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RADIATION DAMAGES IN MATERIALS – PART II

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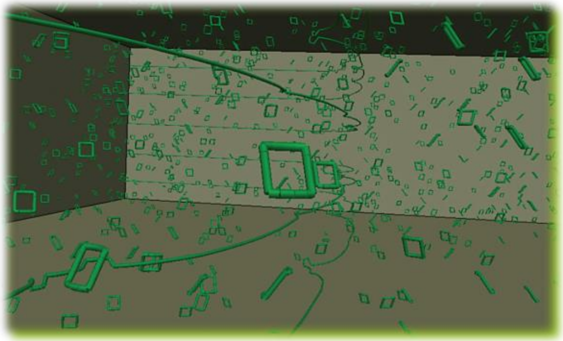
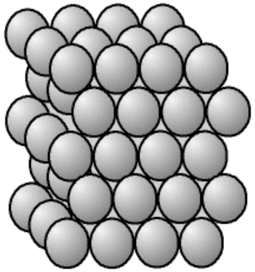


Outline

1. Background on alloys and radiation effects
2. Radiation hardening: example of PWR pressure vessel steel
3. Radiation swelling: example of fast reactor cladding
4. Creep irradiation: example of fast reactor cladding… cont'd
5. Radiation growth: example of LWR Zr-alloy cladding
6. Conclusions

Structure of metals

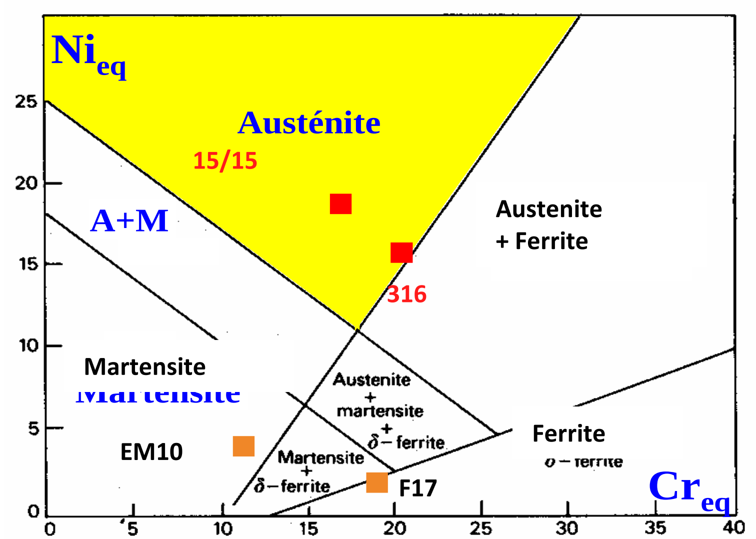
Crystalline structure = precise pattern of atoms following a unit cell that is periodically reproduced



Steels

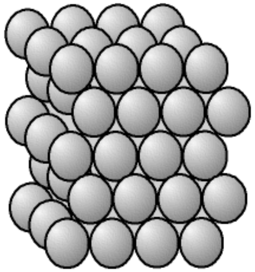
- Steels = Iron + Carbon + alloying major and minor elements (Cr, Ni, etc.) that participate to the global mechanical and chemical properties and to the radiation resistance

- Steels can have different crystalline structures depending on
  - the type and quantity of alloying elements
  - temperature
  - fabrication route (thermo-mechanical treatments)



1. Basics of crystalline structure

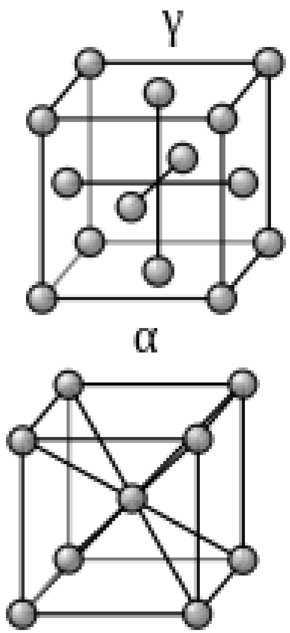
Structure of metals

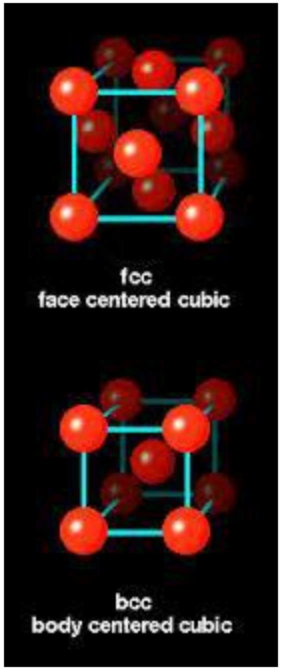


Crystalline structure = precise pattern of atoms following a unit cell that is periodically reproduced

