/dpa (austenitics); 0.2%/dpa (F/M)

- **Ti-stabilised austenitic steels exhibit increased incubation dose**
- **Ferritic/martensitic (F/M) steels start to swell only in excess of 180 dpa**

"Low temperature" embrittlement is "the" limiting issue for F/M alloys





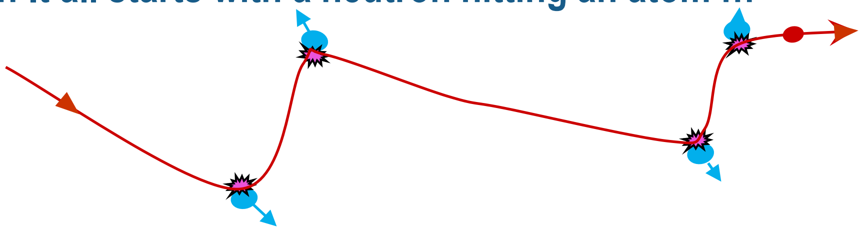


Y. Chen, Nucl. Eng. & Technol. 45 (2013) 311

- **When irradiated below 400°C the yield strength increases significantly**
- **This leads to shifts of the ductile-brittle trasition temperature (DBTT) -~120°C in the example**

Embrittlement is caused by nanometre-scale defects that form under irradiation

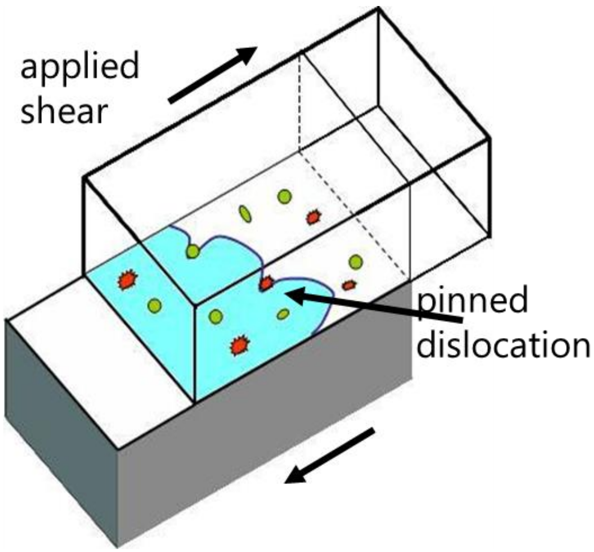
… It all starts with a neutron hitting an atom …

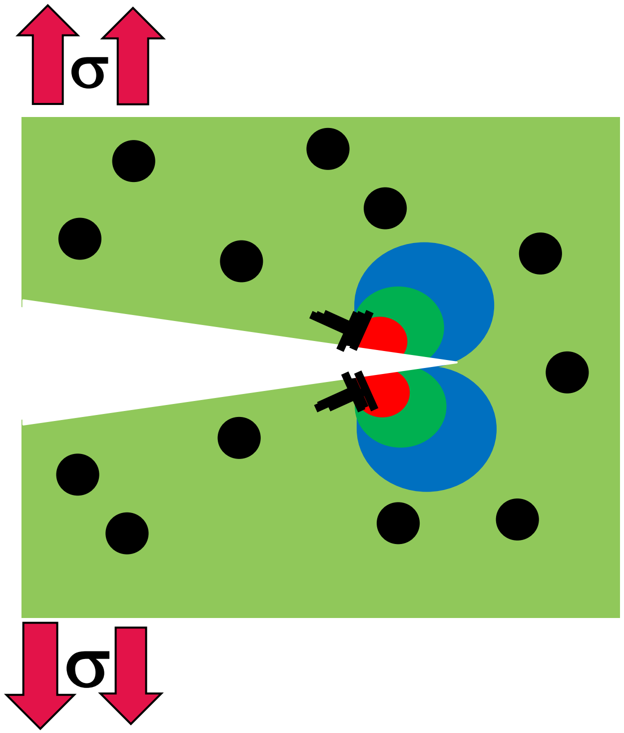


Displacement cascade

Migrating defects

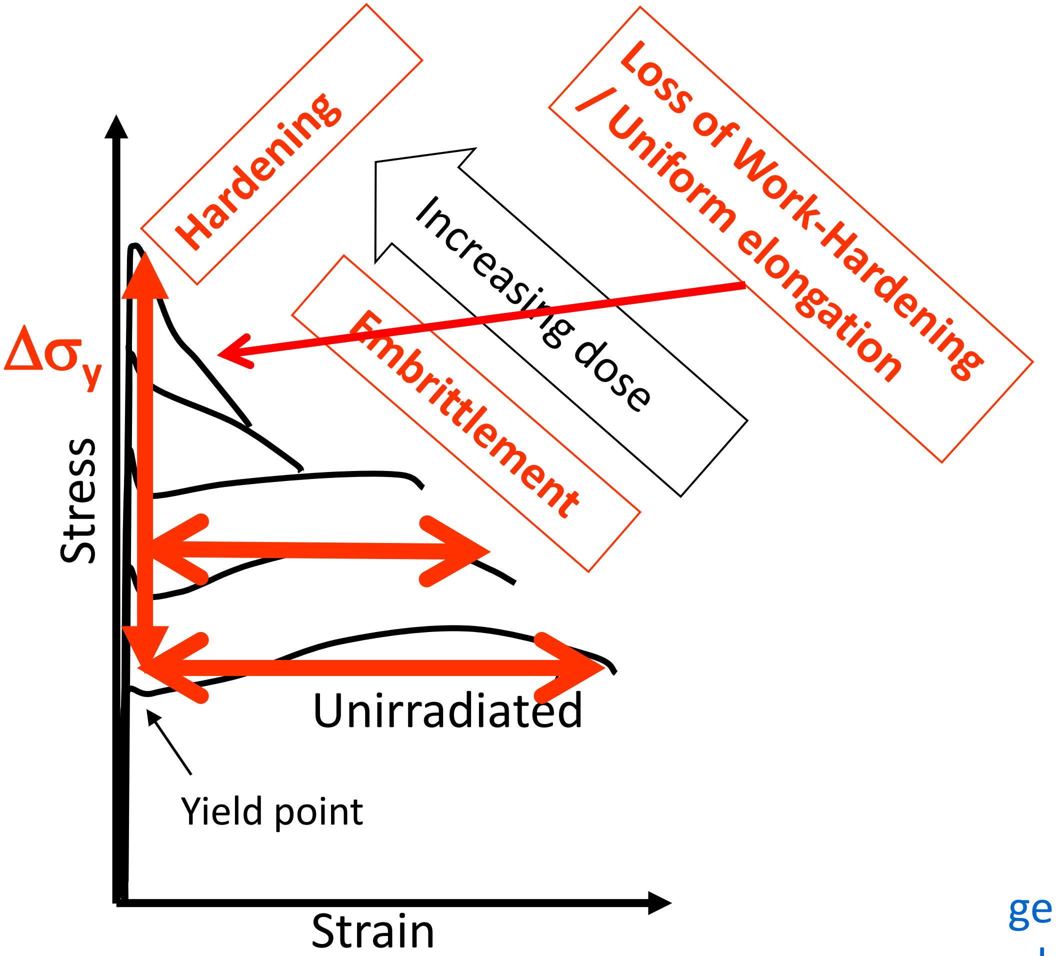
Radiation-hardening





Radiation-embrittlement *is generally a consequence of hardening*

Hardening, embrittlement but also loss of work-hardening / uniform elongation



Zinkle & Singh, J Nucl Mater 351 (2006) 269

When loss of elongation happens, generally bands clear of defects in which plastic flow localises are observed in the electron microscope

