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IRRADIATION EFFECTS

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Irradiation Effects

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Technical challenges for Gen IV

- **Identify acceptable dose-temperature windows for materials**

- Can reactor lifetimes approach one century ?

- Evaluate maximum allowable burn-up limits for LWR fuels

- **Establish technical feasibility for Gen-IV fuels and materials**

- Higher temperature operation for high thermodynamic efficiency

(New specifically tailored in- and ex-core structural materials may be needed)

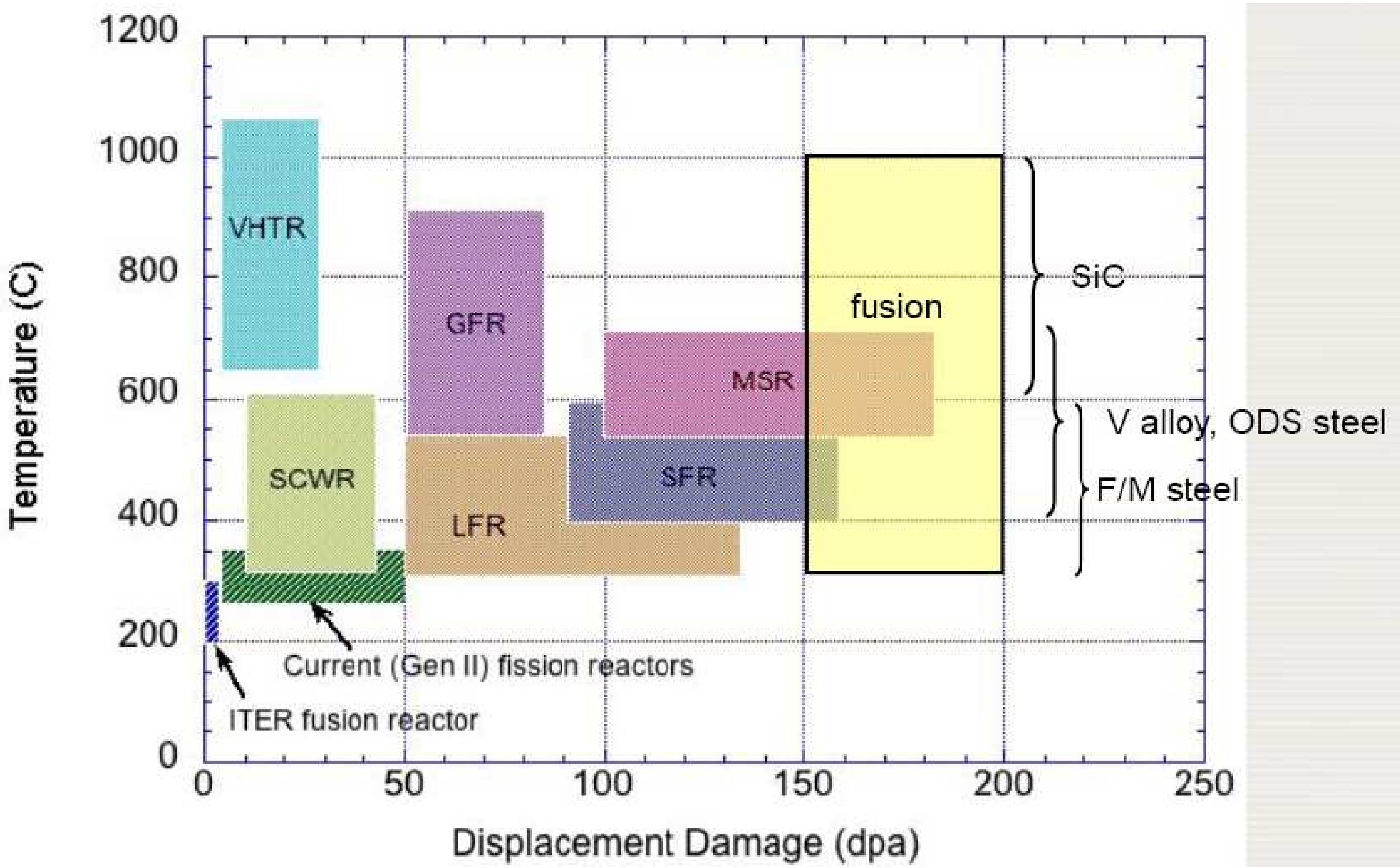
- Establish engineering database for high temperature gas cooled reactor materials (fuels, structures)

- Effect of actinides and fission products on fuel fabricality and irradiation performance (Exploration of fuel recycle options)

- **Science-based options for fuel disposition (once-thru and recycle approaches)**

Introduction

Overview of projected operating temperatures and damage levels for structural materials in 4th generation fission reactors



Irradiation damage by neutrons

- **Inelastic interactions – neutron reacts with the nucleus producing nuclear interactions (capture, (n, α), fission).**

Positively searched for operation of the reactor – e.g. neutron absorption for reactivity control

Concern: - (n, α) reactions (fast neutrons) - continuously dope the alloys with He

- AIC PWR control rods – swelling induces hoop stresses in SS cladding

- **Elastic interactions - neutrons hits the nucleus and transfers only part of its momentum and kinetic energy**

The main mechanism of irradiation damage, depending on its relative value:

Low value of energy transferred – increase of vibration amplitude, impact is local source of heat

Energy transferred larger than E_d (20 - 40eV) - target atom can escape from its lattice site

E_d – varies with species and crystallographic orientation (ASTM E-521-96 2003)

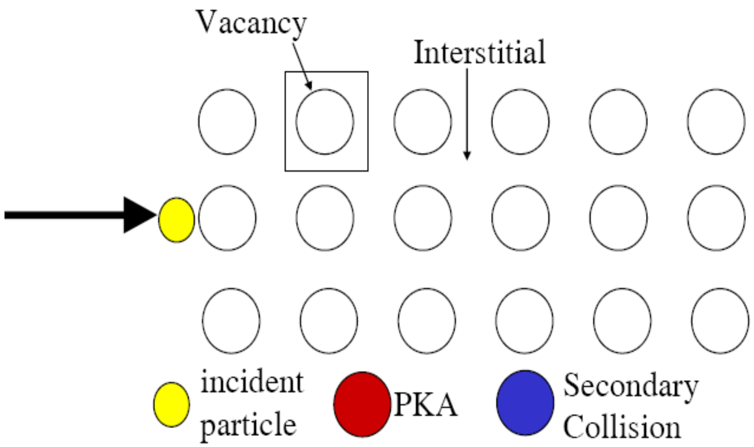
Irradiation damage by neutrons

Following the impact and energy transfer to the target atom:

❑ **Low transferred energy (a few E_d)**

– the final damage a VACANCY and an INTERSTITIAL

- A FRENKEL PAIR (e.g. high-energy electron irradiation)

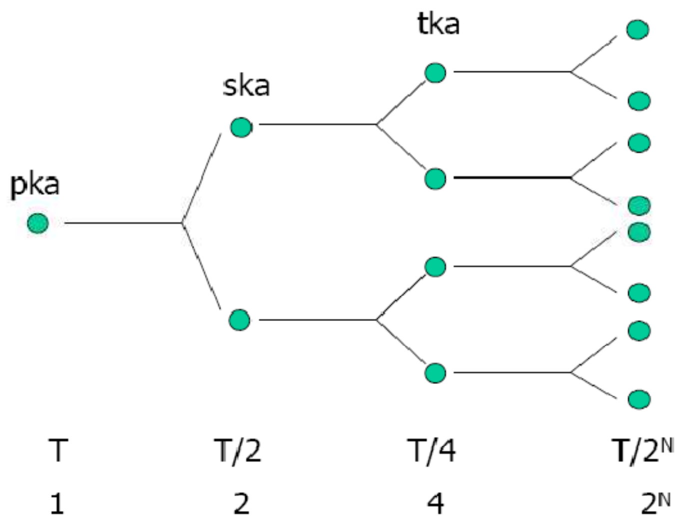


❑ **High values of the energy transferred**

– primary knocked-on atom (pka) interacts with the other atoms of the alloy along its track

-Each interaction transfers ½ its current energy on the secondary target – large number of atoms displaced

- Result: DISPLACEMENT CASCADE



The irradiation by neutrons results in continuous creation of point defects and heat in the bulk of the alloys

Irradiation damage by neutrons – Displ. Cascades

Molecular dynamics computations - advanced description of the behaviour of a cascade

Important features of the cascades:

❑ **The life of a cascade is short – 5-10 ps,**

- **most of PDs recombine**

- **very low number of isolated PD survive to the cascade.**

❑ **The efficiency of the cascade decreases with energy of pka**

❑ **At the end of high energy cascades Clusters of PD of the same types can be formed – small interstitials or vacancy loops, Stacking fault tetrahedra in fcc and etc.**

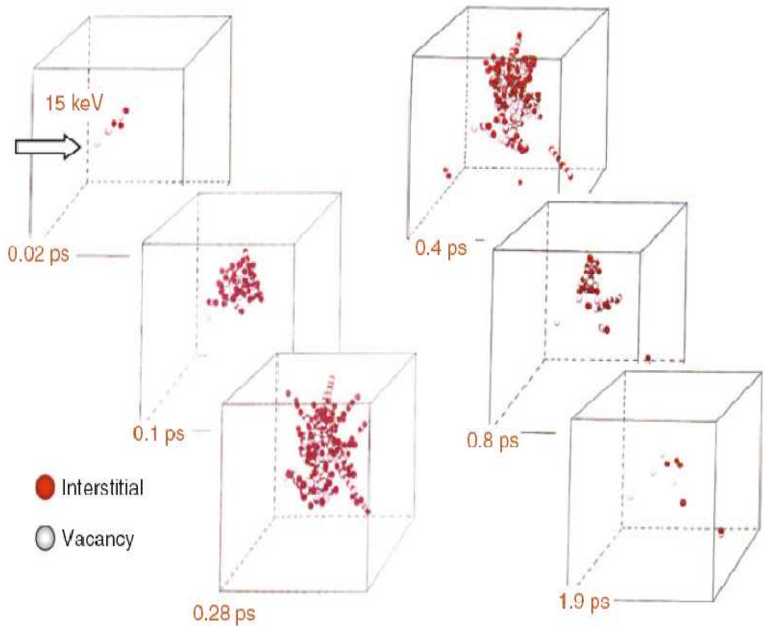


Figure 3
Time evolution of a displacement cascade in Fe (15 keV) (Molecular dynamics simulation, after Souidi et al. 2001)

The number of remaining point defects and clusters of interstitials or vacancies are the initial conditions for thermal evolution of the alloys under irradiation.

Irradiation damage by neutrons – computation of damage

It is common to characterize the full spectrum of neutrons for each irradiation only by its fast neutron contribution

- **In fast reactors – *E > 0.1 MeV* for 1dpa (Fe) – 2 x 10^{25} n.m^{-2}**
- **LWR – *E > 1 MeV* for LWR: 1dpa (Zr) – 5 x 10^{24} n.m^{-2}**

Computation of the damage requires an accurate knowledge of the neutron flux history and spectra at the exact location (pressure vessels of LWR - ASTM Standard E-693-01 2007)

Irradiation to the same fluences, expressed as *E > 1 MeV* and *E > 0.1 MeV* – different damages, the latter being smaller, by a factor of about two.

- **Fuel cladding materials**

-Quantity related to the fuel irradiation: The fuel BURN-UP (BU) (MW.d.t^{-1}):

LWR: 1dpa (Zr) – for BU ~ 4-5 GW.d.t^{-1}
SFR: 1dpa (SS) – for BU ~ 1 GW.d.t^{-1}

Neutron fluences (n · m^{-2}) for 1 dpa in various reactors

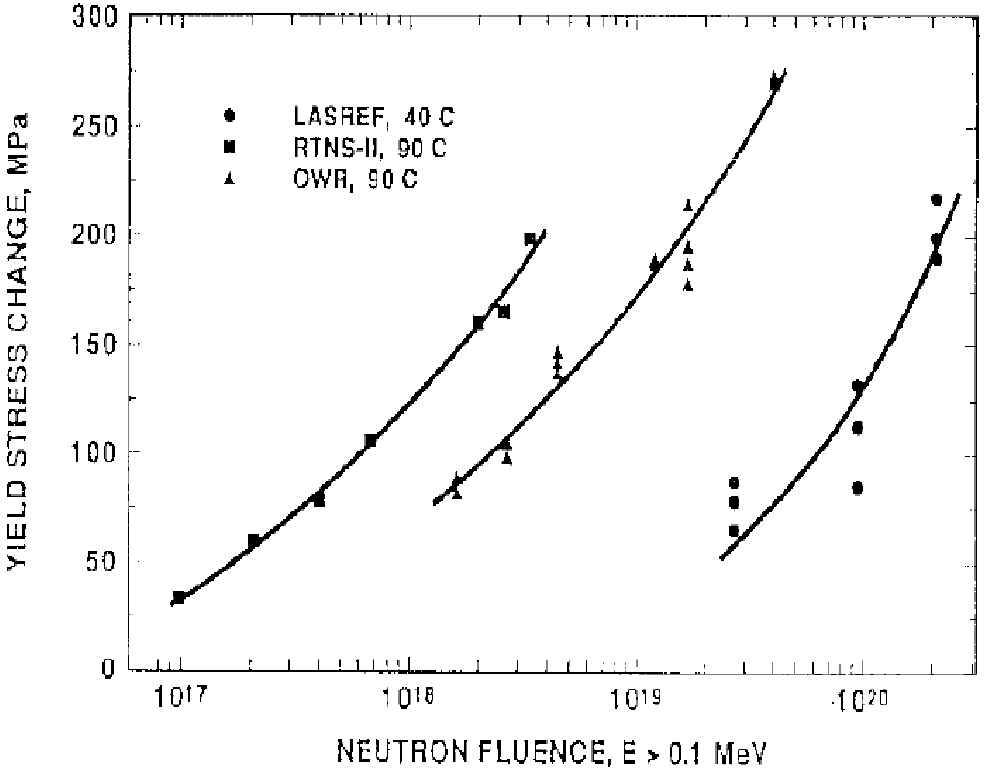
Type of reactor	Atom	$E > 0.1$	$E > 1$ MeV
LWR pressure vessel	Fe		7×10^{24}
PWR BWR (steam 40%)	Zr	1.3×10^{25}	6.4×10^{24}
Na fast reactor	Fe Cr	$\approx 2.2 \times 10^{25}$	
HT GCR	Si C	0.8×10^{25}	

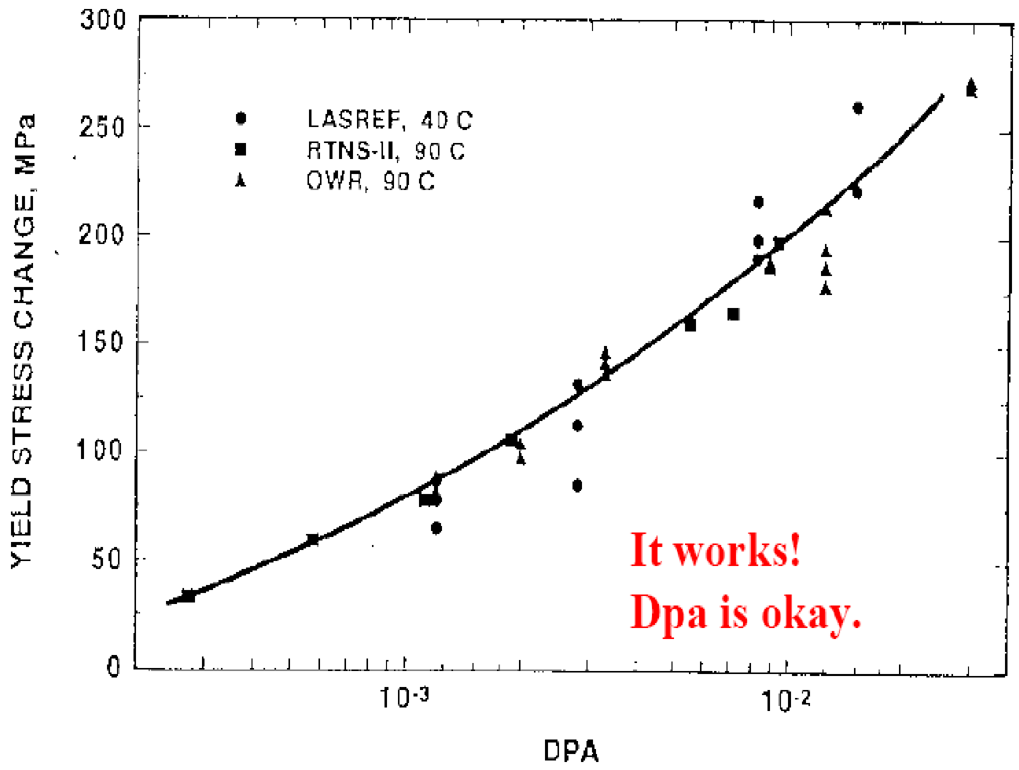
Irradiation damage by neutrons – computation of damage

Radiation induced strengthening of 316 SS:

A) Radically different neutron spectra

B) Fluences converted to Displacement per atom - dpa



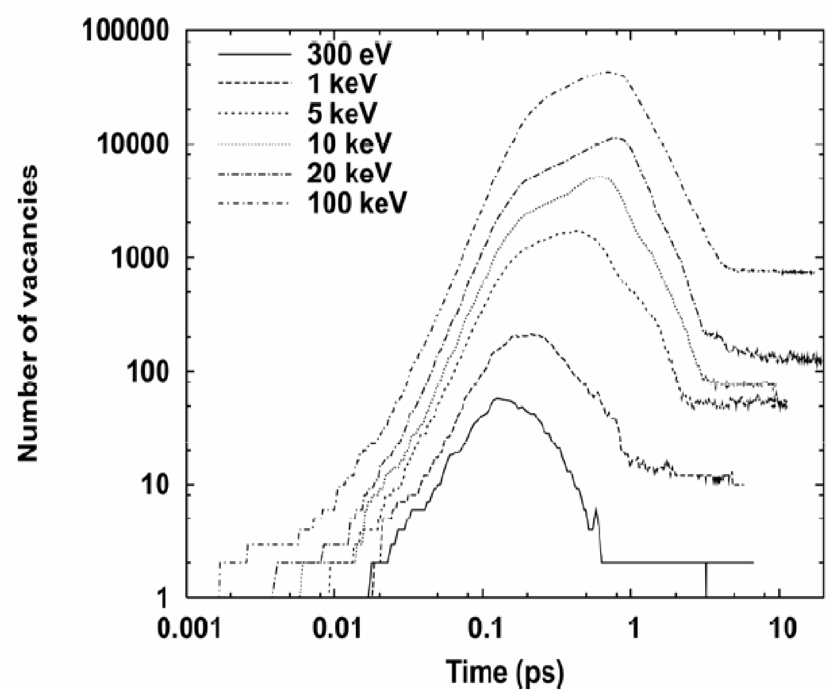


Irradiation damage - dpa

1 displacement per atom (dpa) corresponds to metastable displacement of all atoms in the material

Initial number of atoms knocked off their lattice site is ~ 100 times the dpa value

Thermally activated diffusion of the defects produced by irradiation - large amount of recombination (typically 99 to 99.9 % recovery of the initial displacement damage).