Steels

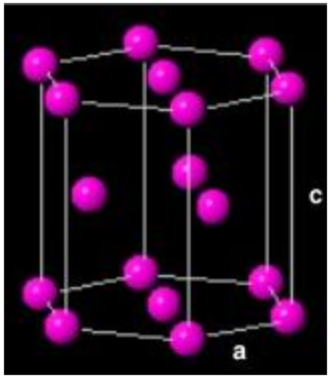
- Two important steel types:
  - Austenitic (gamma)
  - Ferritic and martensitic (alpha and alpha')





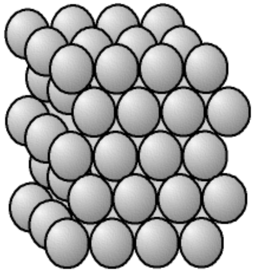
Zirconium

- Hexagonal close pack structure (hcp)



1. Basics of crystalline structure

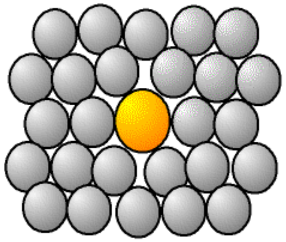
Structure of metals

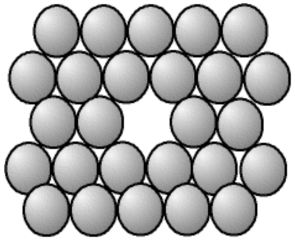


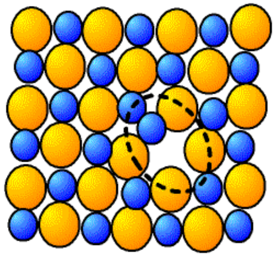
Crystalline structure = precise pattern of atoms following a unit cell that is periodically reproduced

This crystal is not perfect !

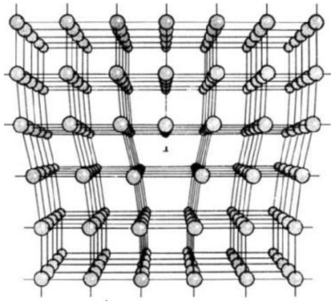
- Extra atoms (interstitials)
  or lack of atoms (vacancies)
  = point defects