Operating temperature for nuclear materials is limited by thermal creep

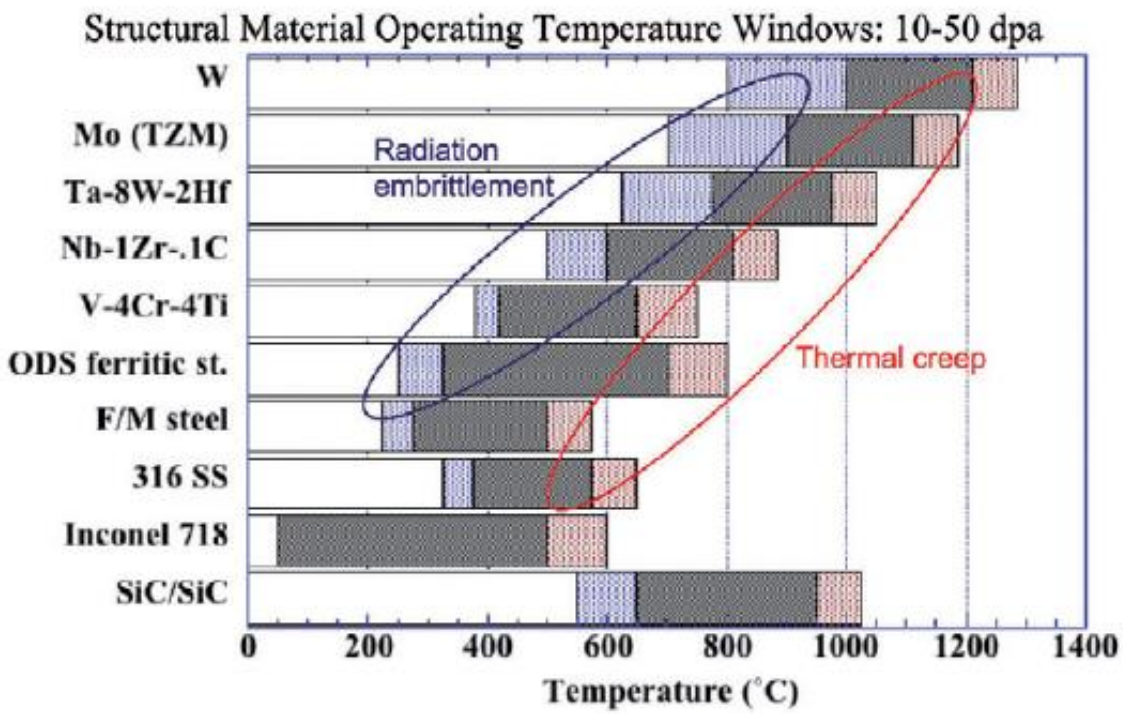
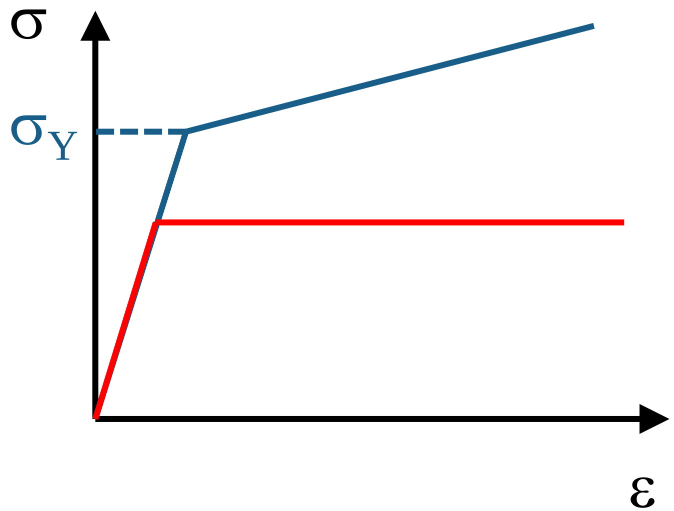


Fig. 5 Estimated operating temperature windows (dark shaded region)[30,79] for structural materials in nuclear energy systems for damage levels of 10 to 50 dpa. The light blue and red regions represent lower and upper temperature uncertainty bands.

S.J. Zinkle, J.T. Busby, Materials Today 12 (2009) 12

Lower bound:
Low T embrittlement

Upper bound:
Thermal creep



Routes to increase high temperature resistance in steels and metallic alloys in general

- Tune composition to:
  - Increase solution strengthening
  - Obtain more favourable volume fraction of phases (thermodynamic modelling)
    - Delay/avoid Laves phase formation &/or their coarsening
    - Delay carbide coarsening
  - Optimize thermo-mechanical treatments to refine carbides and their distribution
- Introduce strengthening inclusions
  - Oxide dispersion strengthening (ODS)
    - Powder metallurgy
- For nuclear
  - Achieve this with *neutron-compatible elements* (activation)
  - Take into account *alloying element influence on behaviour under irradiation* (low T embrittlement, swelling, …)

… and for very high temperatures? Ceramics!

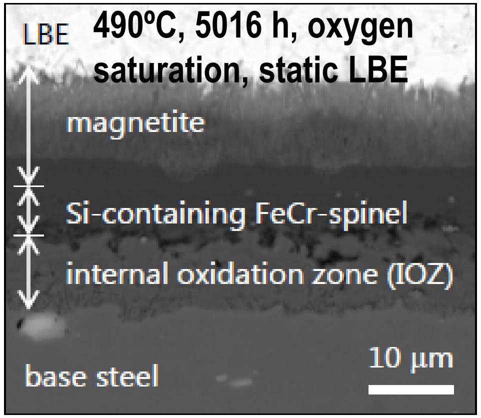
- When the operating temperature is expected to be in the range 800-1000°C or above
  - Use of ceramics becomes necessary*
    - Graphite (VHTR)
    - SiC/SiC (GFR, VHTR, but also cladding for GenII/III, SFR, LFR …)
    - Max phases
    - Ceramic thermal barriers (Al_2O_3)
  - Problems
    - Inherent brittleness / pseudoductility
    - Difficult to establish safe design rules
      - Yet graphite has been used for a long time in GenI/II NPP

*Ni-based alloys are not suitable as core material: He production from Ni, swelling and especially embrittlement

EERA JPNM
JOINT PROGRAMME ON NUCLEAR MATERIALS

Corrosion mechanisms in liquid metals

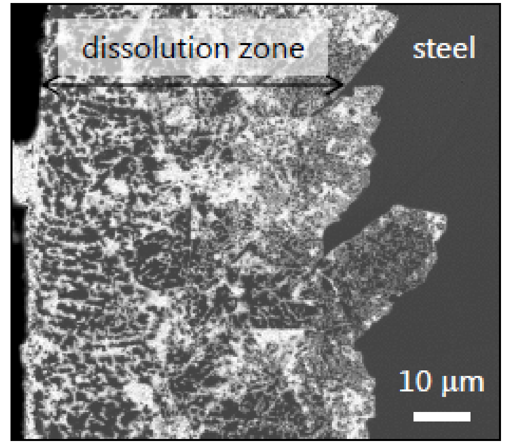
1. Oxidation



- Multi-layered oxide scales form in contact with O-containing LBE on steel surface
- If protective at service conditions, oxide scales minimize further attack of steel by LBE

Variables: Steel type, microstructure & composition, type of fluid, oxygen content, exposure temperature and time

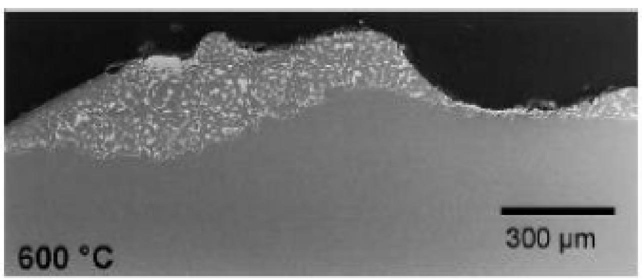
2. Dissolution



- Loss of steel alloying elements (Ni, Mn, Cr)
- LBE penetration
- Ferritization of dissolution zone due to loss of austenite stabilizers (Ni, Mn)