❑ **Requirements for structural materials in advanced nuclear energy systems (~ 100 dpa exposure):**
~99,95 % of "stable" displacement damage must recombine

❑ **Two general strategies for radiation resistance can be envisioned:**
- **Noncrystaline materials**
- **Materials with a high density of nanoscale recombination centers**

Irradiation damage – PD clustering

Institute for Energy

- ❑ The continuous irradiation → a steady state creation of PDs resulting from the final evolution of the cascades
- ❑ The remaining defects → either isolated defects or small clusters of interstitials or vacancies

Interstitials – highly mobile, migrate well bellow room temp.

Vacancies – diffuse more slowly, mobility activated at higher temperatures (250-600 K)

- ❑ Isolated point defects can migrate anywhere in the alloy and interact with any other crystal defect.

Clustering of Interstitial → planar dislocation loops

Clusters of vacancies → 2D and 3D defects e.g. dislocation loops and cavities (SWELLING)

The trapping of PDs on dislocations – climb of these dislocations inducing IRREVERSIBLE STRAIN:

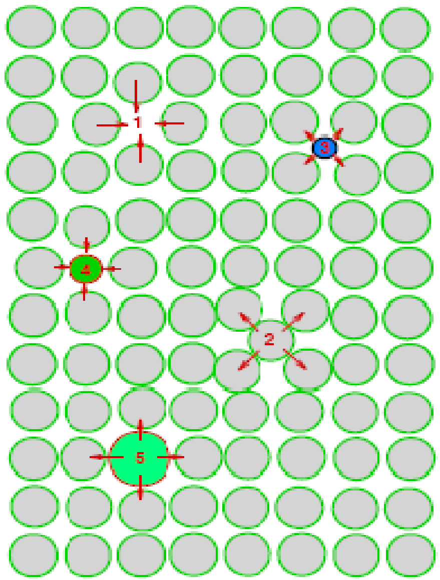
- isotropic results in a macroscopic strain

- anisotropic (applied stress) IRRADIATION CREEP

Important feature of the microstructural change

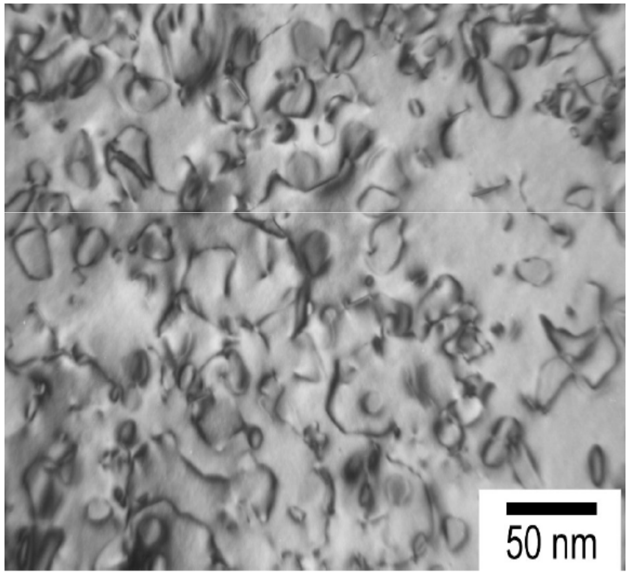
Neutron irradiation can produce in metals and alloys:

- ❑ **Point defects – self-interstitials and vacancies**
- ❑ **Defect clusters – dislocation loops and stacking fault tetrahedra (SFT's)**
- ❑ **Cavities – voids and gas-filled bubbles**

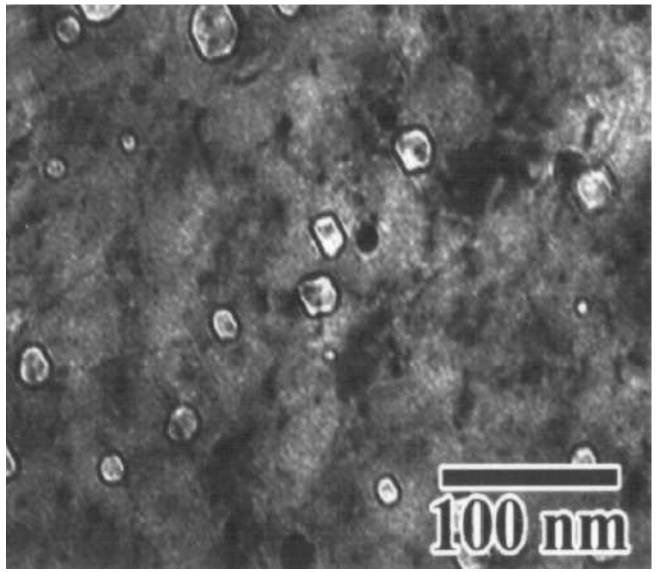


Schematic representation of different point defects in a crystal. (1) vacancy; (2) self-interstitial; (3) interstitial impurity; (4), (5) substitutional impurities. The arrows show the local stresses introduced by the point defects.

www.people.virginia.edu/~lz2n/ mse201/mse201-defects.pdf



Dislocation loops 316L SS



Voids 316L SS

Microstructural changes – temperature dependence

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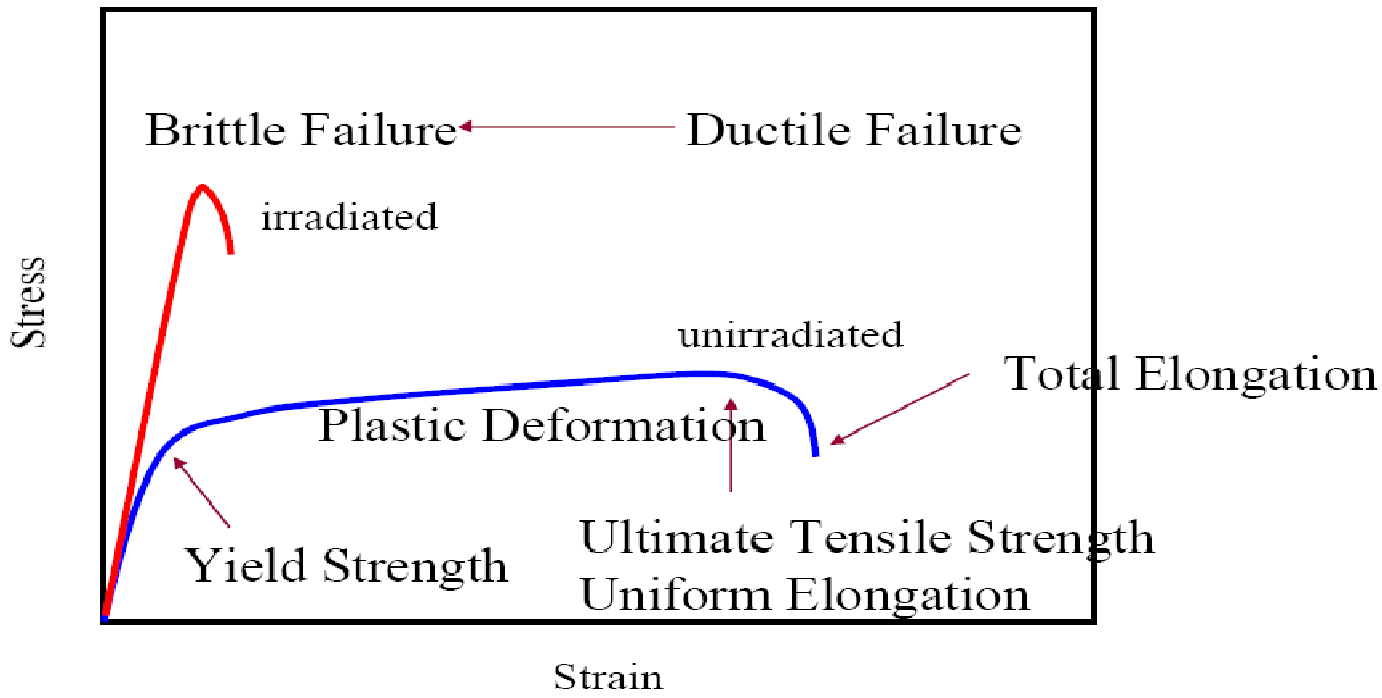
The temperature-dependent microstructural changes induced by neutron irradiation give rise to five major categories of radiation damage:

- Radiation hardening and embrittlement (<0.4 T_M, >0.1 dpa)
- Radiation induced segregation and precipitation (0.3 - 0.6 T_M, >10 dpa)
- Void swelling (0.3 - 0.6 T_M, >10 dpa)
- Radiation induced creep (0.2 - 0.5 T_M, >10 dpa)
- High temperature He embrittlement (>0.5 T_M, >10 dpa)

Two major practical consequences of high level of radiation hardening:

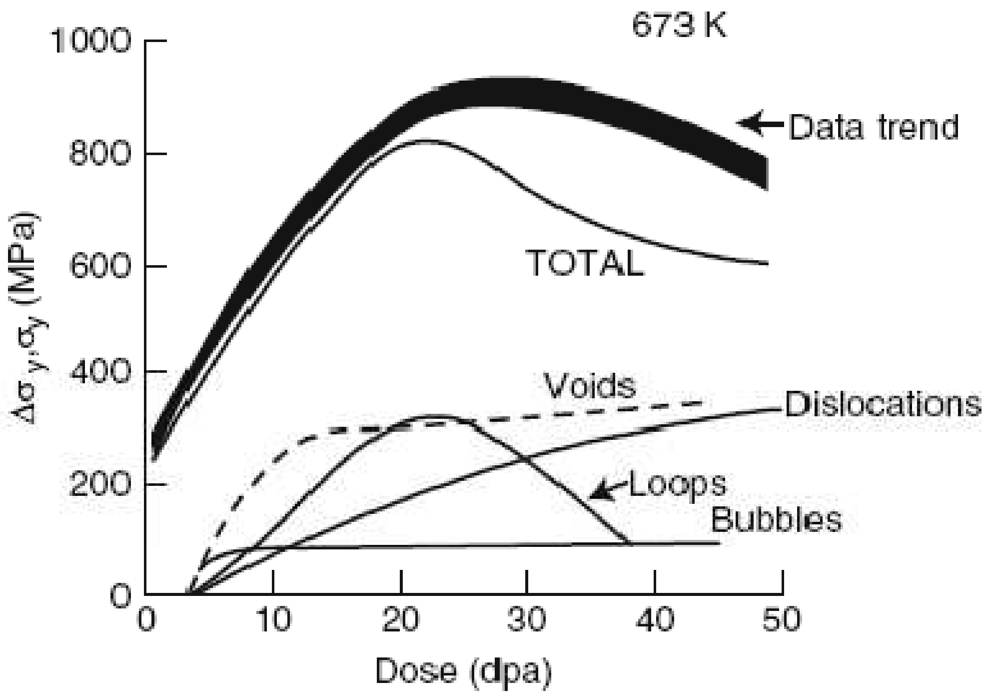
- **A reduction in uniform elongation under tensile test conditions**
- **Reduction in fracture toughness and a potential shift in ductile-brittle transition temperature above the operating temperature (BCC alloys)**

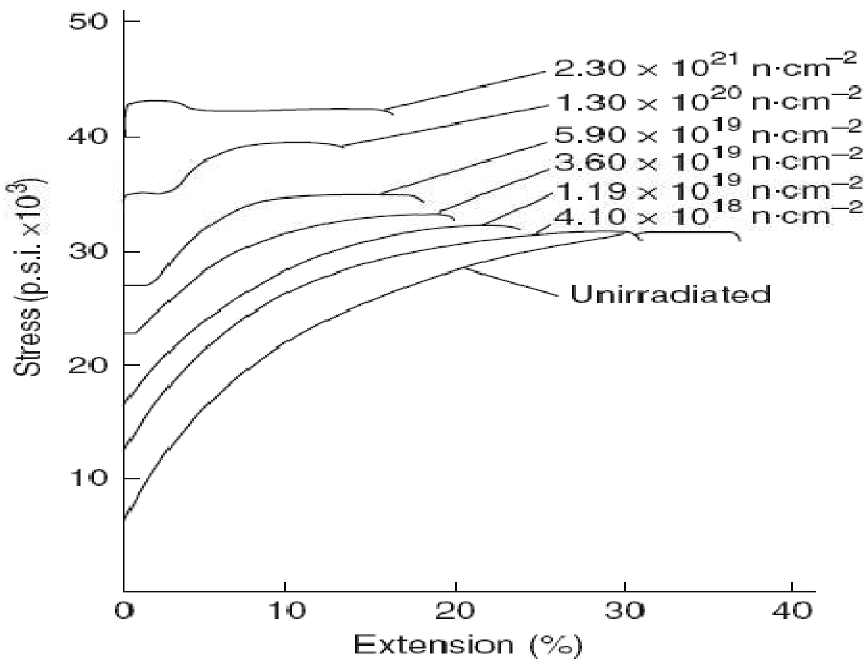


Loss of ductility is the concern

Radiation hardening

- **The basic mechanisms of radiation hardening are related to the formation of various types of PD clusters that act as pinning centers for the dislocations.**
- **Large changes in plastic behaviour are often observed:**
  - **Strain hardening is drastically reduced**
  - **The plastic deformation is more localized**
  - **The uniform and total ductility are reduced**





Radiation hardening - Explanation

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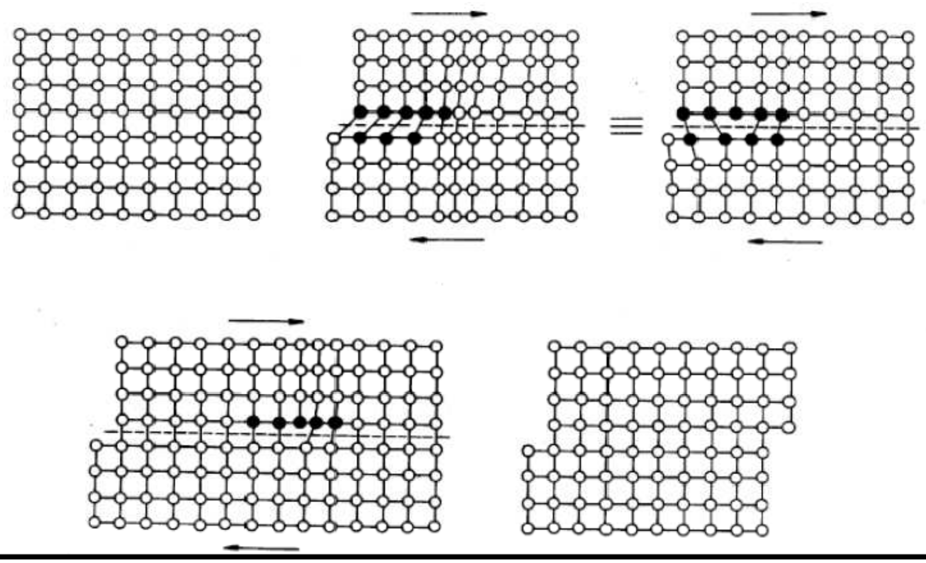
The different behaviour of the dislocations during plastic strain for unirradiated and irradiated materials