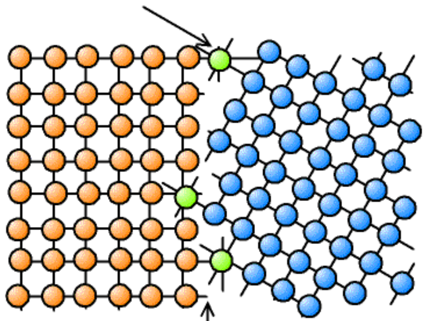




- Staking fault = dislocations

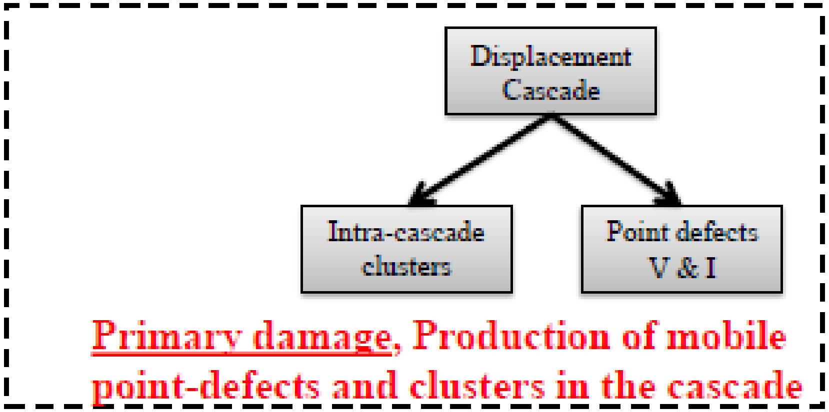


- grain boundaries, interfaces



Radiation effects in metals

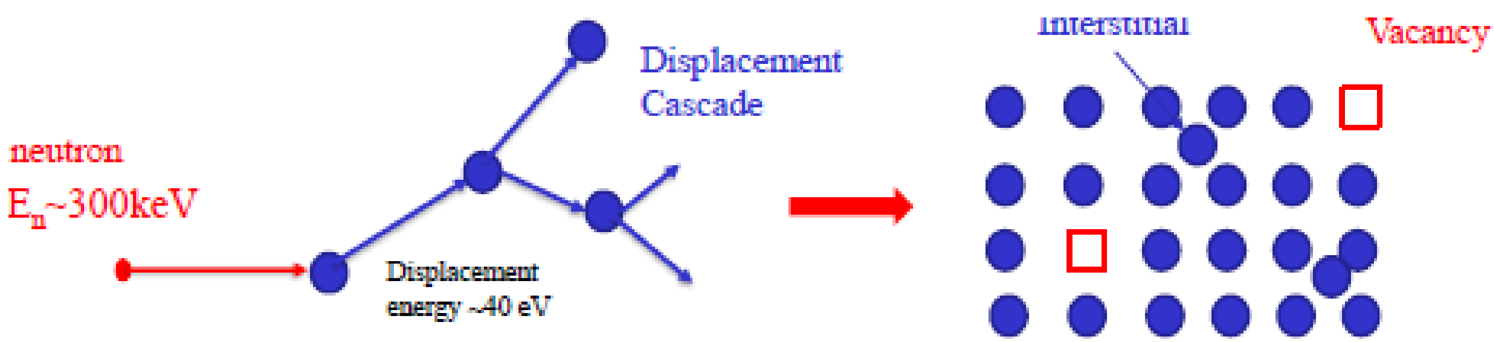
- Neutrons dissipate energy in the matter by colliding atoms



- Primary damage: atoms are expelled from their equilibrium site and collide other atoms
  - Atomic displacement cascade : interstitials + vacancies (Frenkel pair)

→ reorganization / atomic diffusion (thermally activate): some atoms go back to their initial site

→ others remains in the crystalline network as interstitials and vacancies



Radiation effects in metals

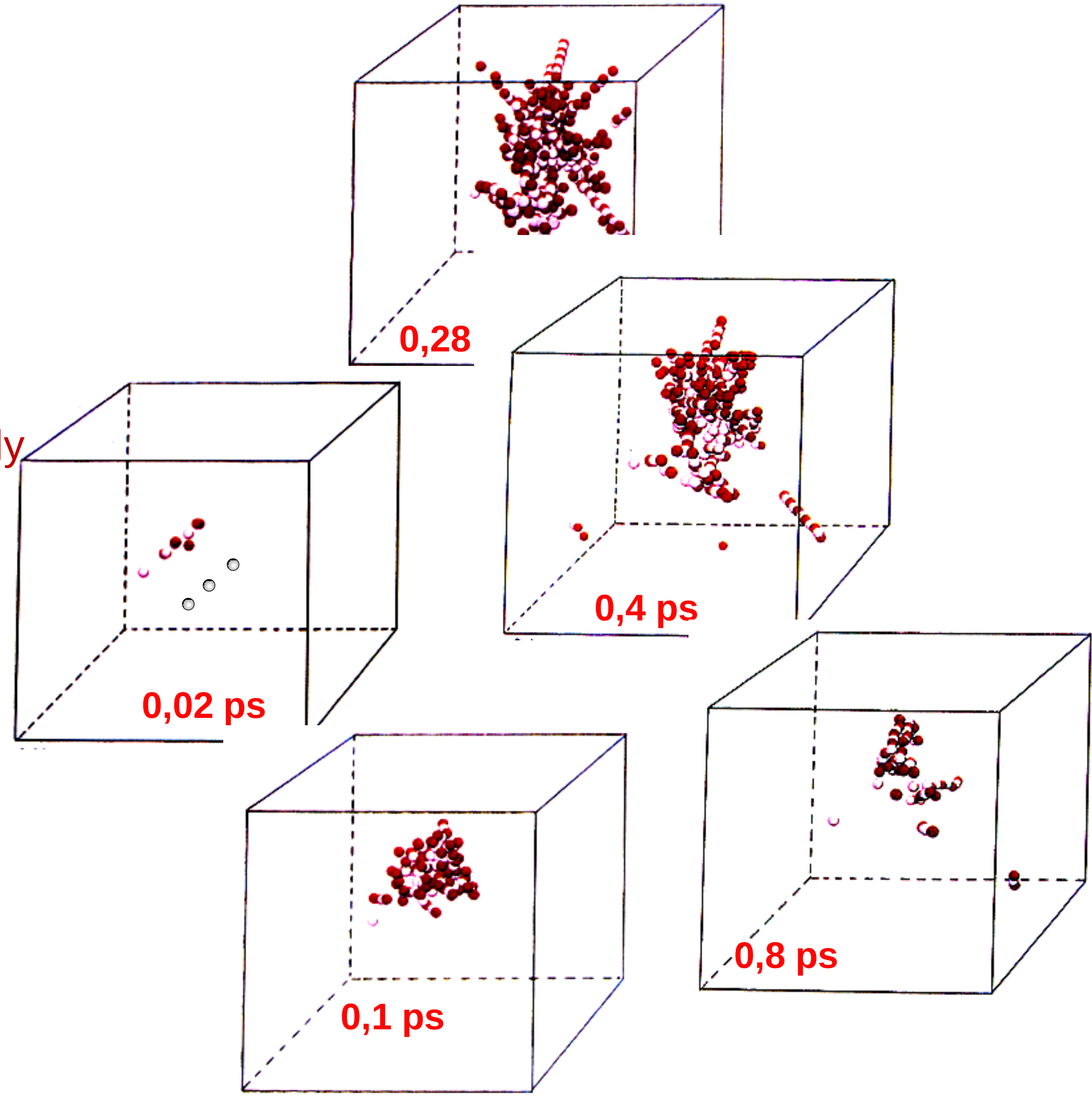
- Computer simulation of displacement cascades (DM)
  - Fast recombination