316L: 500°C, 3282 h, 7.5×10^{-13} < [O] (mass%) < 2.8×10^{-8}, static LBE

3. Erosion



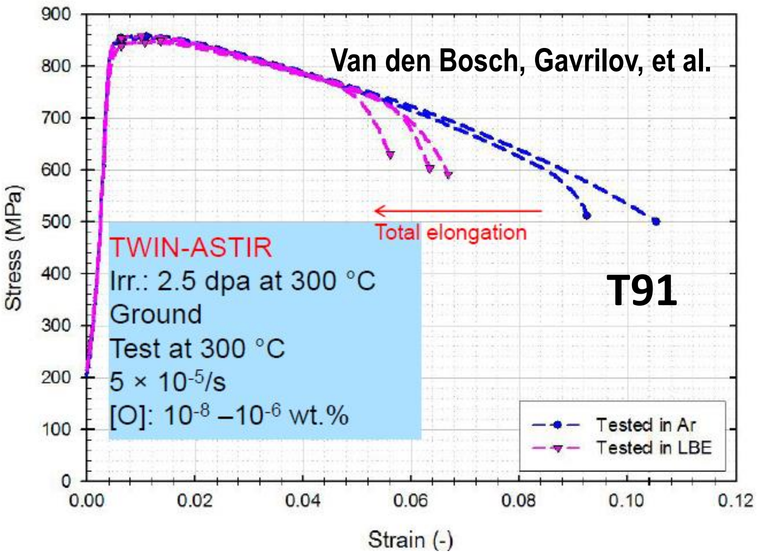
- Severe material loss & compromise of structural integrity
- Observed at high LBE flow velocities, two-phase flow, and sites of flow diversion →fluid velocity = additional variable

316L: 600°C, 2000 h, [O] ≈ 10^{-6} mass%, flowing LBE (v ≈ 2 m/s)
*(Müller et al., Journal Nuclear Materials **301** (2002) 40-46)*

Characterization of corrosion effects requires mapping as function of several variables

Liquid metal embrittlement (LME): what is it?

- **Definition**: A ductile metal experiences drastic loss in tensile elongation or undergoes brittle fracture when exposed to specific liquid metal
- Only a specific liquid metal embrittles a specific metal or its alloys (couple)
- Liquid metal/solid metal couples initially recorded as immune to LME were then found susceptible changing testing procedures: **large scatter in data**
- Influencing factors:
  - Temperature
  - Strain rate
  - Solid metal microstructure
  - Solid and liquid metal composition
  - Oxygen content in the liquid
- Key factor: wetting
- Mechanisms: still unclear & debated



- **ALL F/M steels show susceptibility to LME in heavy liquid metals (Pb_{liq}, PbBi = LBE)**
- The contact with LM can **affect also other mechanical properties**: fracture toughness, fatigue, creep, creep-fatigue

Corrosion/Dissolution/Erosion in Na_{liq}, Pb_{liq} and its alloys, He gas

- Liquid sodium: does corrode and erode, but these processes are considered under control and not limiting safety
  - Oxygen as low as possible
    - *The no. 1 safety problem with Na is the reaction with water*

  NB: LME affects also austenitic steels in contact with Na_{liq}, but stable contact is difficult to establish

- Liquid lead and its alloys: corrosion, dissolution, erosion and above all LME are serious problems for safety
  - Active oxygen control: if oxygen too low oxide layer does not stabilize, if too high PbO starts to form
  - LBE data for 316L/1.4970 (austenitic steels) show that T must be <400°C: inacceptable for LFR → mitigation strategies are necessary
  - LME data for 316L/1.4970 (austenitic steels) suggest immunity; however dissolution of Ni in Pb_{liq} may lead to ferritisation and thus exposes to LME even in the case of austenitic steels

- He: high T corrosion and erosion, mitigation strategies may be necessary

Mitigation strategies against cor/e/rosion

- The protection against corrosion/dissolution/erosion is given by the stability of the **oxide layer** formed on the surface between solid & liquid metal
  - Typically high-Cr steels have improved corrosion resistance

- If the oxide layer is insufficient, possible solution are **coatings**
  - Ceramic coatings, typically Al_2O_3, maybe Max phases, applied with different techniques or obtained via surface changes of composition (*surface engineering*)

- More "metallurgical" promising solution: **alumina forming alloys** (AFA)
  - Austenitic (but also F/M) steels that contain a significant amount of Al, to form a self-healing protective alumina layer
  - Require tuning the whole composition to <u>promote alumina formation</u>

Summary

- F/M steels swell much less (and conduct better) than austenitic steels, but suffer from low T radiation embrittlement and more severe loss of elongation, are less resistant to thermal creep and are susceptible to LME

→ Austenitic steels are the candidate materials for prototypes; improved F/M steels may be candidates for future systems

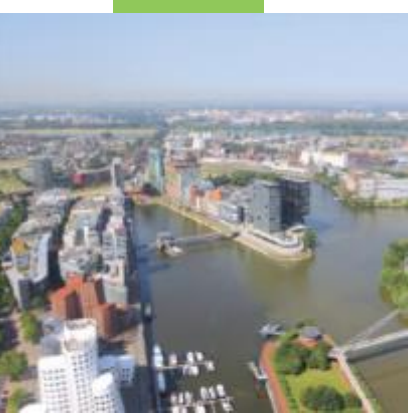
- When the range is 800-1000°C or more, ceramics enter into play

- Corrosion/erosion in Na is handled; it is more serious in Pb_{liq} and its alloys

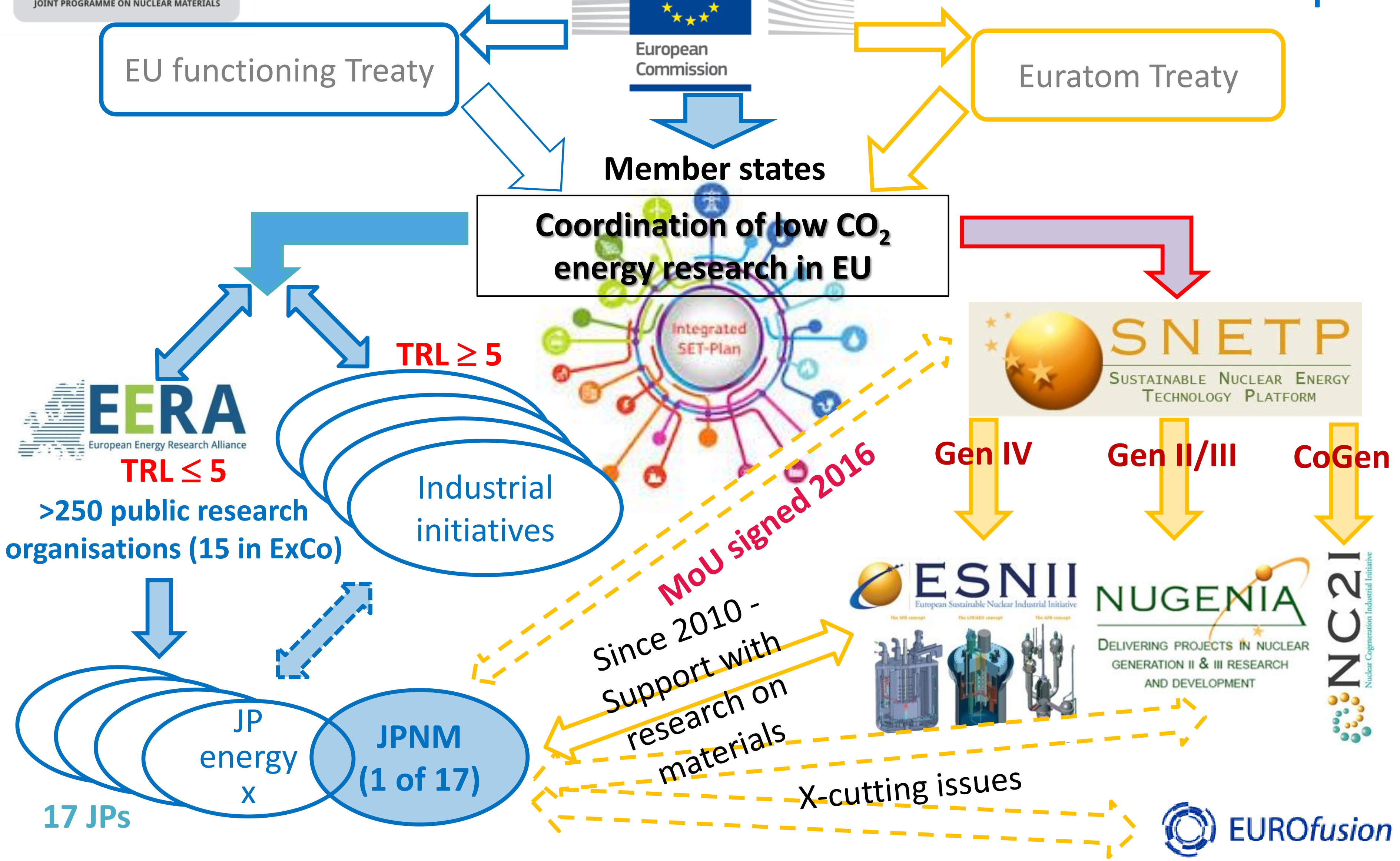
→ Ceramic coatings and/or surface modifications and/or self-healing alumina layers (AFA) by Al addition are needed to improve corrosion resistance for contact with heavy liquid metals