- **Unirradiated – the interaction of dislocations with obstacles leads to the multiplication of dislocations – strain hardening, large uniform elongation, necking**

- **Irradiated – the interaction with the irradiation-induced obstacles leads to annihilation of these defects - easy localized plastic deformation**

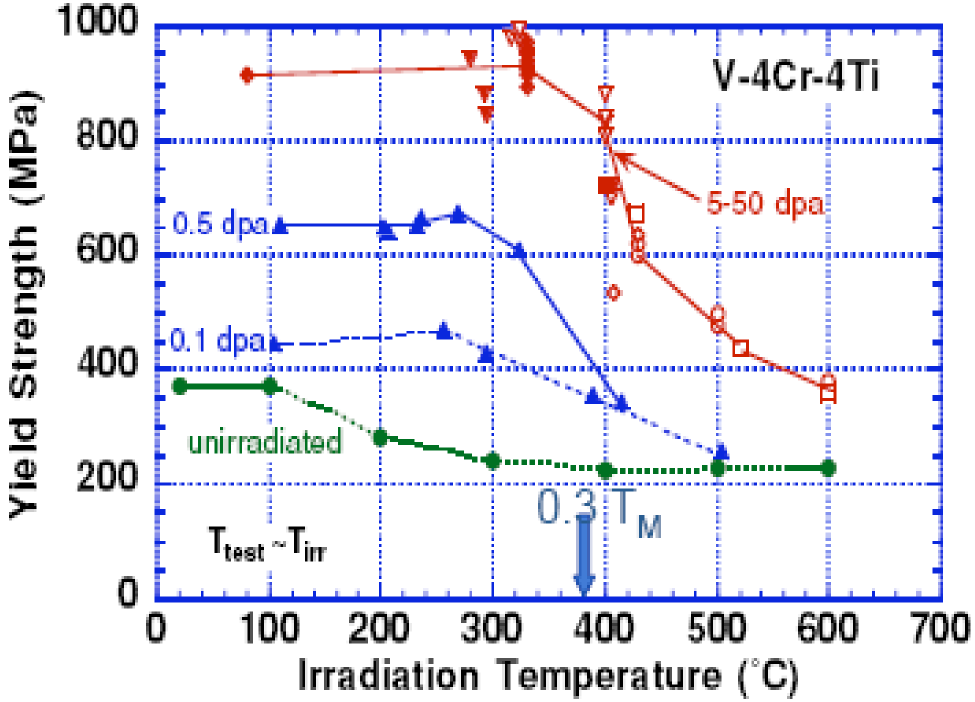


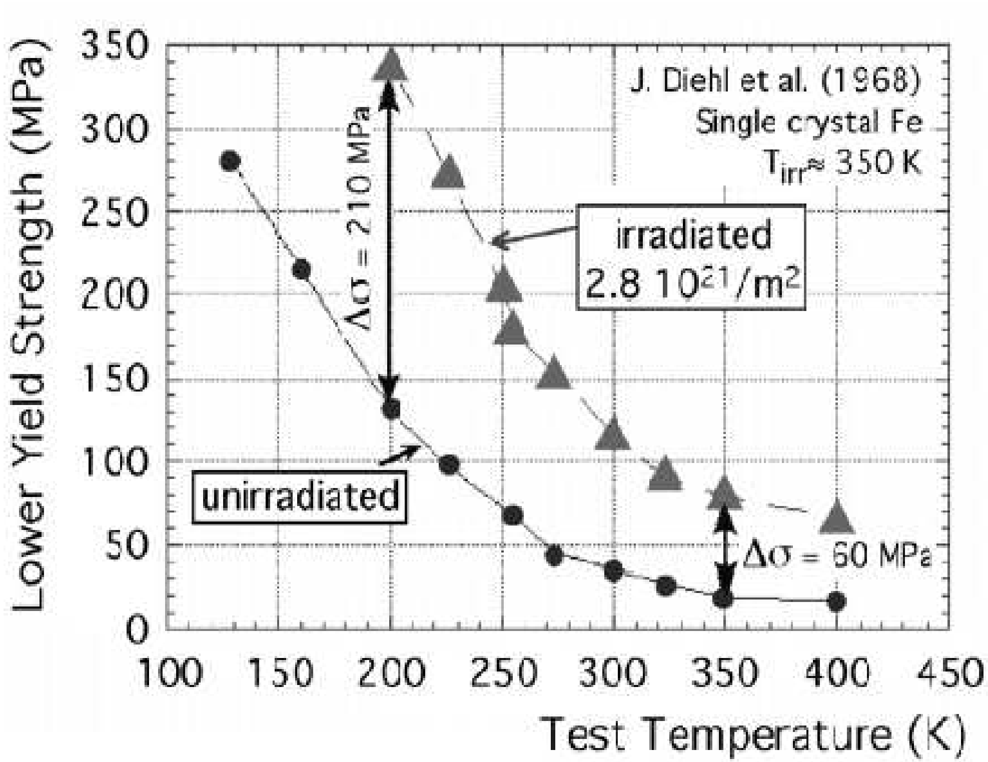
Hartmut Leipner

If the top half of the crystal is slipping one plane at a time, then only a small fraction of the bonds are broken at any given time and this would require a much smaller force.

Radiation hardening – Temperature and dose

- **Temperature dependence – the ↓ the ↑ the embrittlement**
- **Dose dependence - ↑ the stronger irradiation effect, strong tendency to saturation**



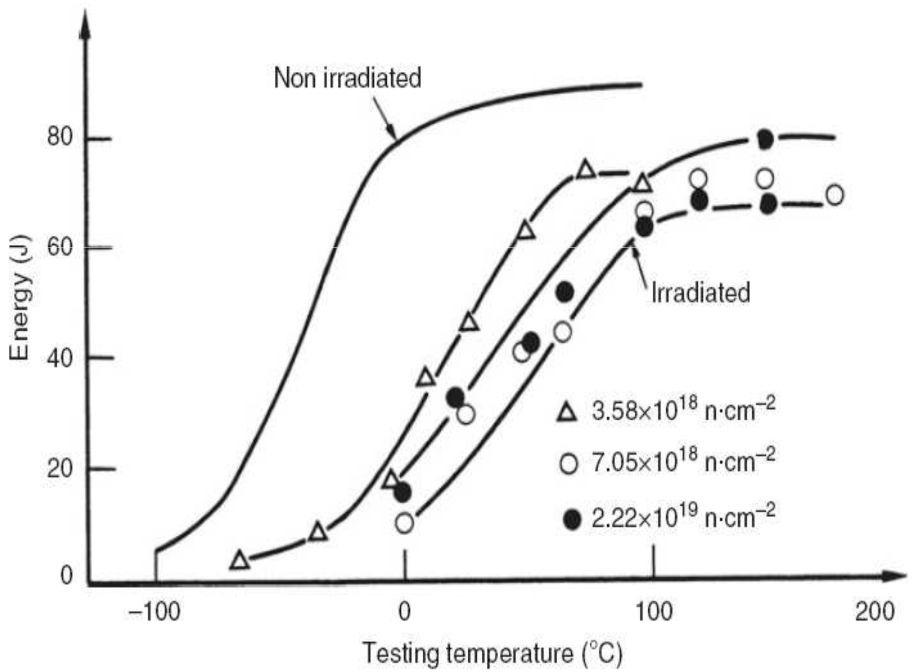


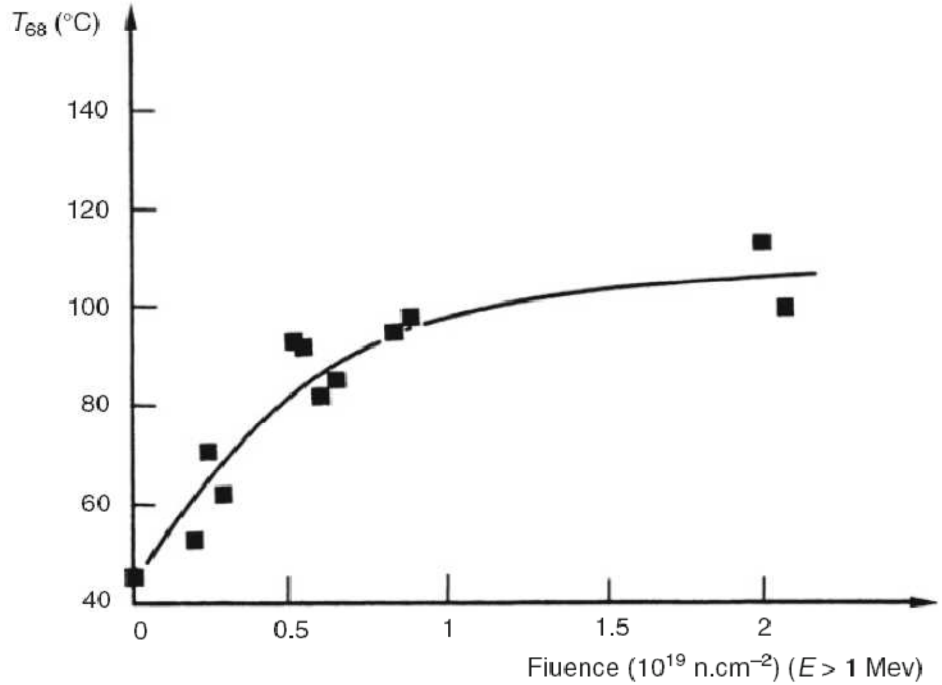
High hardening and loss of uniform elongation occurs for irradiation and test temperatures <0.3 T_M

Irradiation embrittlement

Pressure vessel – a critical property of the steel is its capability to resist:

- Crack propagation → Strict requirements with respect to fracture toughness and the DBTT
- Brittle fracture





Irradiation at temperatures below 0.3 to 0.4 T_M causes:

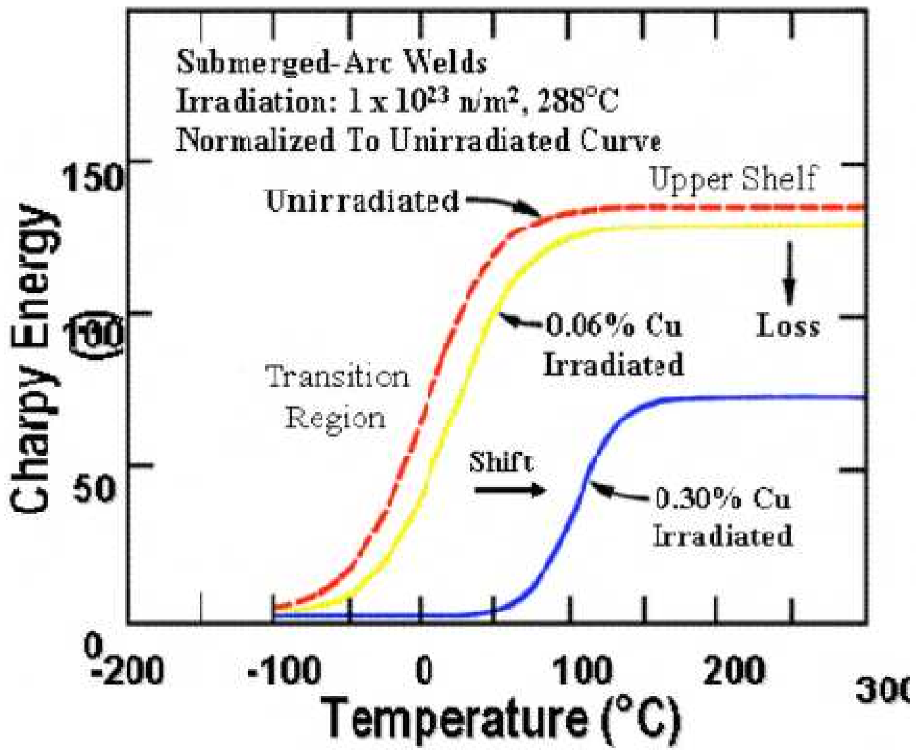
- Increases in the ductile to brittle transition temperature
- Reduction in upper shelf toughness of alloys such as 9-12% Cr F/M steels.

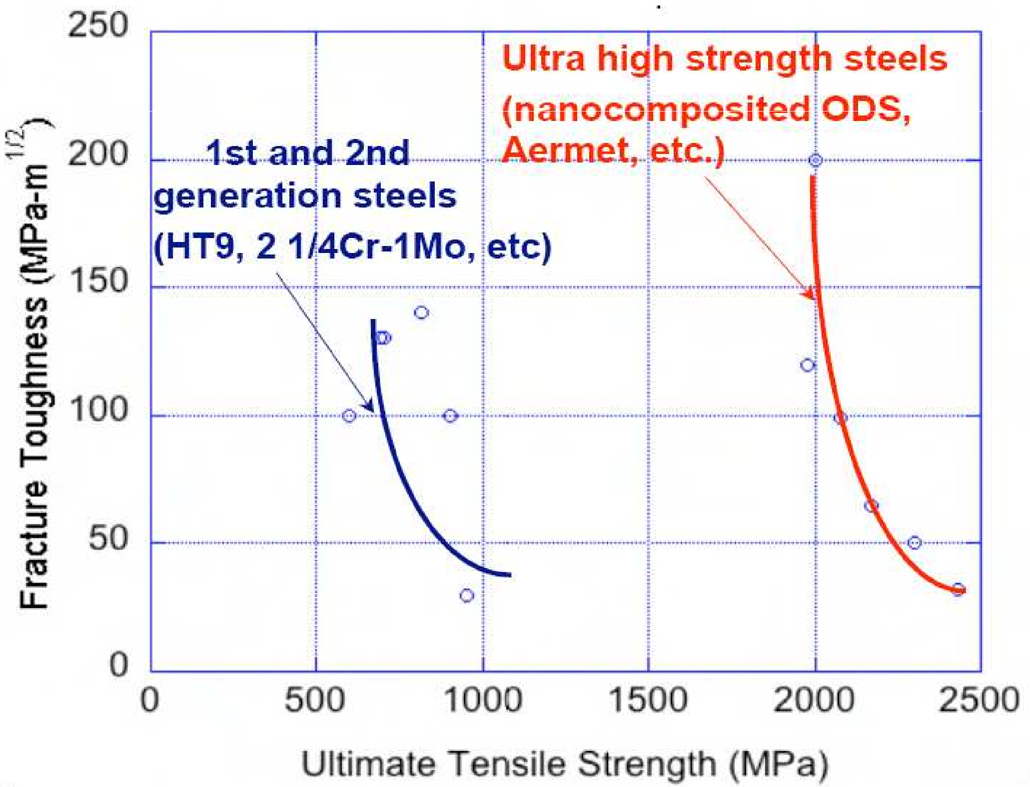
Radiation Hardening - Fracture Toughness

Radiation hardening can increase the flow stress above the critical value for ductile fracture – low fracture toughness

Two strategies to mitigate degradation in fracture toughness:

- **Specific alloying modification** - to reduce radiation hardening (e.g. low-Cu RPV steels)
- **Metallurgical changes** - to increase the critical stress for initiation of brittle cleavage fracture



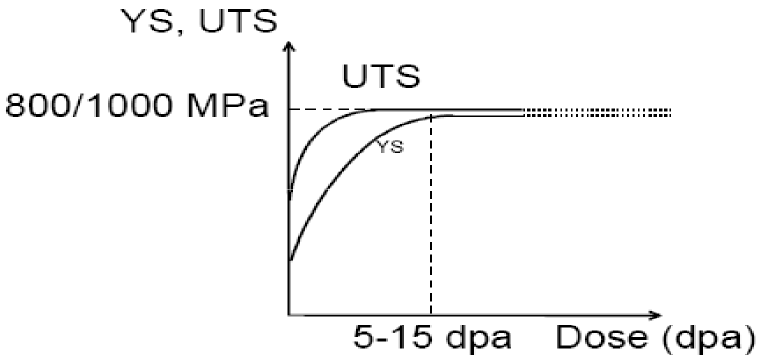


Radiation Hardening - Austenitic Stainless Steels in LWR

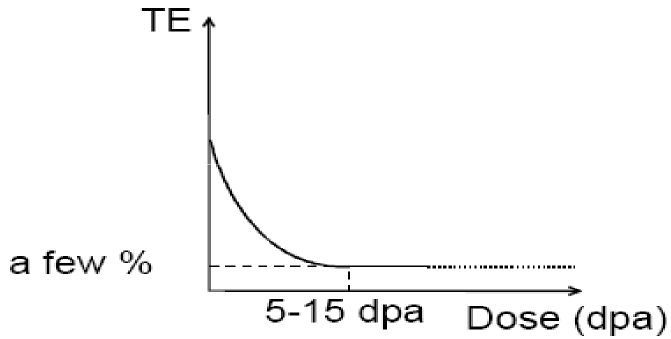
SS components close to the core – high neutron fluxes (e.g. baffles 2 dpa.y^{-1})

High density of dislocation (Frank) loops → ↑ in mechanical properties:

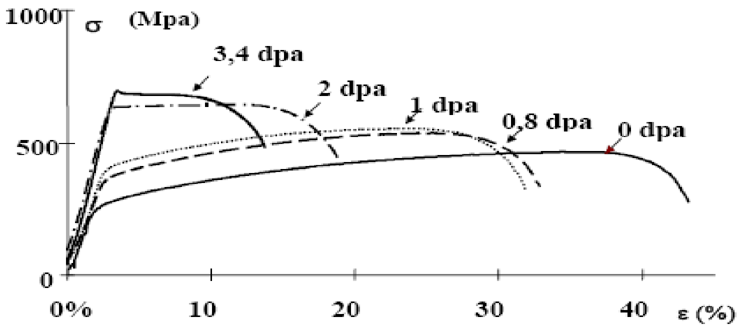
- **The YS and UTS close to 1000 MPa**
- **>10-20 dpa saturation occurs**



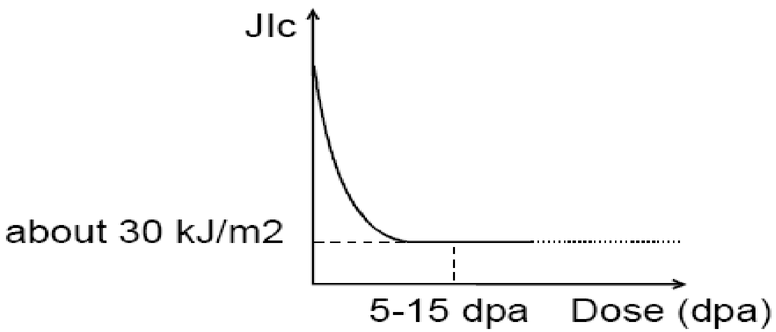
a) Yield strength - Ultimate tensile strength



b) Total elongation



c) Stress – strain curves



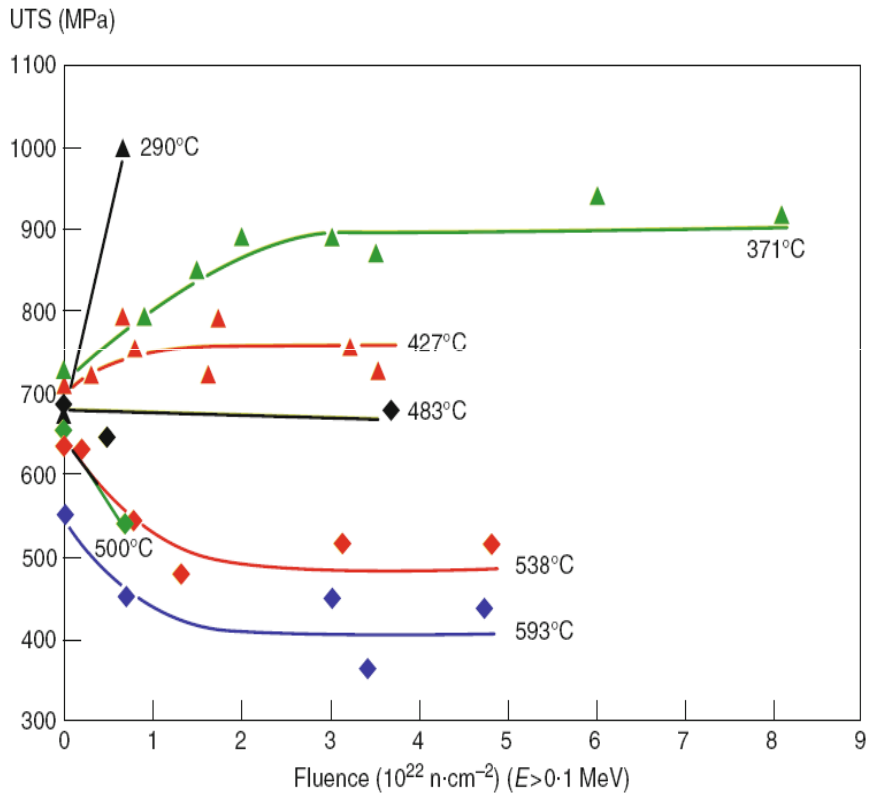
d) Fracture toughness

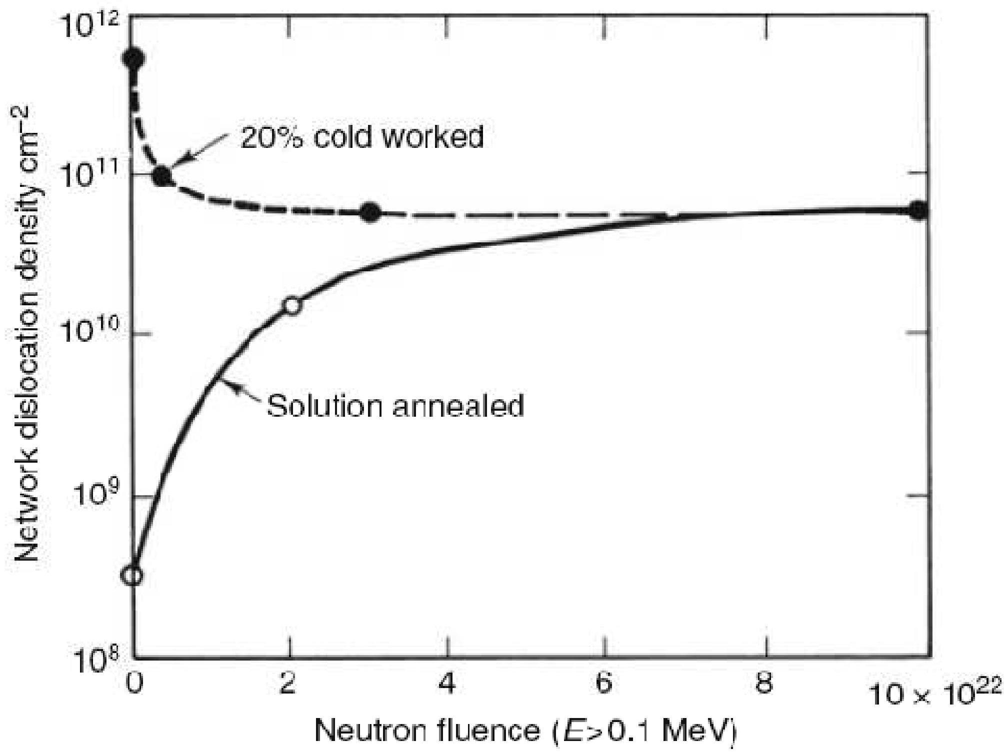
(Pokor 2010)

Radiation Hardening - Austenitic Stainless Steels in LWR

Higher irradiation temperatures:

→ **Recovery of the dislocation network resulting in a reduction of the yield strength**

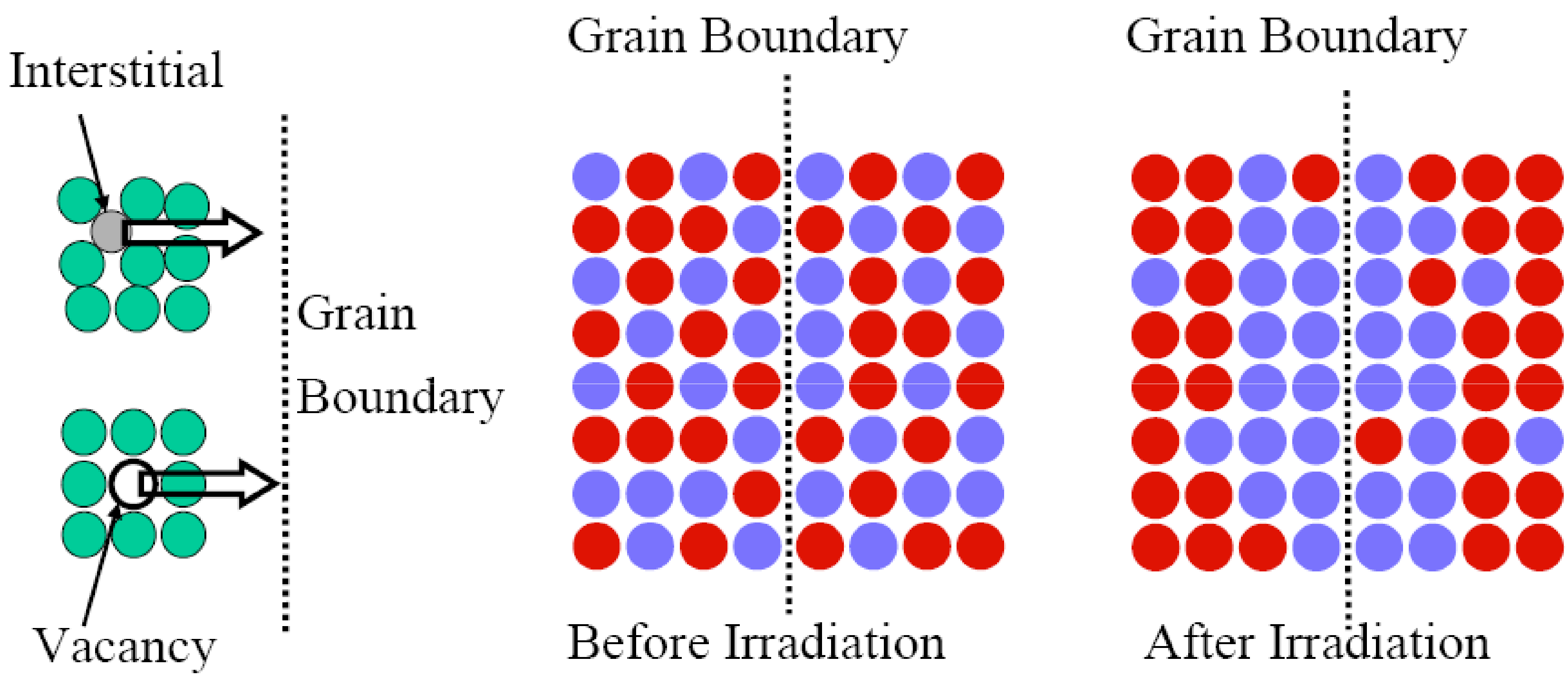




Mechanical properties of 316 SS CW under irradiation (Garner et al. 2010)

Saturation of dislocation density in both SA and 20% CW 316 SS at 500ºC (Brager et al. 1977)

The RIS observed in austenitic SS under specific conditions



Possible RIS Mechanism to Enrich Blue

- (blue) Migrates preferentially as interstitials
- (red) Exchanges preferentially with vacancies
- (blue) Vacancies drag species to sink

$$D^{I}_{Blue} > D^{I}_{Red}$$

$$D^{V}_{Blue} < D^{V}_{Red}$$

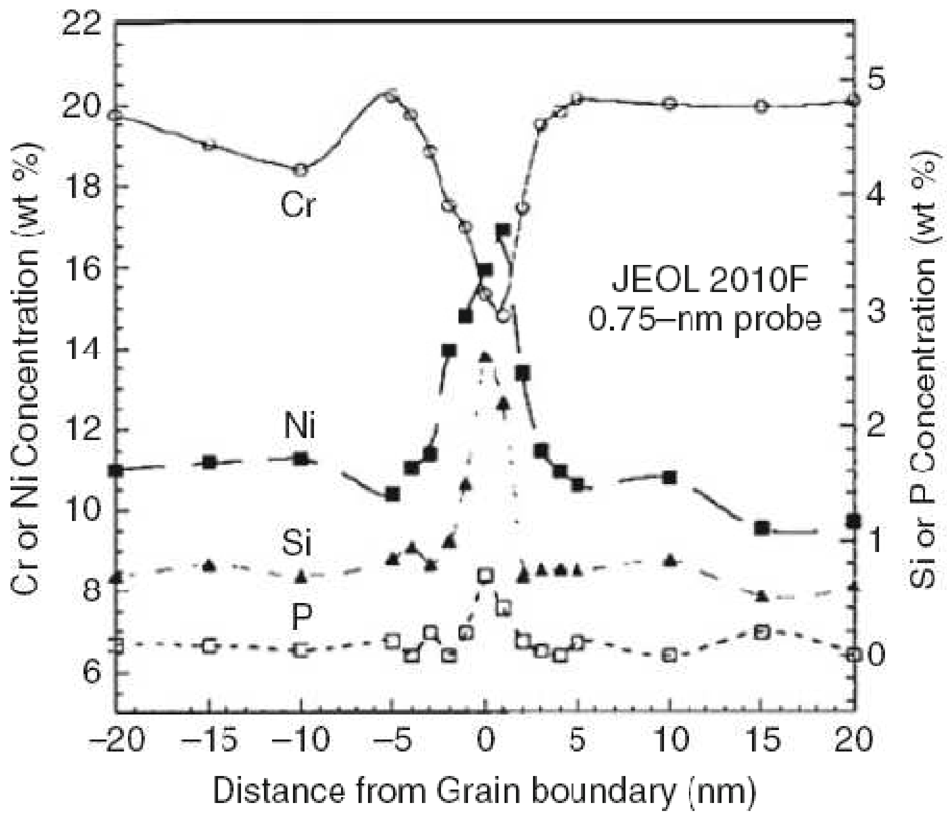
$$D^{V}_{Blue} > D^{V}_{Red}$$

J. Tucker

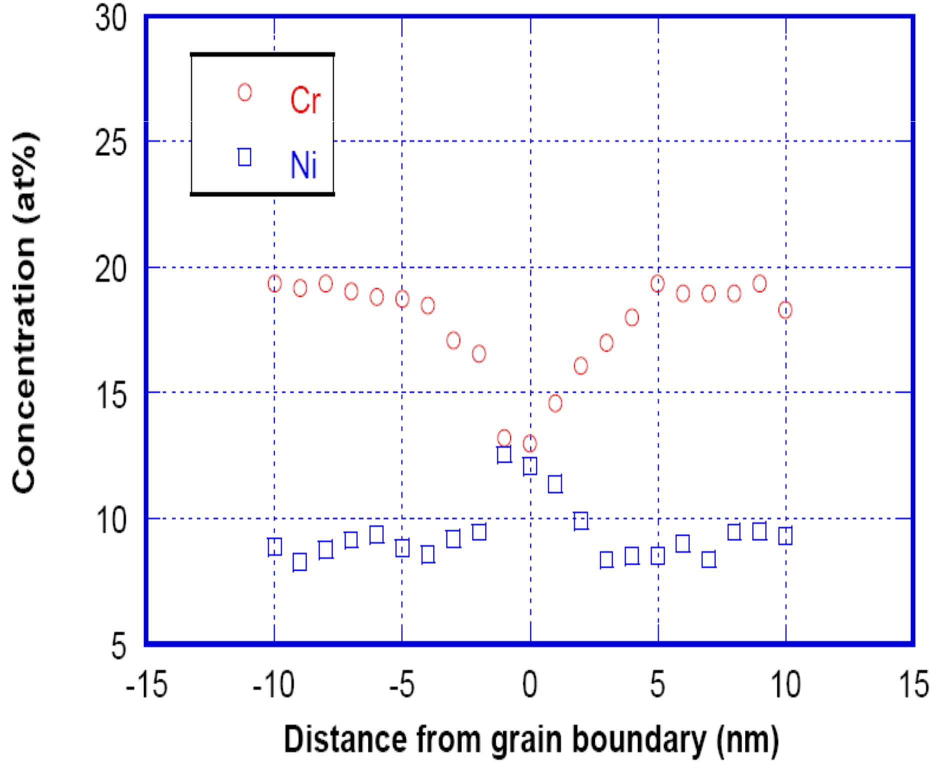
Radiation-Induced Segregation

Very narrow band of high segregations at GBs is observed

- **Interstitials (Si, P and S)** → **dragged by interstitials to GB**
- **"Inverse Kirkendall Effect"** → **migration of vacancies, a flux of atoms opposite direction (the fastest diffusion rates)**