EERA
European Energy Research Alliance
JPNM
JOINT PROGRAMME ON NUCLEAR MATERIALS

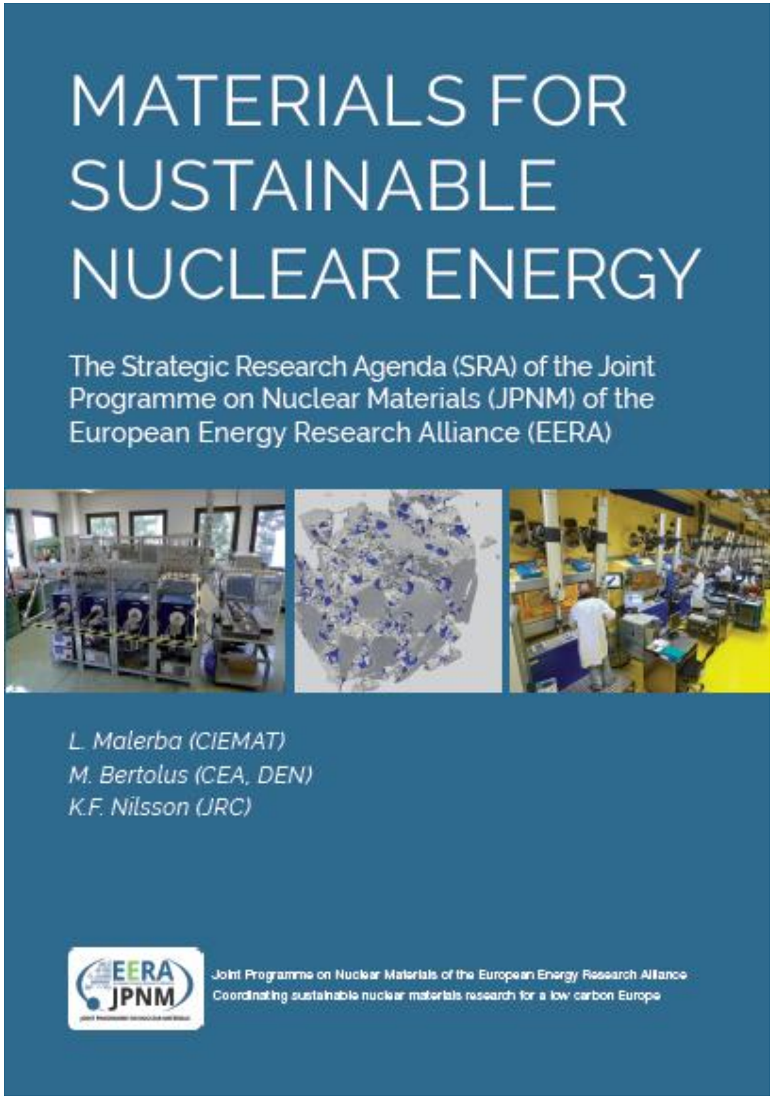
Part III Materials for the ESNII prototypes and beyond



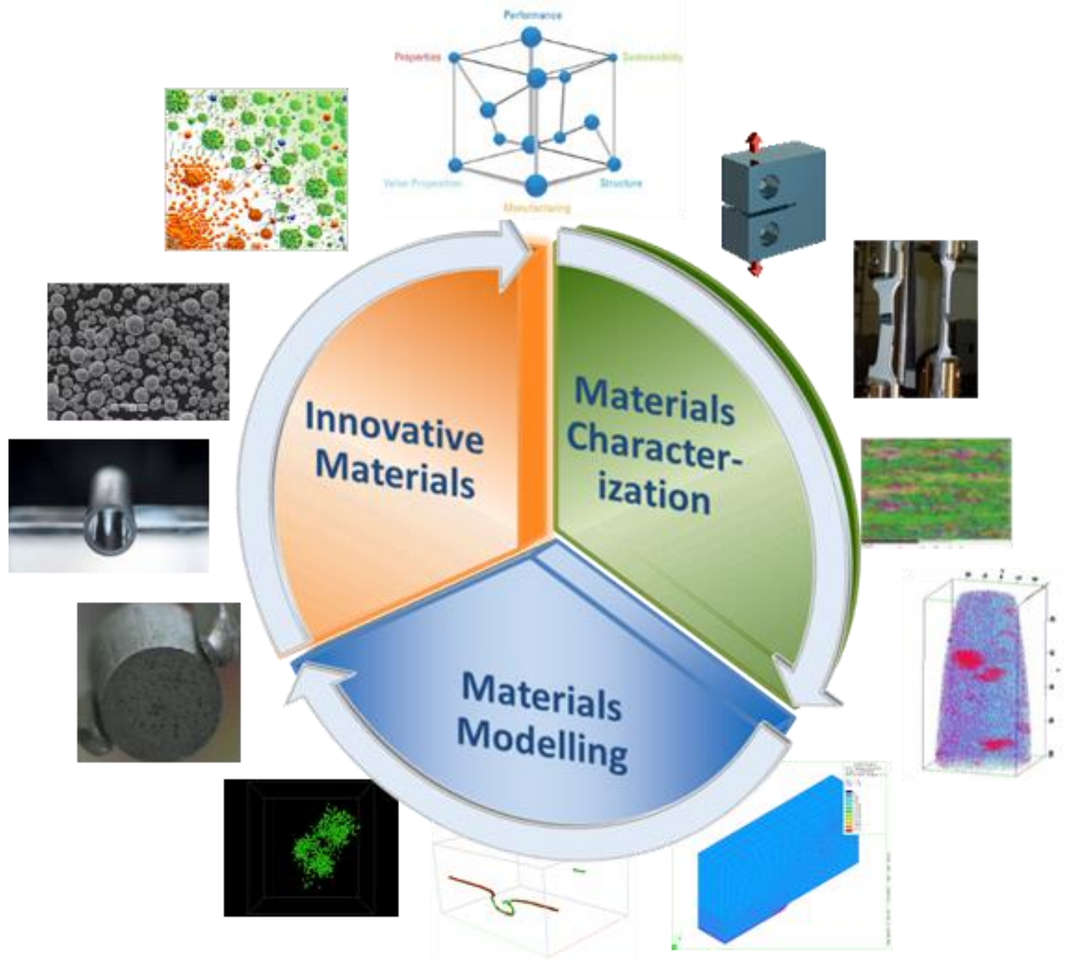

The EERA JPNM: the nuclear materials voice in Europe



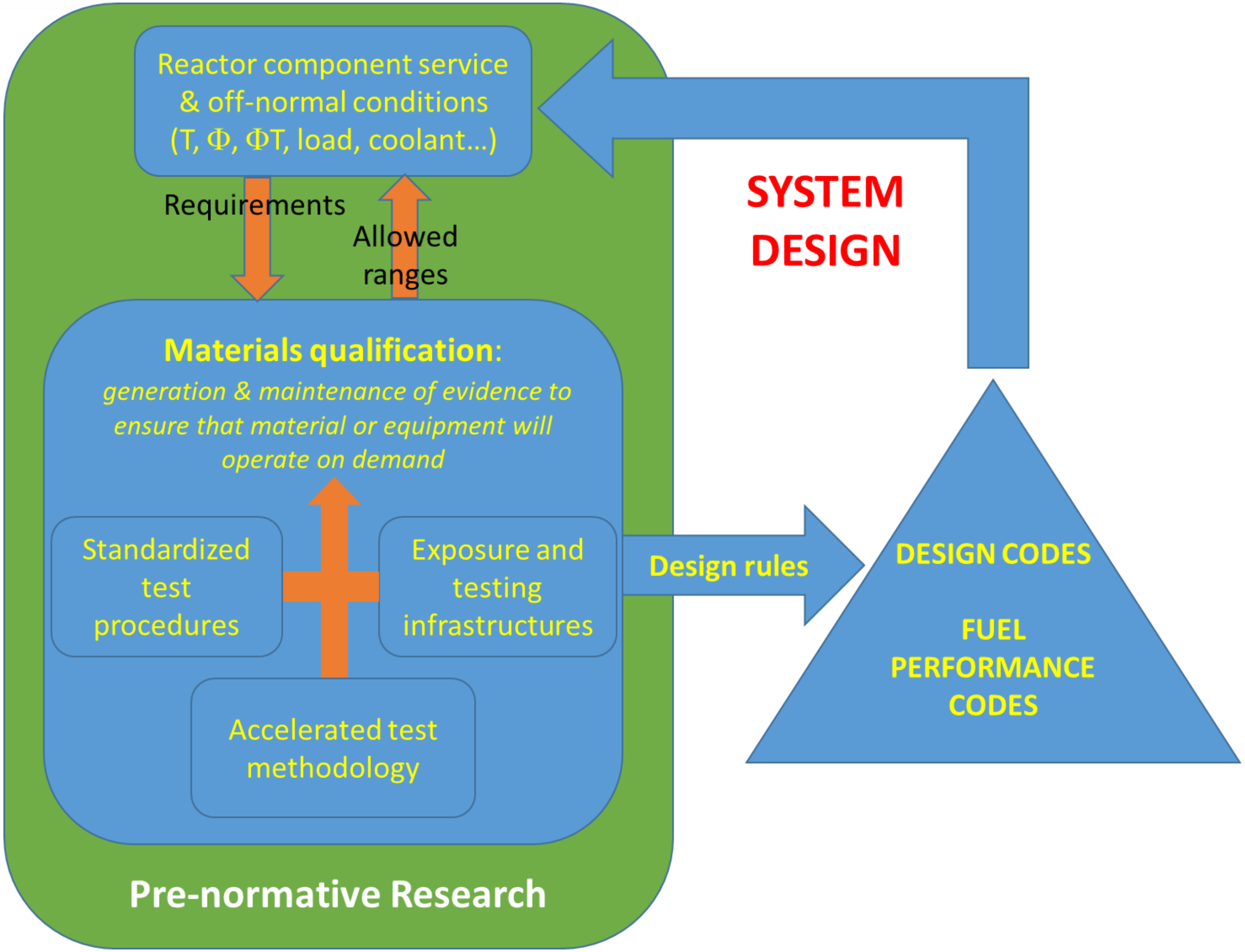
EERA-JPNM Strategic Research Agenda: Materials for sustainable nuclear energy



http://www.eera-jpnm.eu/?q=jpnm&sq=nboard



Approaches: Materials Qualification in Support of Design



Approaches: Development of New Materials Solutions

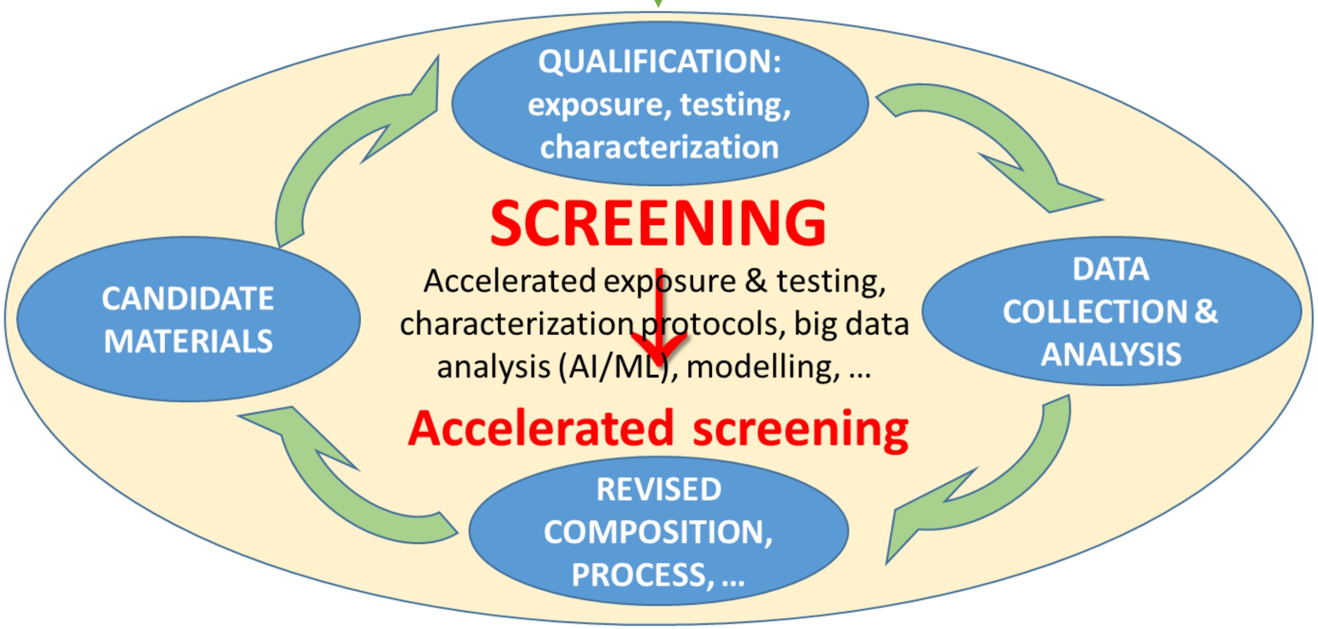
DRIVING FORCE

Insatisfactory design-driven mitigation:
→ Too short component availability & lifetime

Need to improve economics/safety margins

NEW MATERIALS SOLUTIONS

- Different material of same class
  - Material protection
  - Improved properties (composition, architecture, manufacturing, processing, ...)
- Entirely different material
- **Targeted material development**