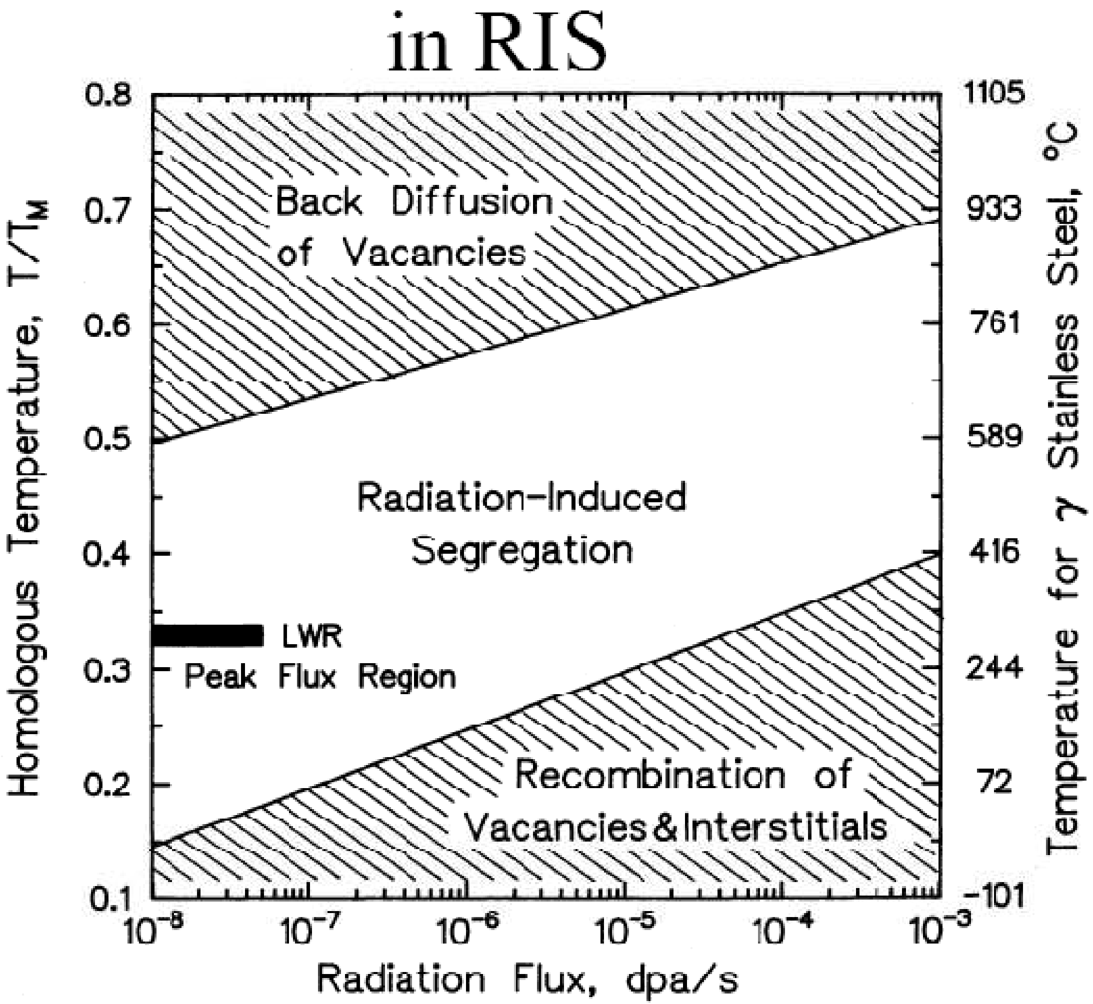


304 SS, proton irradiated at 400°C to a dose of 1.0 dpa



The competition between PD generation and recombination

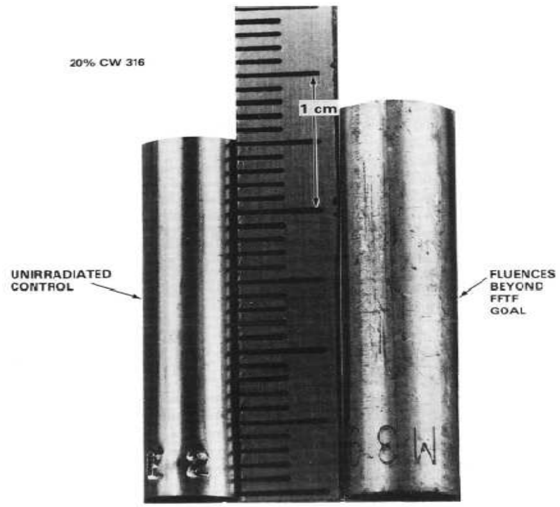
→ **temperature/dose rate dependence of RIS**



Swelling

Swelling – a homogenous decrease of the density of the alloy leading to:

→ **Increase of the dimensions of the components**



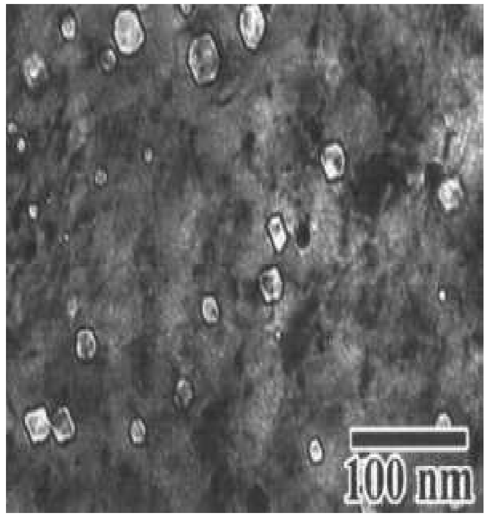
- **Occurs after a minimum incubation time**
- **Is significant in a given temperature range**
- **Is strongly dependent on the metallurgical state of the alloy**

The mechanism of swelling:

- **The clustering of vacancies in 3D cavities**
- **Nucleation ↑ by He from (n, α) reactions**

(austenitic SS the high Ni content)

- **The clusters of He atoms – nanometric bubbles**
- **Bubbles grow as cavities**



Critical concern:

→ **The geometry and performance of the core**

Swelling

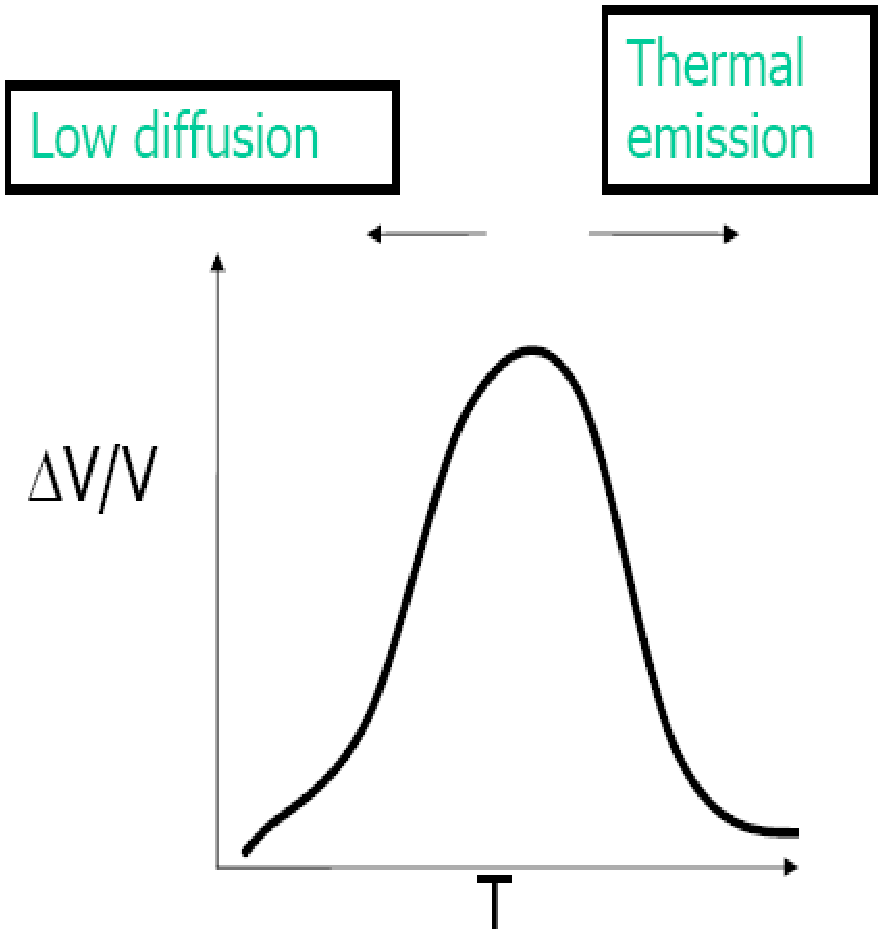
Temperature regimes:

❑ Low temperature – swelling not observed

→ low migration rate of vacancies

❑ High temperature – swelling disappears

→ thermal emission of vacancies from cavities



Most metals swell in temperature range of:

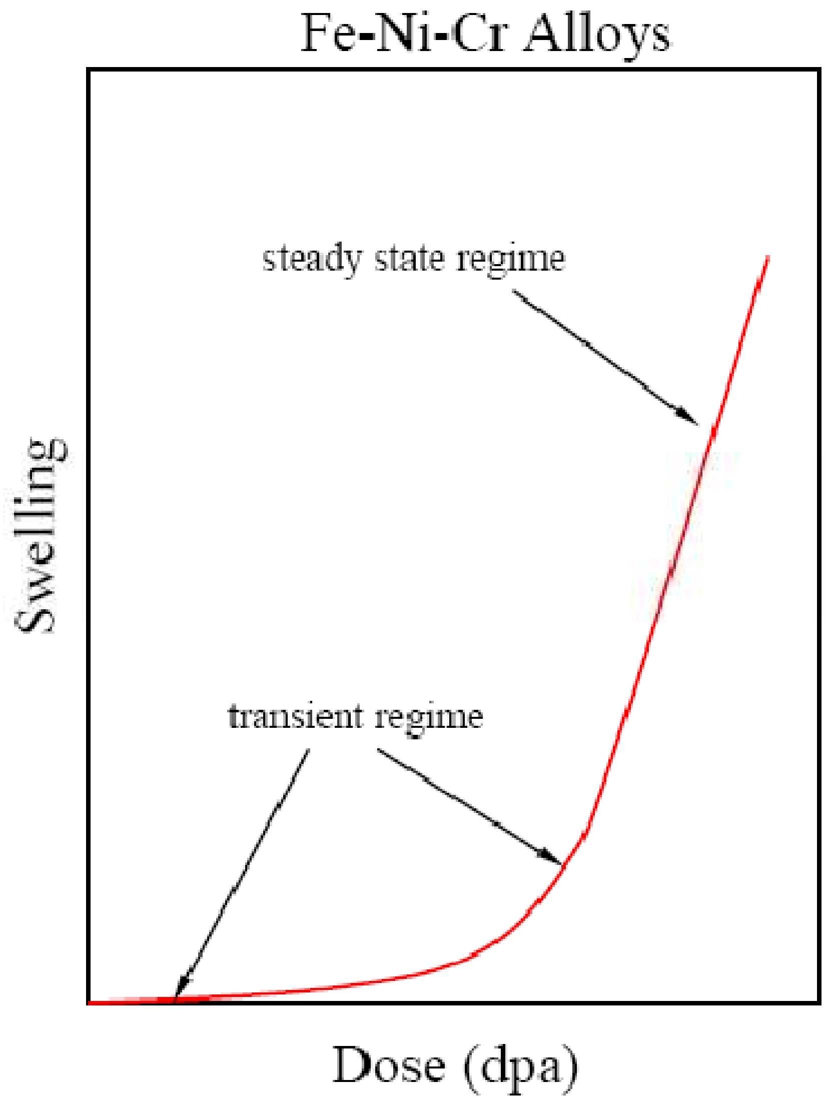
$$0.3T_M < T < 0.55T_M$$

Swelling

The crystal structure affects the swelling behaviour

A constant swelling rate of:

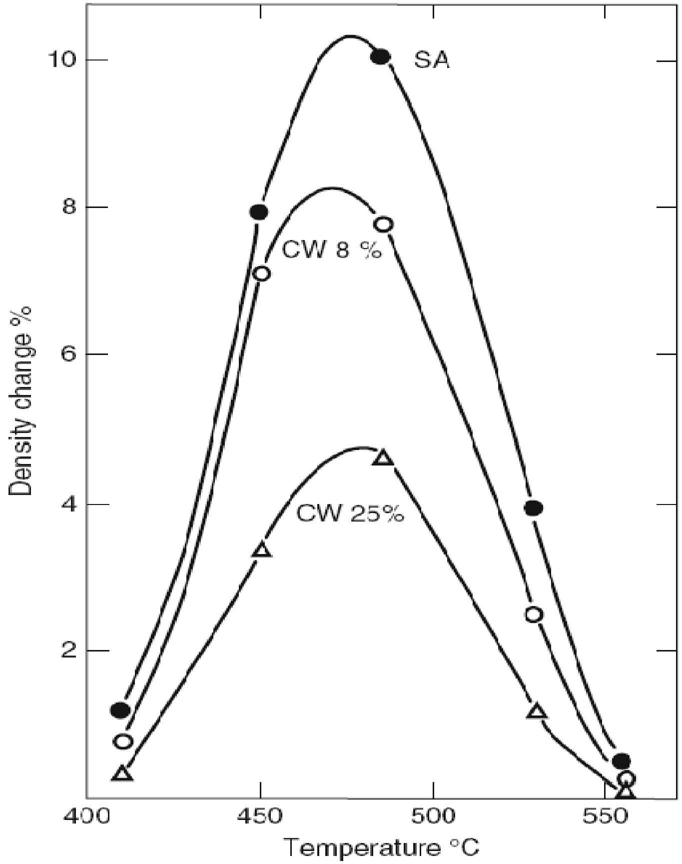
- ~ 1% x dpa^{-1} in austenitic SS
- < 0.1 x dpa^{-1} in ferritic steels (e.g. Fe-Cr alloys)
- Lower swelling rates for more complex alloys
- Hcp alloys (Zr, Ti, etc.) no swelling



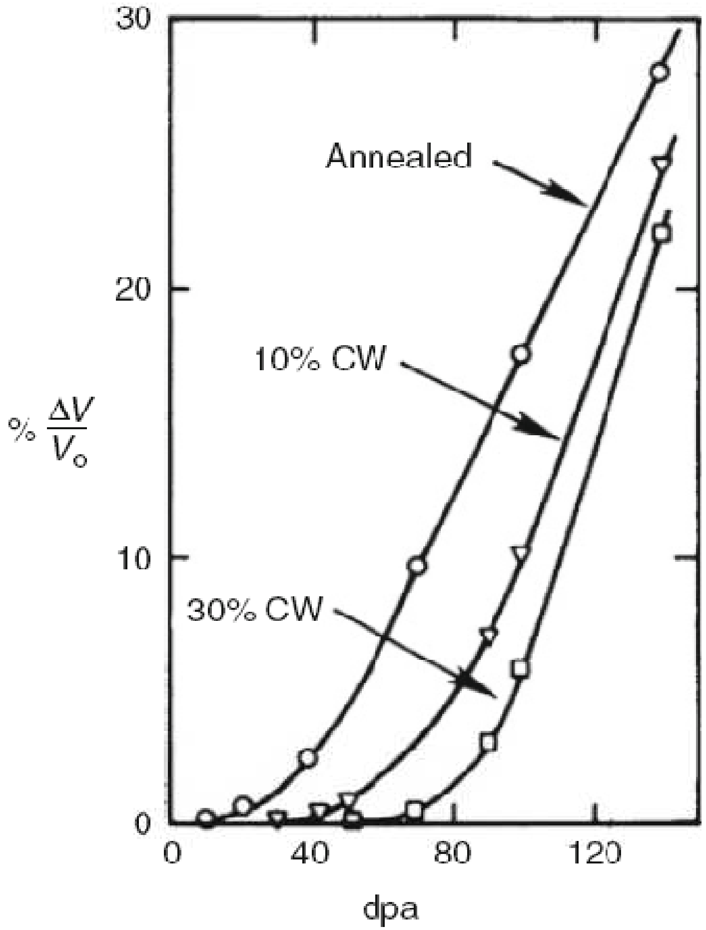
Swelling

Reduction of swelling is obtained in cold worked materials

by delay of the onset of swelling



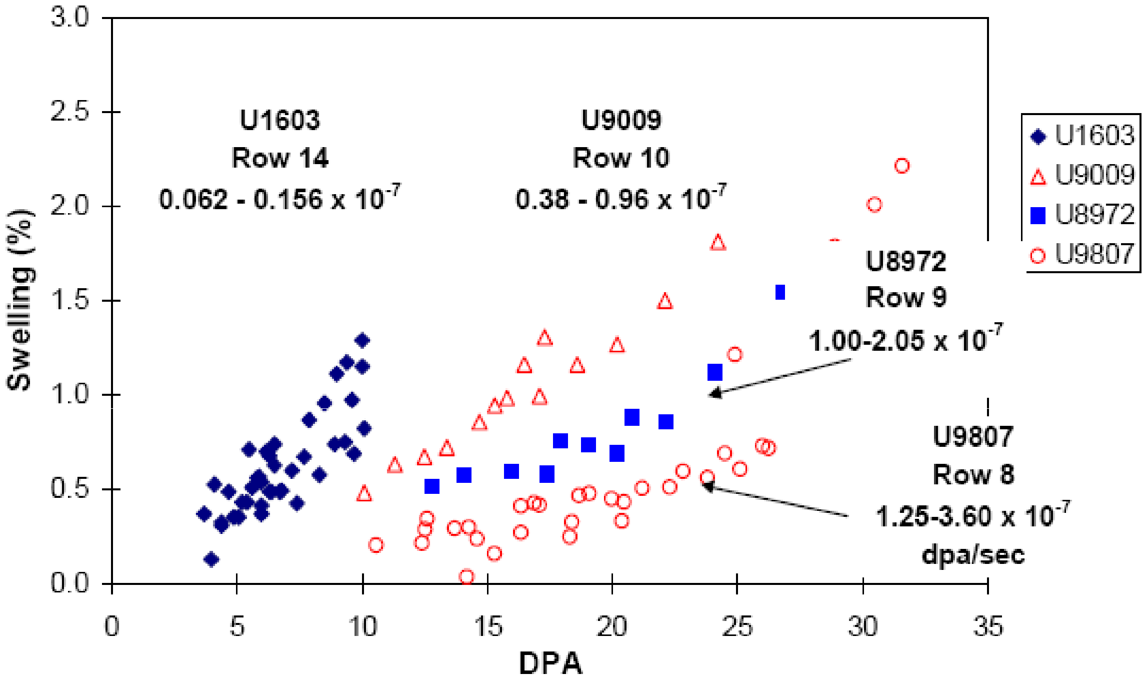
316 SS irradiated at SFR (Dupouy 1978)