SCREENING

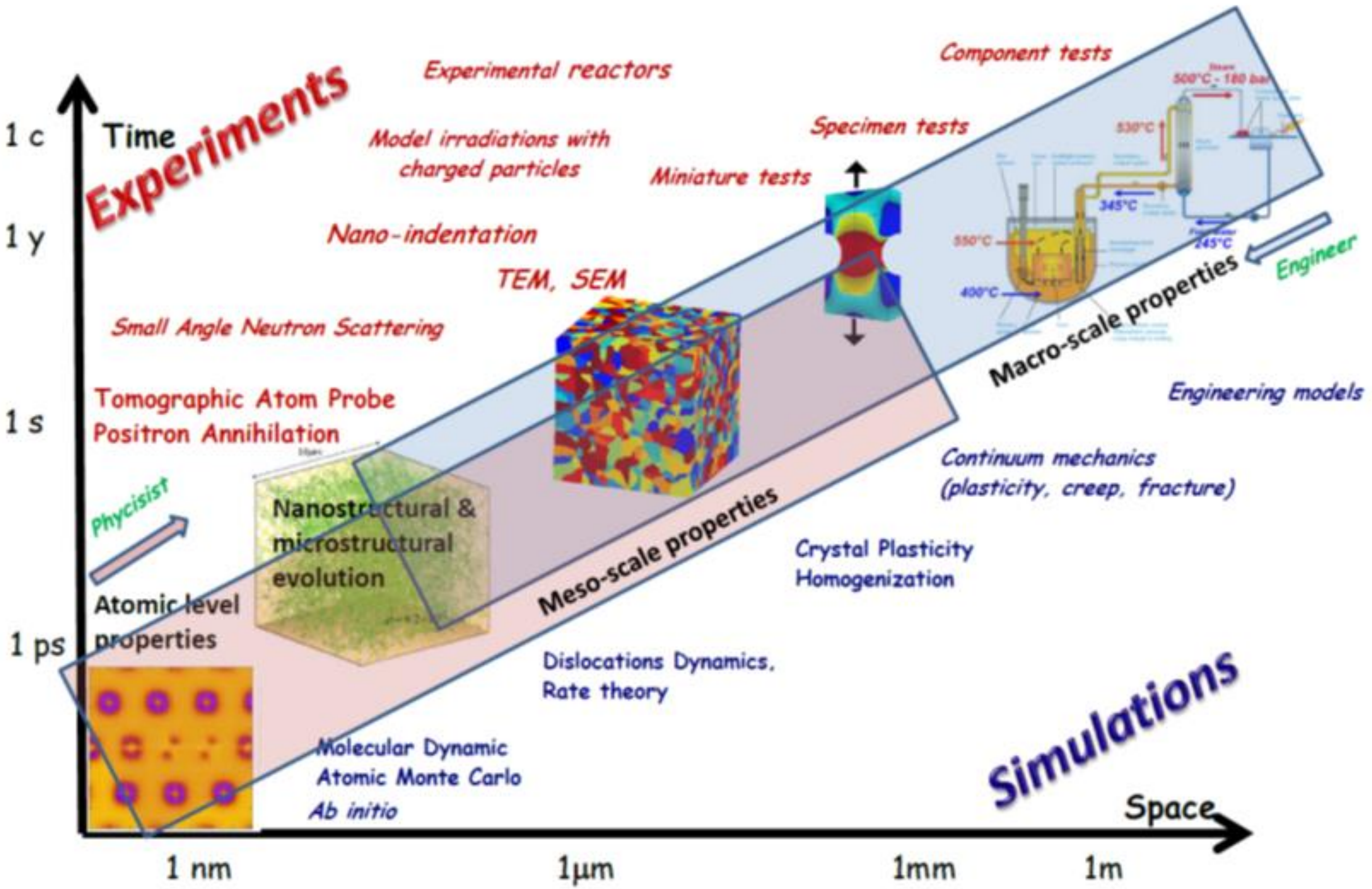
Joining, industrial production upscaling, ...

SELECTION

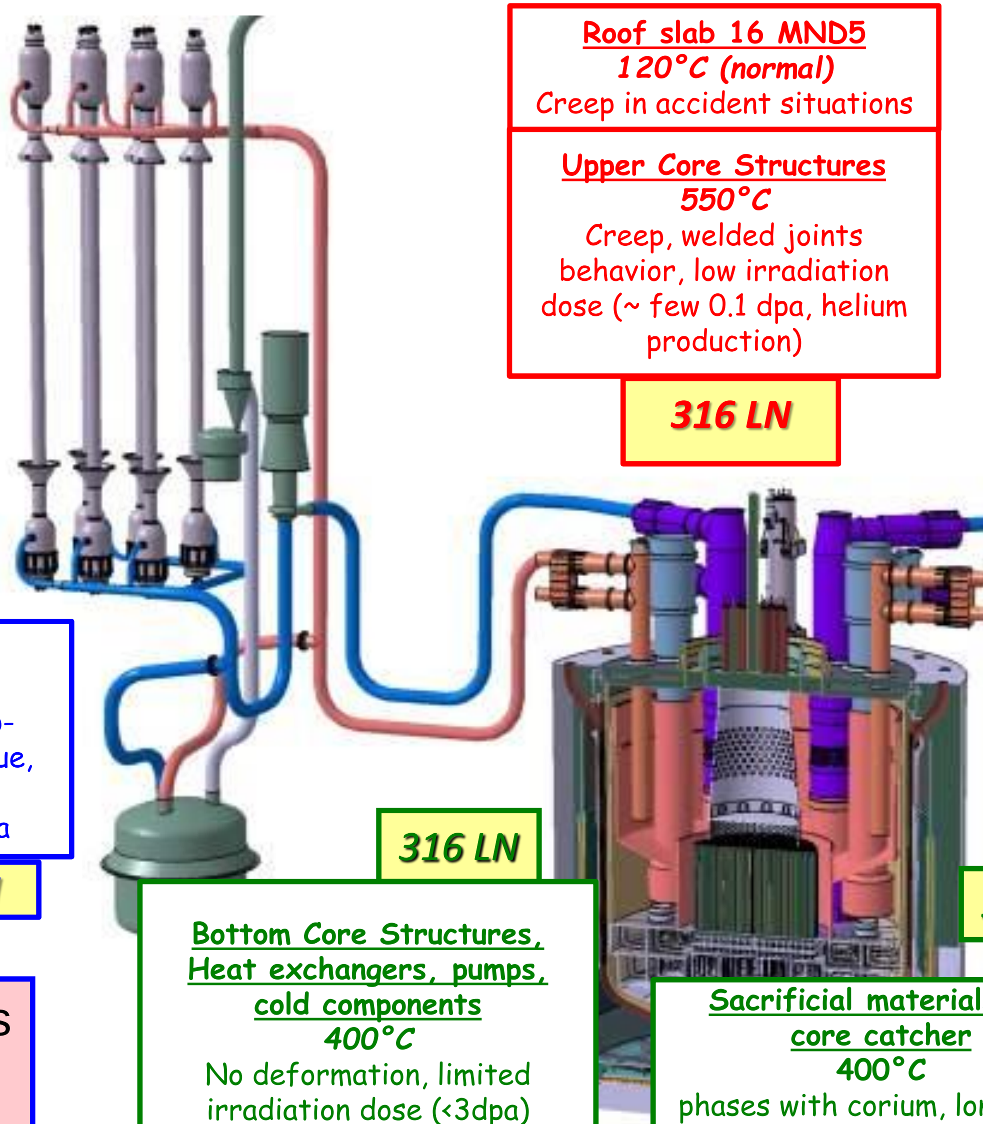
Full qualification, codification, monitoring

APPLICATION

Approaches: Materials Modelling and Microstructural Characterization



ASTRID: 316LN everywhere, except fuel assembly



Na-gas Heat Exchanger or Steam Generator
330 – 530°C
aging, welding, compatibility with environment

316 LN
Alloy 800

Roof slab 16 MND5
120°C (normal)
Creep in accident situations

Upper Core Structures
550°C
Creep, welded joints behavior, low irradiation dose (~ few 0.1 dpa, helium production)

316 LN

Fuel Assemblies
400 – 650°C
Irradiation, mech. strength

- **1er core (110 dpa) AIM1 cladding ,EM10 wrapper tube**
- Advanced Cores
  AIM2 cladding (130 dpa)
  F/M ODS cladding (180 dpa)

Circuits – pipes
350– 550°C
Creep, fatigue, creep-fatigue, thermal fatigue, aging, welding, compatibility with Na

316 LN

Courtesy of M. Leblanc, CEA

316 LN

Bottom Core Structures, Heat exchangers, pumps, cold components
400°C
No deformation, limited irradiation dose (<3dpa)

316 LN

Sacrificial materials for core catcher
400°C
phases with corium, long term behavior

Vessel
400°C
No deformation, negligible creep,
negligible irradiation

Fixed structures
Life time
40 → 60 yrs

ASTRID fuel assembly

Core components:
- **Fuel cladding (critical component)**
- **Wrapper tube**

Courtesy of M. Leblanc, CEA

ASTRID demonstrator
480-700°C, 110 dpa

- Use of reference materials benefiting from a large feed-back from the previous French SFRs (Rapsodie, Phénix, SuperPhénix)
- **Austenitic steels (cladding),**
  F/M steels (wrapper tube),
  B_4C (absorbers).
- Improving the description of their behavior (swelling, high temperature)
- **Qualifying the materials regarding the specificities of ASTRID core**

Future SFRs
480-700°C, 180 dpa

- Use of advanced materials with improved properties
- **ODS ferritic/martensitic steels (cladding),**
  SiC/SiC composites (wrapper tube),
  Innovative absorbers and reflectors.
- R&D to develop/fabricate suitable grades
- **Qualifying these materials in ASTRID**

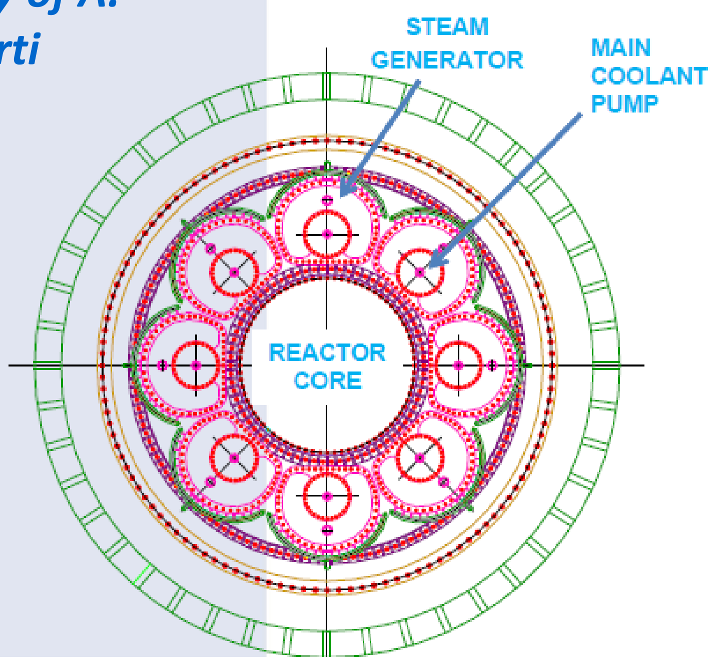
Lead fast reactor prototype

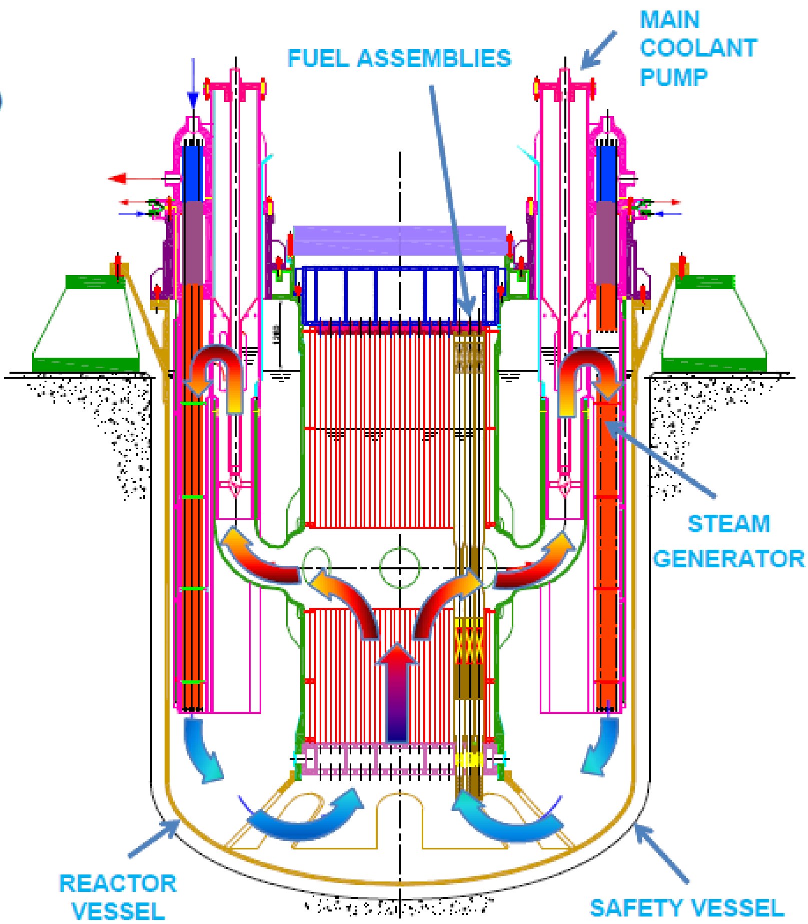
ANSALDO NUCLEARE
Ansaldo Energia Group

ALFRED - Reactor Configuration from LEADER

Power:	*300 MWth (125 Mwe)*
Primary cycle:	*400-480 °C*
Secondary cycle:	*335-450 °C , 180 bar*

Courtesy of A. Alemberti





LFR materials

Component	Min./Max Temp. Normal Operation (long term) °C	Max Temp. Accident Conditions (transient) °C	Max. Lead velocity (m/s)	Max. Radiation damage (dpa/y)	Max. Radiation damage (dpa)	Materials and coatings for low oxygen concentration (10^{-6} / 10^{-7} wt%)		
						Material	Coating	Notes
Reactor Vessel	380÷430	500	0,1	< 10^{-5}	0,0002	AISI316L AISI316LN	No	(*) Ta coating needs lower oxygen content
							Ta* (CVD)	
							Al diffusion coating/Pack Cem.	
Inner Vessel	380÷480	550	0,2	0,1	2,1	AISI316L	No	
							Ta* (CVD)	
							Al diffusion coating/Pack Cem.	
Steam Generator	380÷480	550	0,6	< 10-5	0,0001	T91	No	T91 reference option to maximise heat transfer (less overall surgiface and thermal stresses)
							Ta* (CVD)	
							Al diffusion coating/Pack Cem.	
						AISI316L	No	Stainless steel backup option
							Ta* (CVD)	
							Al diffusion coating/Pack Cem.	
Fuel clad	380÷550	600	2		100	15-15Ti	FeCrAlY buffer layer + Al_2O_3 topcoat (PLD)	
FA Structures	380÷530	550	2		100	15-15Ti	FeCrAlY buffer layer + Al_2O_3 topcoat (PLD)	
						AISI316L	FeCrAlY buffer layer + Al_2O_3 topcoat (PLD)	
DHR Heat Exchanger	380÷430	500	0,2	< 10-5	0,0001	AISI316L	No	SS reference option
							Ta* (CVD)	
							Al diffusion coating/Pack Cem.	
							FeCrAlY buffer layer + Al_2O_3 topcoat (PLD)	
						T91	No	T91 backup option
							Ta* (CVD)	
							Al diffusion coating/Pack Cem.	
							FeCrAlY buffer layer + Al_2O_3 topcoat (PLD)	
Primary Pumps	380÷480	550	15÷20	< 10-5	0,0001	AISI316L	No	Pump Placed in cold leg
							Al diffusion coating/Pack Cem.	FeCrAlY buffer layer + Al_2O_3 topcoat (PLD)
							Ta* (CVD)	
							MAXTHAL (Ti_3SiC_2)	

Again 316L(N) and 15-15Ti dominate (*T91 only for steam generator*)