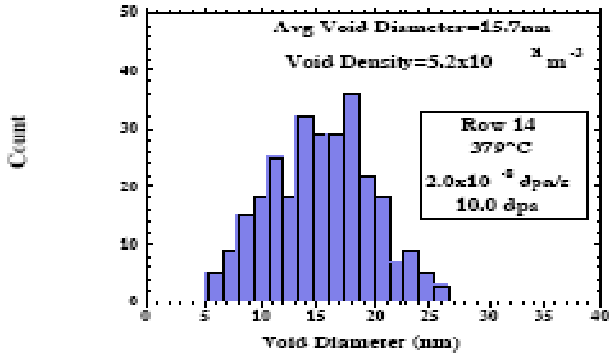


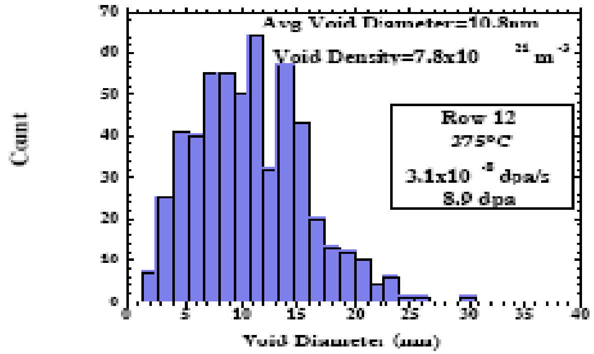
15-15 austenitic SS irradiated at 650ºC with 1MeV Cr ions (Garner 1993)

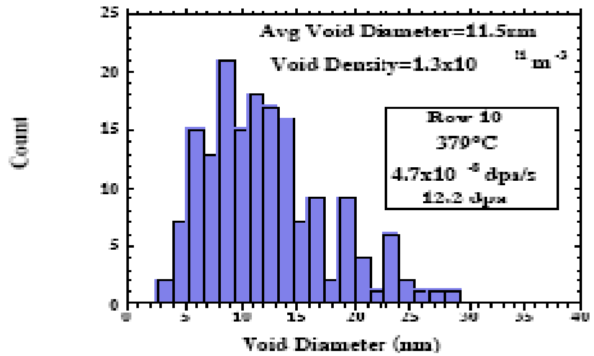
Swelling

Dose rate







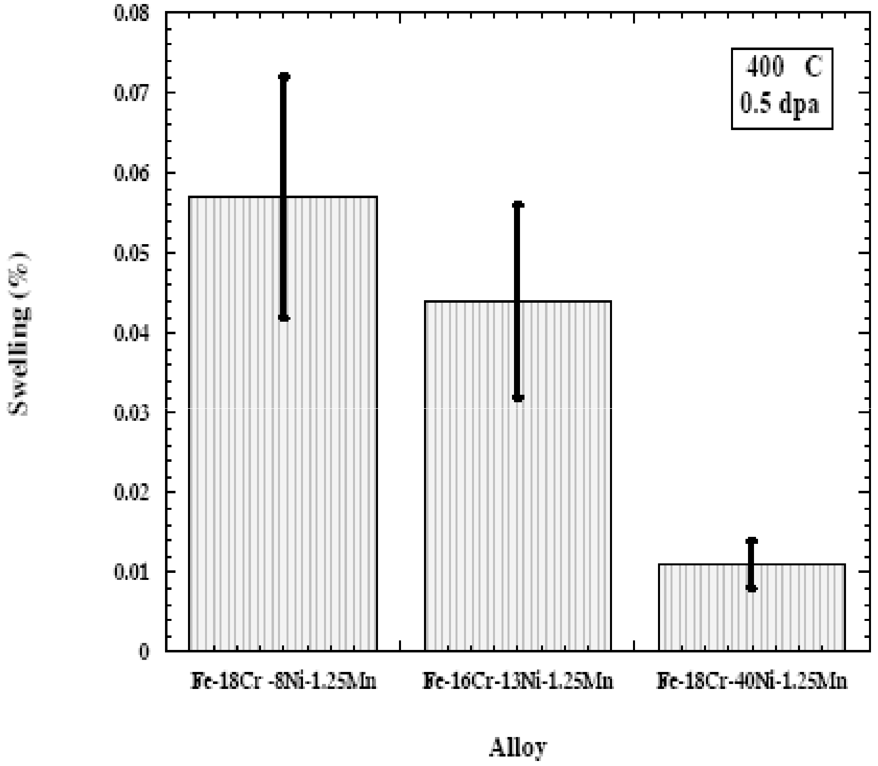


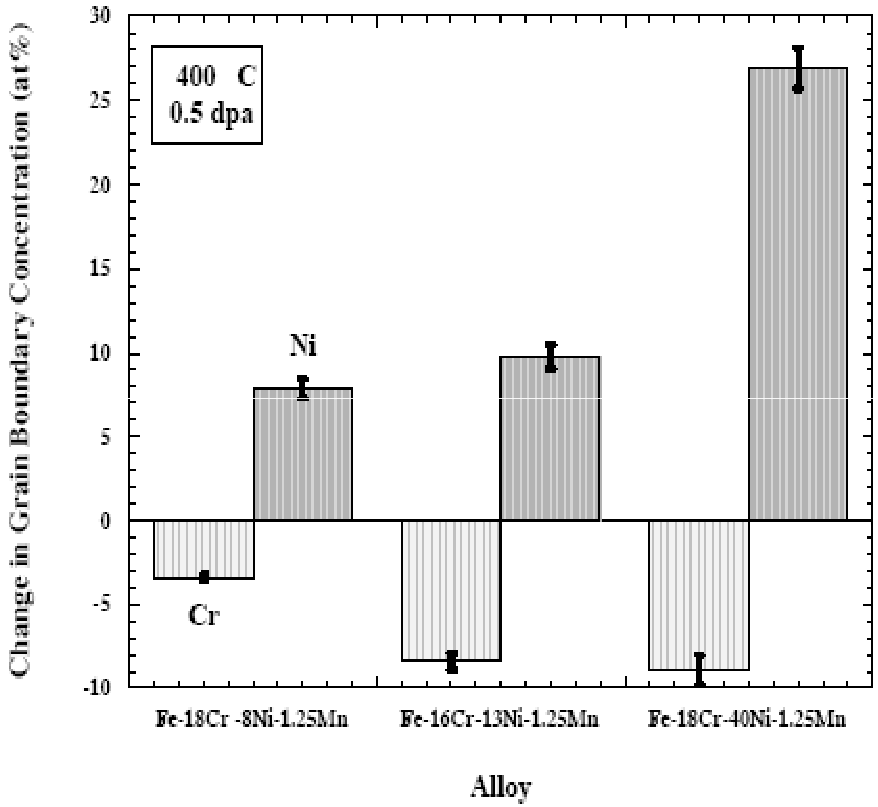
High dose rate:

→ **the peak swelling at higher temperatures**

(Allen 2010)

Segregation and swelling

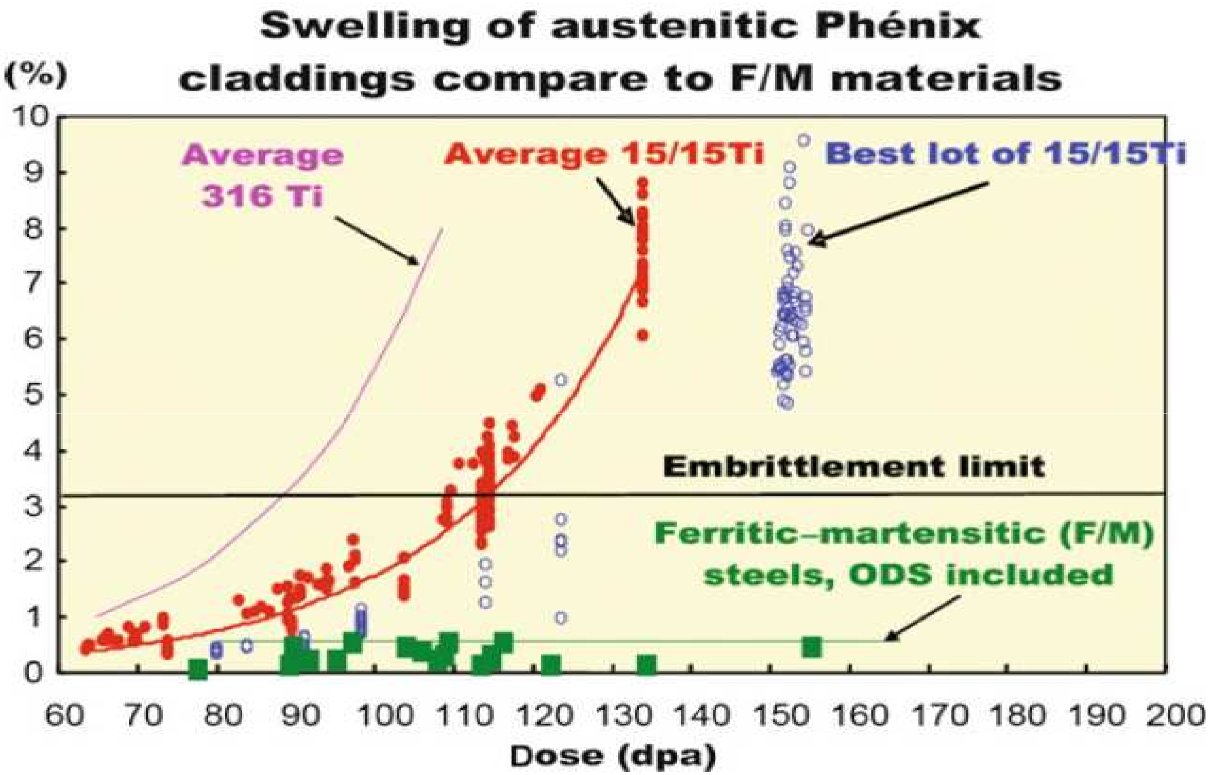




Increasing the Ni content reduces the swelling

(Allen 2010)

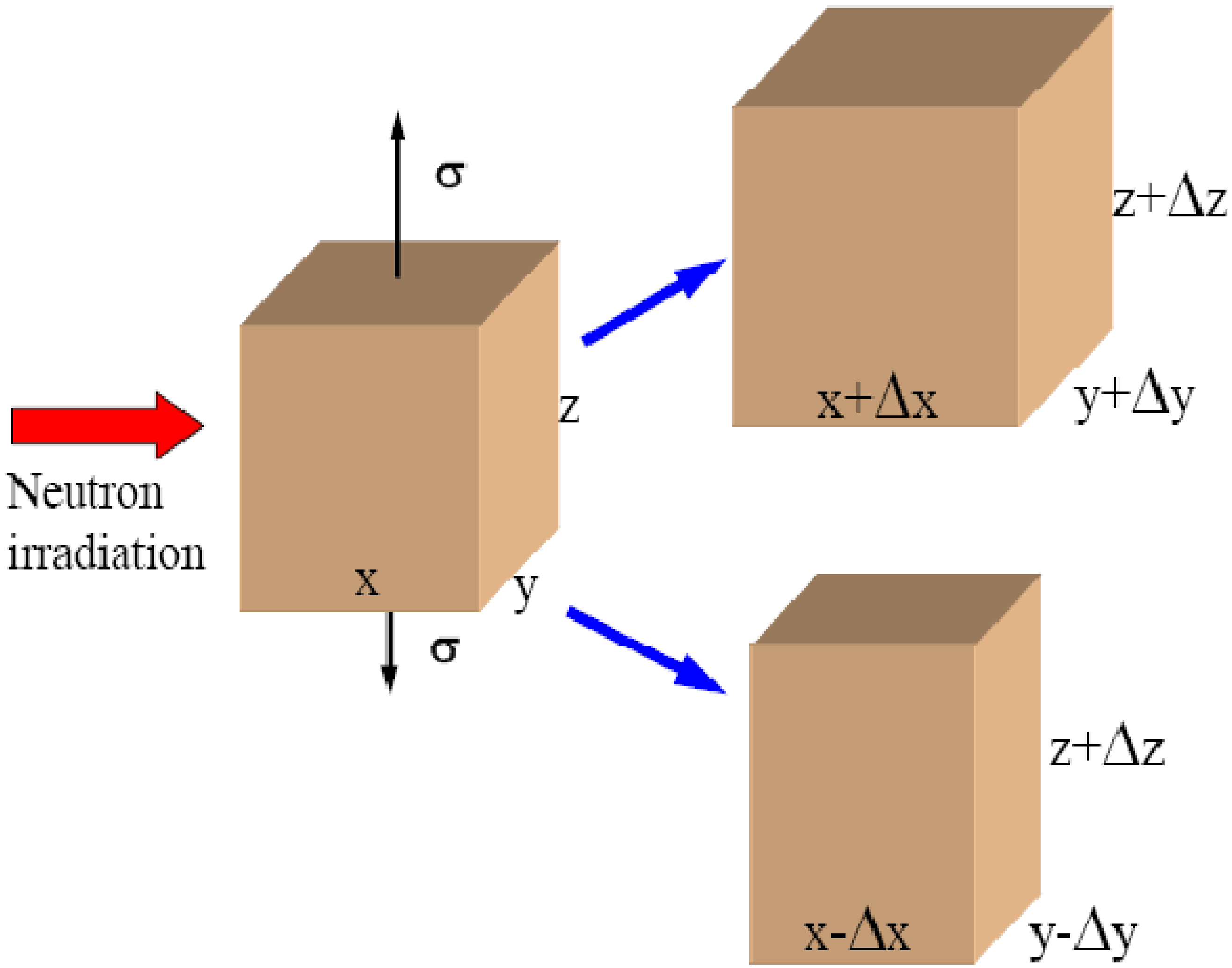
Historical evolution of the performance of SS cladding for the French SFR



The Gen-IV will require very low swellings at very high doses

(Allen 2010)

Swelling & Irradiation Creep



Swelling:
- Volume increase
- Isotropic Shape Change
- Radiation-induced

Creep:
- No volume change
- Shape Change
- Radiation-influenced

Irradiation Creep

❑ Primary state:

- Short-term transient state, poorly characterised

❑ Secondary state:

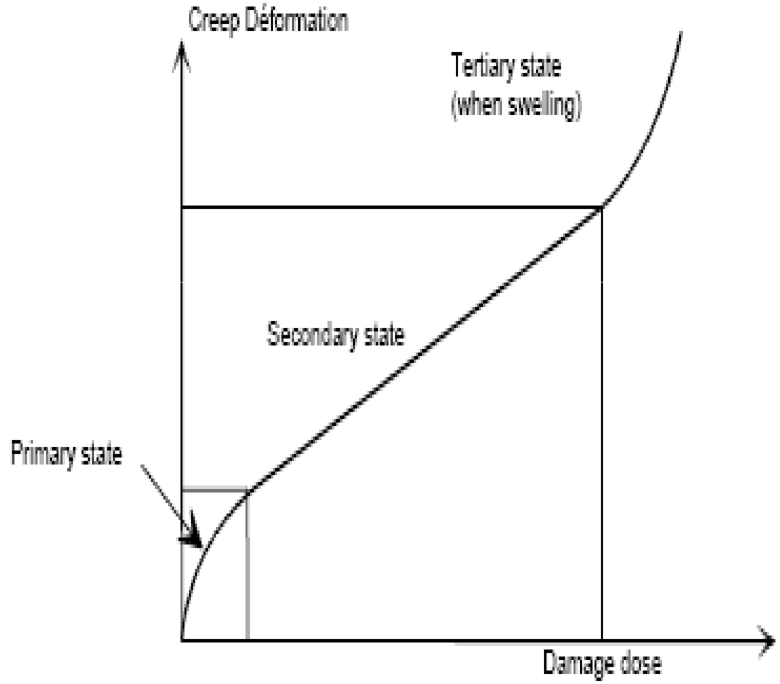
- Retained until swelling appears on the material
- Plastic deformation proportional to the dose and the stress:

$$d\varepsilon_F = B_0 \,.\, \sigma \,.d\varphi$$

εF : Creep deformation

φ : Fluence

Bo : Creep compliance



❑ Tertiary state:

- Creep rate acceleration due to swelling:

$$d\varepsilon_F = (B_0 + D \,.\, dV/d\varphi) \,.\, \sigma \,.d\varphi \,,$$

dV/dφ : Instantaneous swelling rate

❑ Radiation produced point defects increase diffusion and allow creep at lower temperatures