Coatings (Ta, Al_2O_3, FeCrAlY, …) are explicitly mentioned

Evolution of prototype through three stages, with different materials

Courtesy of A. Alemberti

Three stages of ALFRED

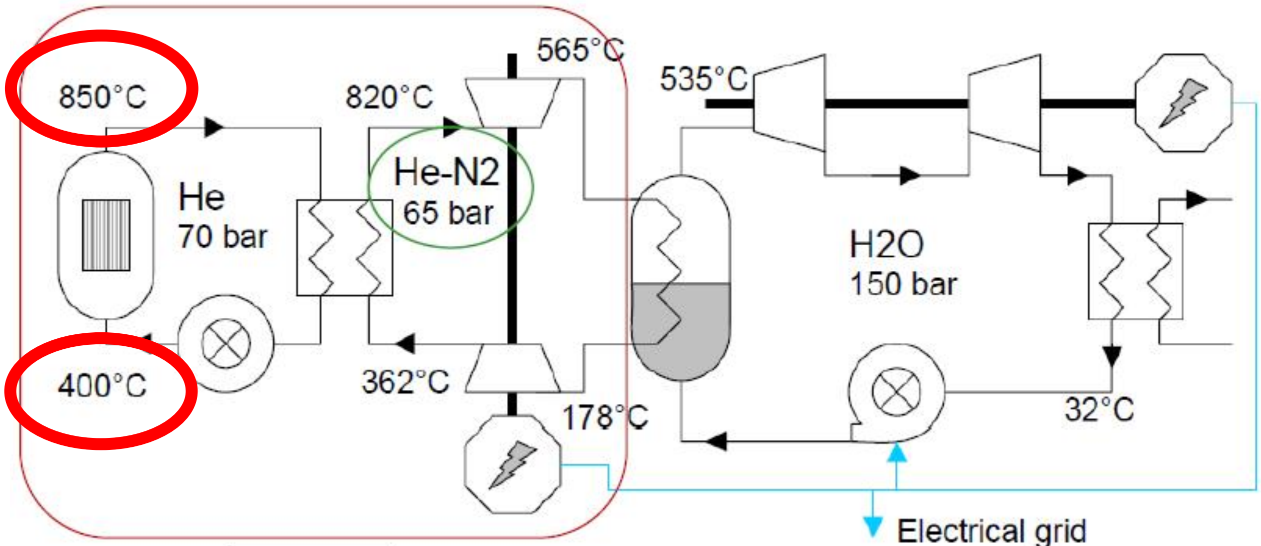
- 1st stage: low temperature
  - Proven technology, proven materials, oxygen control, low temperature
  - Aimed at in-core qualification of PLD Al_2O_3 coating for cladding
- 2nd stage: medium temperature – coating on cladding by default
  - Need for fuel assembly replacement, same steam generator and primary pumps
  - Aimed at in-core qualification at higher temperature
- 3rd stage: high temperature – advanced heat exchanger, AFA of improved F/M steels
  - Replacement of main components for improved performances
  - Representative of FOAK conditions for LFR deployment

	1st stage	Final stage
Power	100 MWth	300 MWth
Thermal cycle	390-430°C	400-520°C
Coolant chemistry control	$10^{-6} \div 10^{-8}$ O2 wt.%	Same, applicable to low T regions
Materials	316L, 15-15Ti	Relying on coating or innovative materials

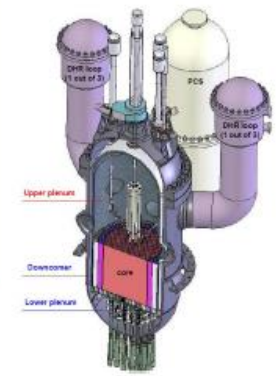
ALLEGRO: the gas fast reactor prototype materials are still largely to be defined

- **Indirect cycle** (FP7 GoFastR)
  - Target core outlet T: 850 → 780 [°C]
  - Main HX sec. outlet T: 820 → 750 [°C]
  - Intermediate gas circuit: ~80N2-20He

Courtesy of L. Belovský, UJV



ALLEGRO



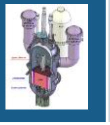
Vessel: 316L or 9Cr-1Mo F/M

Cladding: 15-15Ti (low T core) → F/M ODS (?) → V or Mo alloys, SiC/SiC

Other in core components: **?**

Above core thermal barrier: Al_2O_3 or SiC/SiC (?)

Materials by system

PHASES → / SYSTEMS ↓		ESNII demonstrator		FOAK (prototype)	Commercial deployment
		As licensed (phase I)	Evolving (phase II)		
SFR (ASTRID) 	Periodically Replaced Comp.	AuSS: 15-15Ti – AIM1 (cladding) F/M: EM10 (wrapper)	AIM2 or F/M ODS (cladding)	TMT F/M or F/M ODS	TMT F/M, F/M ODS, perhaps SiC/SiC
	Permanent Structural Comp.	AuSS: AISI316L(N); 800SPH		AuSS: AISI316L(N); TMT F/M	
ADS (MYRRHA) 	Periodically Replaced Comp.	Cladding: 1.4790; structures: 316L(N)	Coated 15-15Ti (FeAl, FeCrSi, FeTa, MAX phases, …) or AFA	N/A	
	Permanent Structural Comp.	316L(N)			
LFR (ALFRED) 	Periodically Replaced Comp.	Cladding and structures: (Al_2O_3 coated) 15-15Ti (AIM1)	Cladding and structures: Al_2O_3 Coated 15-15Ti or AFA	Cladding: AFA or FeCrAl ODS Structures: AFA	AFA or FeCrAl ODS, or (coated) Mo-ODS, or SiC_f/SIC ,
	Permanent Structural Comp.	316L(N)		AFA or ferritic steel lined with AFA	
GFR (ALLEGRO) / (V)HTR 	Periodically Replaced Comp.	GFR: T<550°C: 15-15Ti (cladding) – AFA? HTR: TRISO (SiC)	GFR: T> 850°C: SiC/SiC (cladding) / HTR TRISO (SiC)	SiC/SiC, perhaps Mo-ODS/ HTR TRISO (SiC)	
	Permanent Structural Comp.	GFR : T<550°C: 316L(N) – AFA ? HTR: graphite	GFR: 550<T<850°C: AFA, FeCrAl ? HTR: graphite	GFR: AFA or FeCrAl, perhaps , Mo or V alloys HTR: graphite	

EERA
European Energy Research Alliance
JPNM
JOINT PROGRAMME ON NUCLEAR MATERIALS

From fundamental science to application: Short and long term perspectives

SAFETY

SHORT TERM APPLICATION

GenIV demonstrators and simulators: non-optimal efficiency, flexibility, progressive performance improvement

Qualification existing materials: codes

Focus: Austenitic steels (including welds) / MOX
(less effort on: F/M steels, Ni-base alloys, MA-fuel)

Issues Structural Materials:
SFR: 60 years design lifetime
LFR & ADS: compatibility with heavy liquid metal
GFR: compatibility with high temperature/pressure He
All: test standardization, health monitoring

Issues Fuel Materials:
All: Margin to melt, fission products & gas, mechanical properties, interaction with cladding/coolant

LONG TERM APPLICATION

Truly GenIV commercial systems: enhanced economy, better use of resources, waste reduction, …

Innovative material solutions

Fabrication/Processing & Screening/Qualification:

Structural Materials:
Improved austenitic steels (low swelling/SFR, AFA/LFR)
Creep-strength enhanced (650°C) & ODS (>700°C) F/M steels (all) / FeCrAl (LFR)
Ceramic coating & surface layer techniques
SiC/SiC cladding (GFR, >850°C)
Prospective ceramic & metallic refractory (high temperature) materials (>850°C); HEA

Fuel Materials:
U/Pu carbides, nitrides, MA-bearing fuel, MSR fuel

Physical Modelling

Advanced multiscale materials modelling & micro-structural/micro-mechanical characterization
Use of ion irradiation for materials screening
(Artificial intelligence/high performance computing coupled to integrated materials test-beds)

Thank you for listening
Any question?