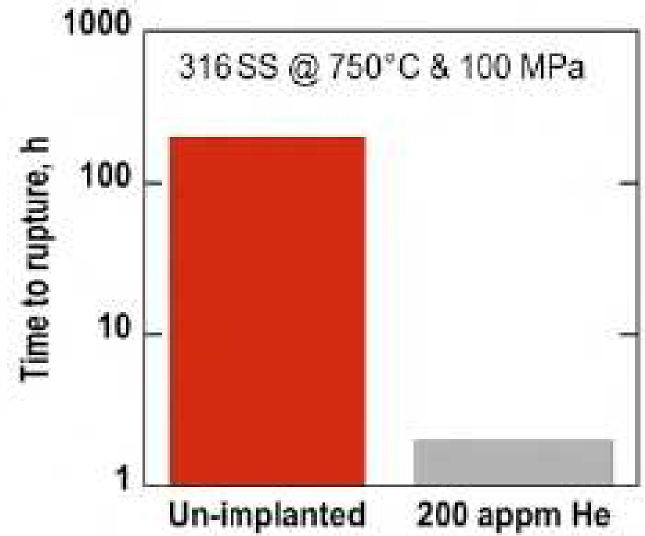
High Temperature Helium Embrittlement

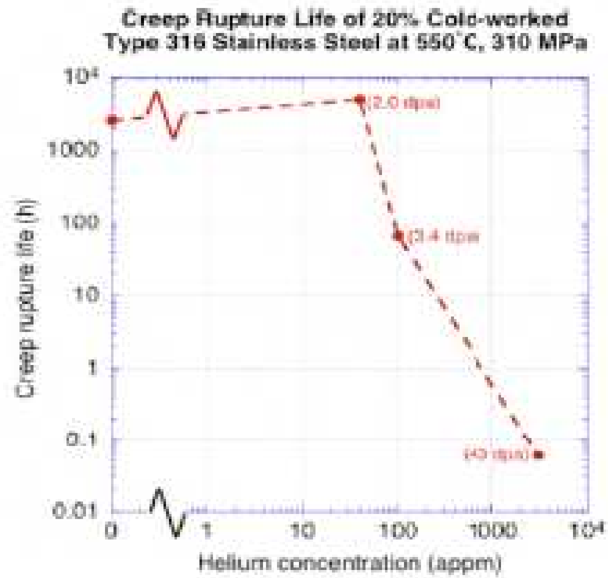
Institute for Energy

Mechanism:

- He ($^{10}B(n,\alpha)^{7}Li$) is highly insoluble
- He preferentially nucleates at grain boundaries in the form of cavities as long as:
  - $T > 0.5\ T_M$
  - Applied stress
- Can lead IG fracture due to transformation of the He bubbles to voids
- The critical amount of He to induce grain boundary embrittlement:
  - ↓ with ↑ test temperature and ↓ strain rate.
  - F/M steels > SSs

- Introduction of high densities of nanoscale that serve to trap migrating He before it can reach grain boundaries had been found to be effective in mitigating He embrittlement

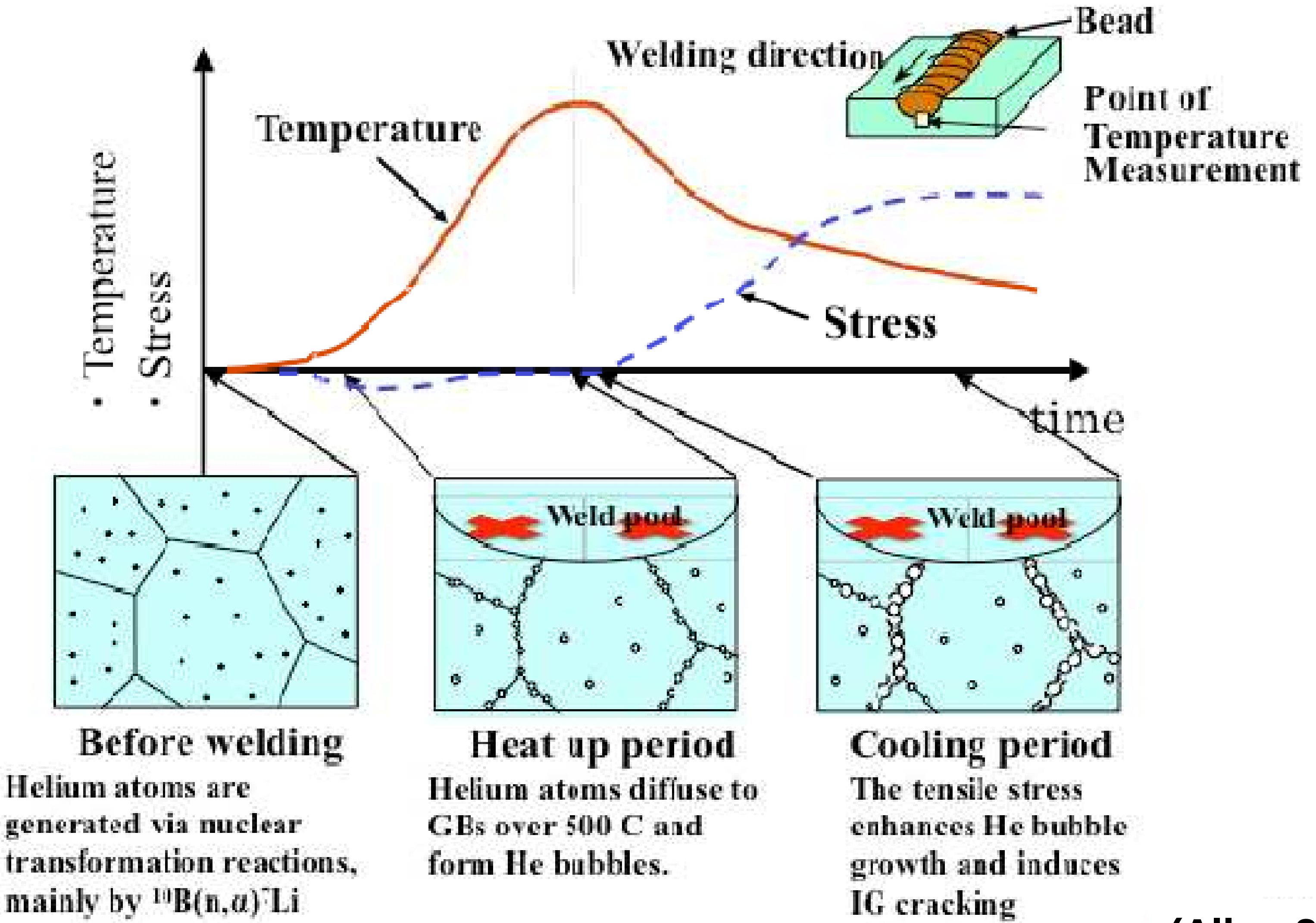




(Allen 2010)

High Temperature Helium Embrittlement

Mechanisms of Helium-Induced Weld Cracking



(Allen 2010)

HPLWR Phase 2 - SCW Loop

- The purpose is to minimize the corrosion of structural materials and hence mass transport of corrosion products within the water/steam cycle
- Strict control of impurities in water/steam cycle
- Transition to SCW causes strong decrease of impurity solubility and hence formation of deposits

- HPLWR – hydrogen water chemistry very likely

During water radiolysis a number of transient and stable products are produced. The initial reaction can be summarized with the equation:

$$H_2O \overset{irr}{\rightarrow} e^-_{aq}, {}^{\bullet}OH, H^{\bullet}, H^+, OH^-, H_2O_2, H_2$$

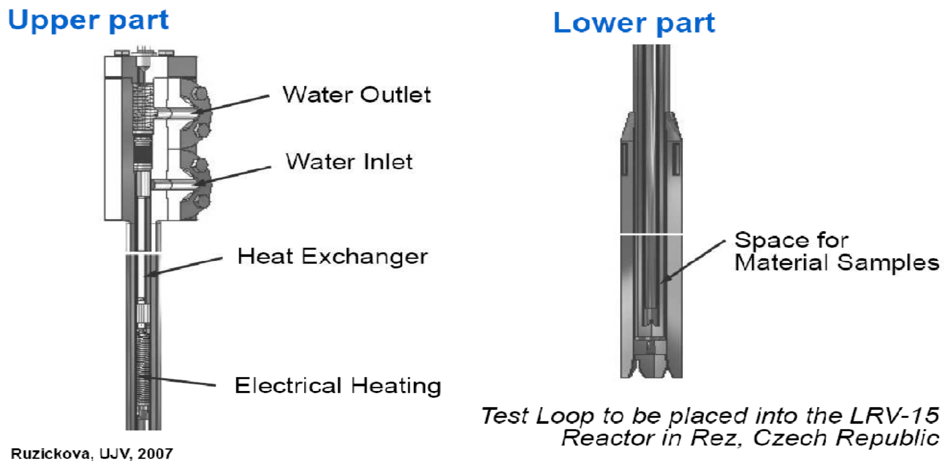
1. The concentration of the transient and stable species may be different
2. Chem. Potential of O_2 and H_2O_2 could affect corrosion potential of the water and thus oxide layer morphology

- The maximum content of radiolytic water decomposition product no higher than BWR (200-300 wppb O_2)
- HWC should reduce the radiolytic oxygen production but it might require much more hydrogen which could cause metal hydriding

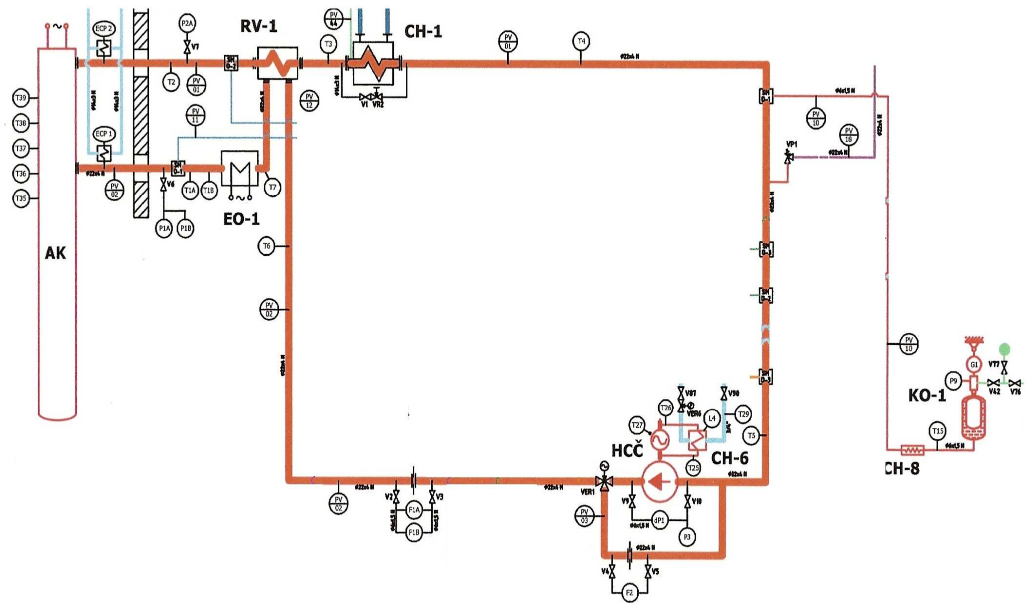
HPLWR Phase 2 - IASCC

Main targets:

- ❑ Corrosion studies
- ❑ Testing and optimization of suitable water chemistry
- ❑ Coolant radiolysis studies
- ❑ Development and testing of sensors



Test Loop to be placed into the LRV-15 Reactor in Rez, Czech Republic



LOOP: MAIN PARAMETERS:

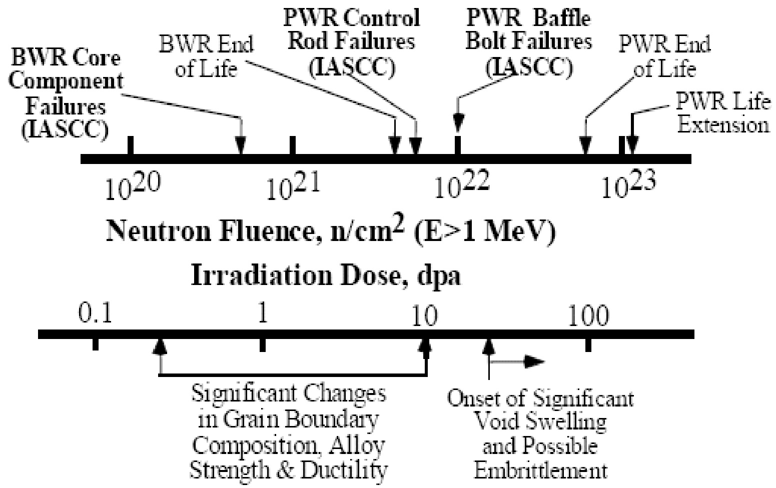
PRESSURE: 25MPa; max. 32MPa.
TEMPERATURE: max. in active channel 600℃; max. in loop 390℃.
FLOWRATE IN ACTIVE CHANNEL: 200kg/h.
FLOWRATE IN LOOP: 200kg/h.
TOTAL VOLUME: 42dm3.
FILTRATION RATE: 30kg/h.
SAMPLING: 0.2kg/h.
ON-LINE MEASUREMENT: 2 x 12kg/h (HIGH-PRESSURE AND LOW-PRESSURE CIRCUITS).

IASCC – Irradiation Assisted Stress Corrosion Cracking

- **Terminology used to describe cracking of materials exposed to nuclear reactor coolant and ionizing radiation.**

- **Like all Stress Corrosion Cracking phenomena it requires critical combinations of applied stress or strain, environmental chemistry and metallurgical structure to occur.**

Microstructure Changes can be Correlated to Irradiation Dose/Fluence



Reactor Type	Inlet Temp (°C)	Outlet Temp (°C)	Maximum Dose (dpa)	Pressure (Mpa)	Coolant
PWR	290	320	100	16	Water
SCWR	290	500	15-67	25	Water
VHTR	600	1000	1-10	7	Helium
SFR*	370	550	200	0.1	Sodium
LFR*	600	800	200	0.1	Lead
GFR*	450	850	80	7	Helium/ SC CO_2

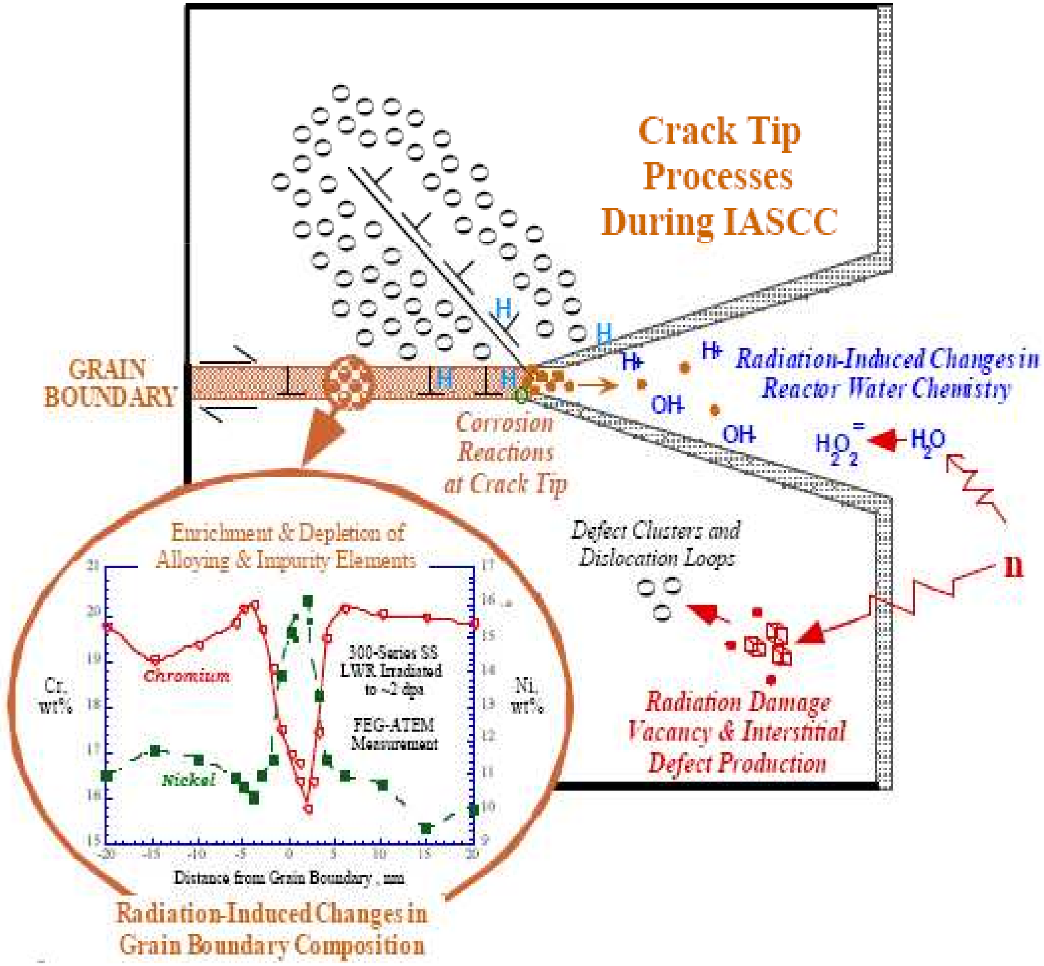
IASCC

❑ IASCC added feature to EAC:

by virtue of atomic displacements, neutron irradiation significantly alters the metallurgical microstructure and ionizing (α,β and γ) radiation can modify the environmental chemistry.

❑ Effects of irradiation on SCC:

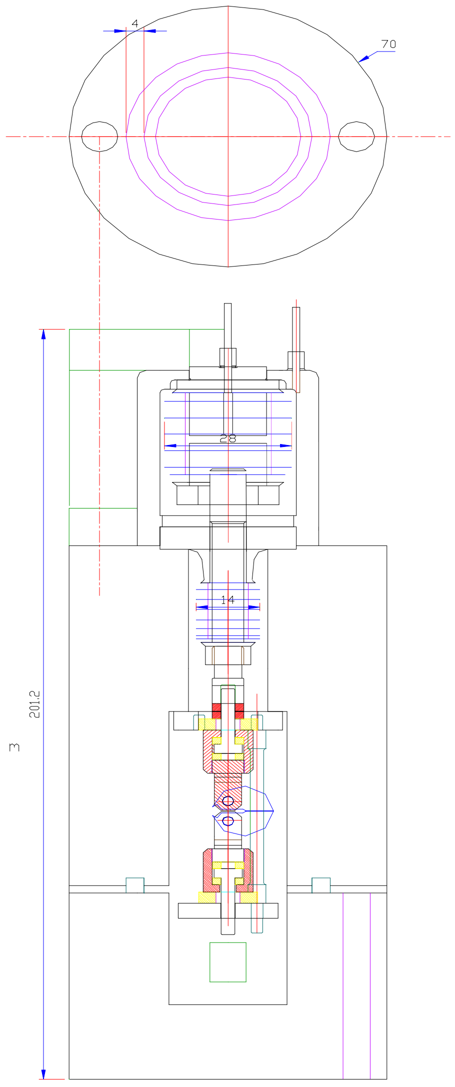
- **primary defects**
- **defects segregation**
- **dislocation interaction**
- **grain boundaries**
- **localized stress and strain**
- **environment**
- **stress relaxation by irradiation creep (beneficial factor for IASCC)**



IASCC

❑ Miniature size of autoclave-bellows system for SCC (Stress Corrosion Cracking) test in LWR, SCWR and LFR environmental conditions.

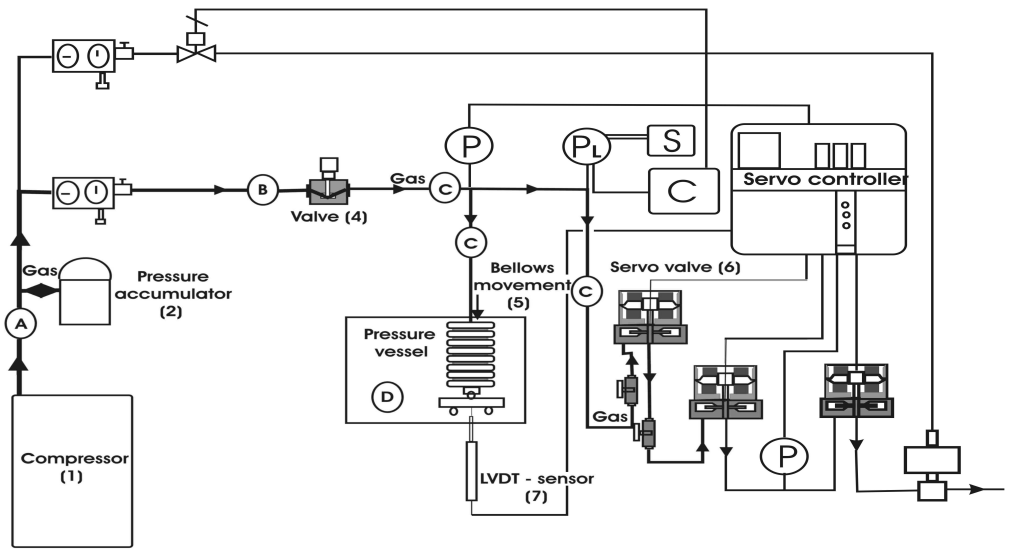
Idea is to design and develop testing system for SCC and IASCC material testing both pre-irradiated (hot cells tests) samples and samples irradiated in-situ (in-pile) facilities.



❑ The main innovation:

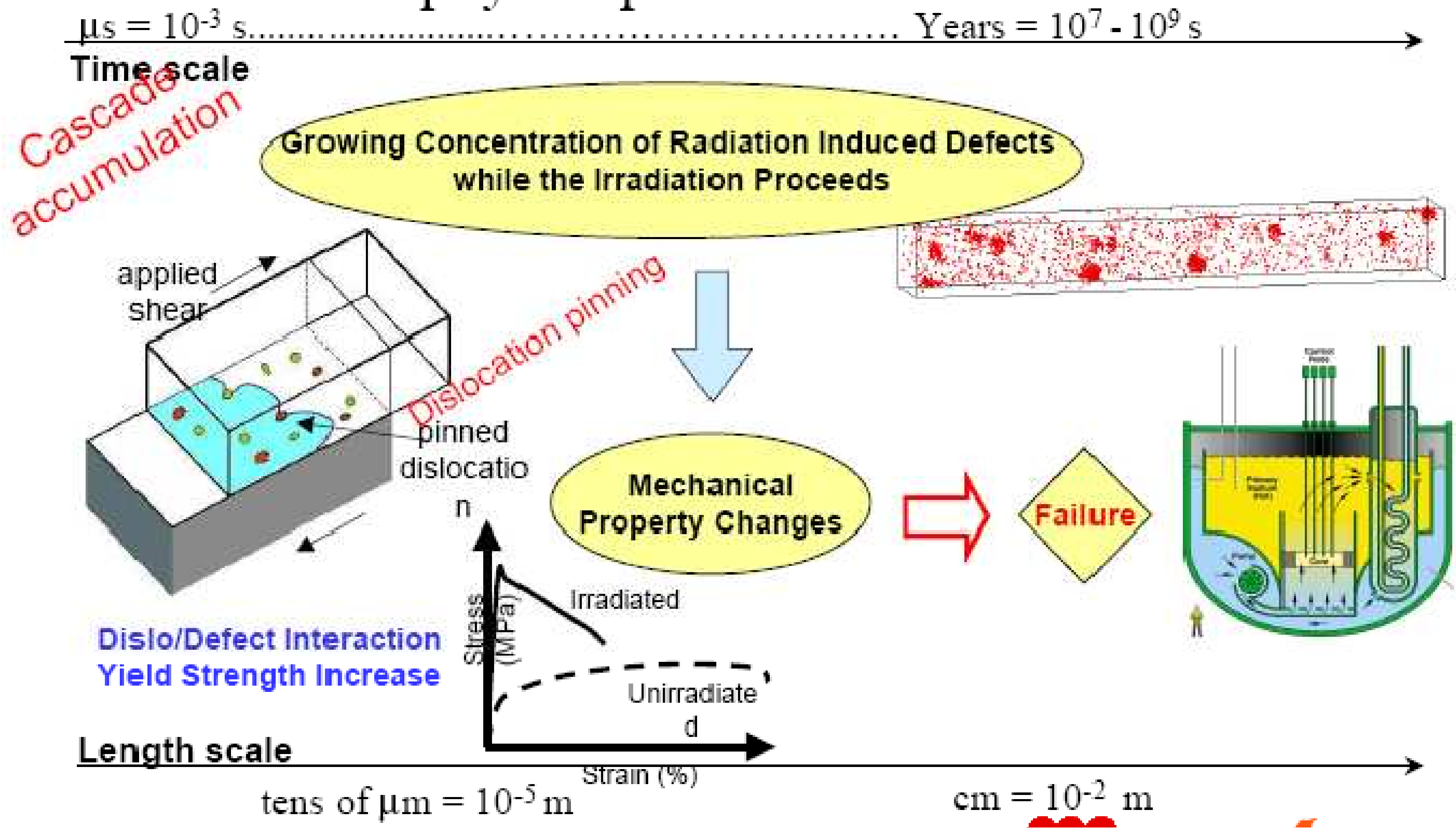
-New design of bellows based loading system

-New design of pressure adjusting loop

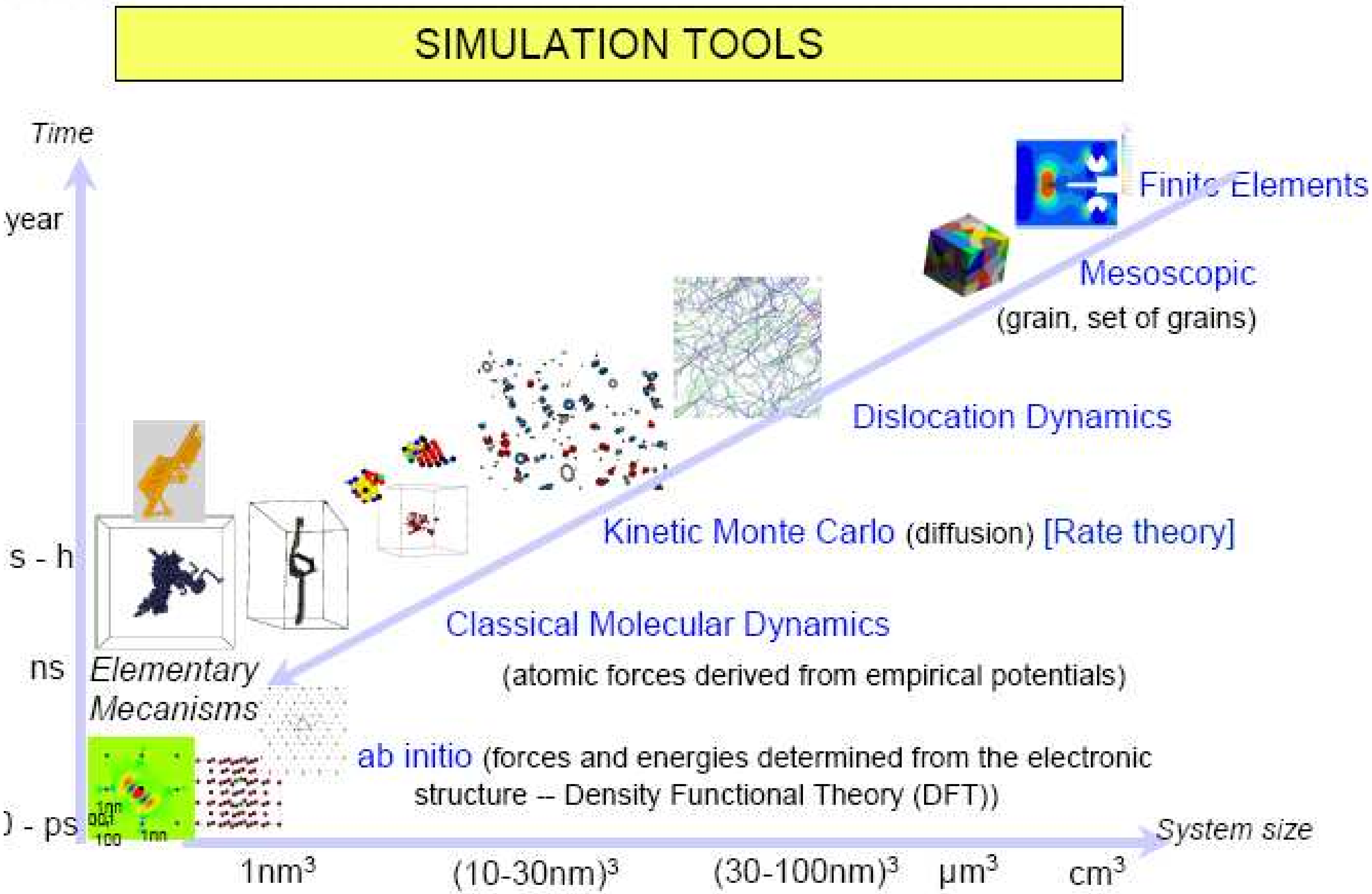


Summary

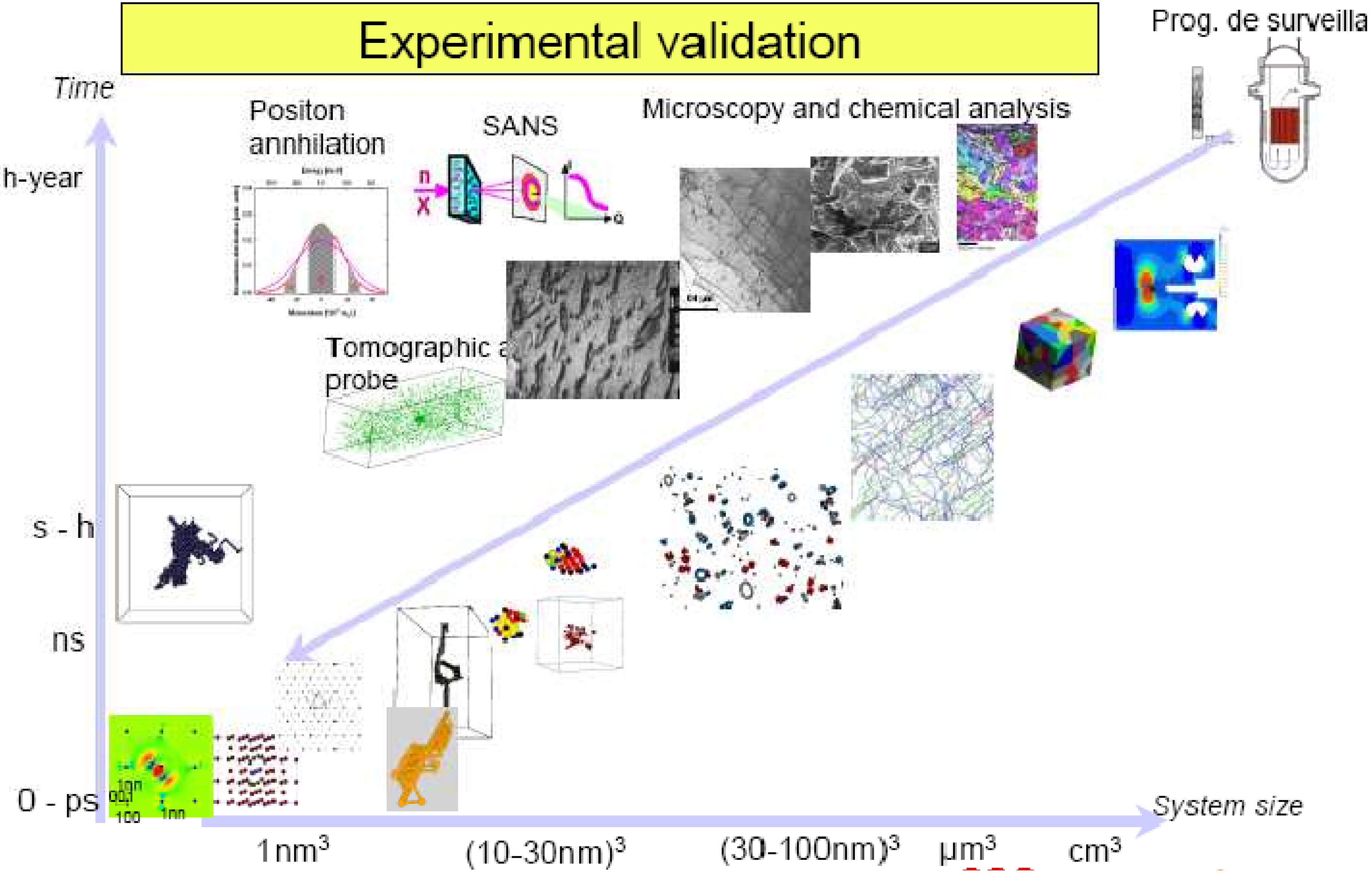
Radiation effects are inherently multiscale and multiphysics phenomena



Summary


SIMULATION TOOLS
Time
year
s - h
ns
) - ps
Finite Elements
Mesoscopic
(grain, set of grains)
Dislocation Dynamics
Kinetic Monte Carlo (diffusion) [Rate theory]
Classical Molecular Dynamics
(atomic forces derived from empirical potentials)
Elementary
Mecanisms
ab initio (forces and energies determined from the electronic
structure -- Density Functional Theory (DFT))
System size
1nm³
(10-30nm)³
(30-100nm)³
µm³
cm³

Summary


Experimental validation
Prog. de surveilla
Time
Positon
annhilation
SANS
Microscopy and chemical analysis
h-year
Tomographic a
probe
s - h
ns
0 - ps
System size
1nm^3
(10-30nm)^3
(30-100nm)^3
µm^3
cm^3