→ Few surviving defects in the crystalline structure:

- Point defects: interstitials and vacancies
- I and V clusters formed directly in the cascade

PKA

15 keV

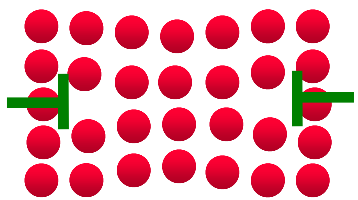


Material damage is quantified in displacement per atom = dpa

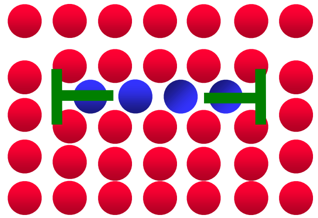
1. Basics of crystalline structure under irradiation

Radiation effects in metals

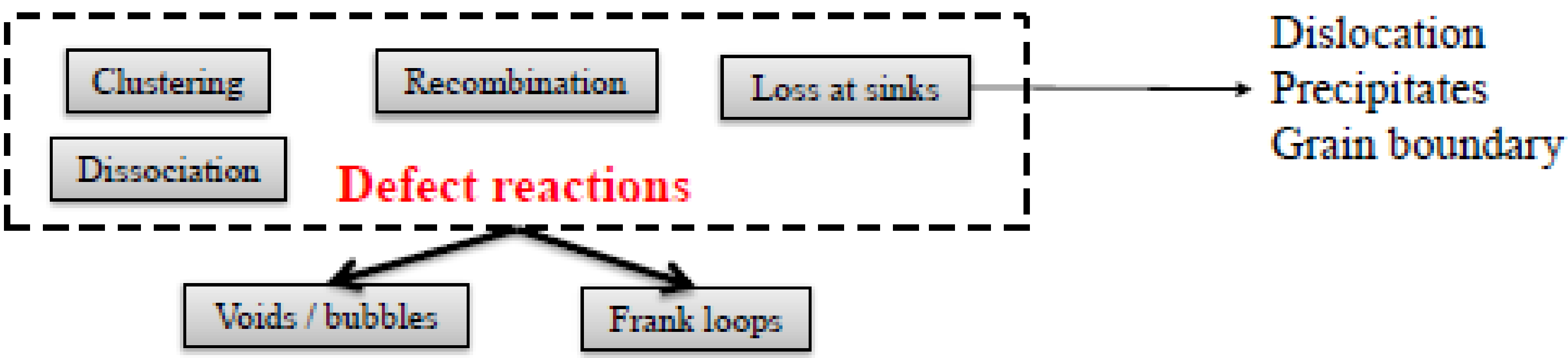
- Defects rapidly evolve with time. Depending on the dose, temperature, material characteristics… these defects
  - Recombine together
  - Annihilate along dislocations and grain boundaries that acts as sinks for defects
  - ➔ Driving force for interstitial annihilation at dislocation is (slightly) higher than for vacancies → bias
  - Group to form clusters



Vacancy clustering -> cavities



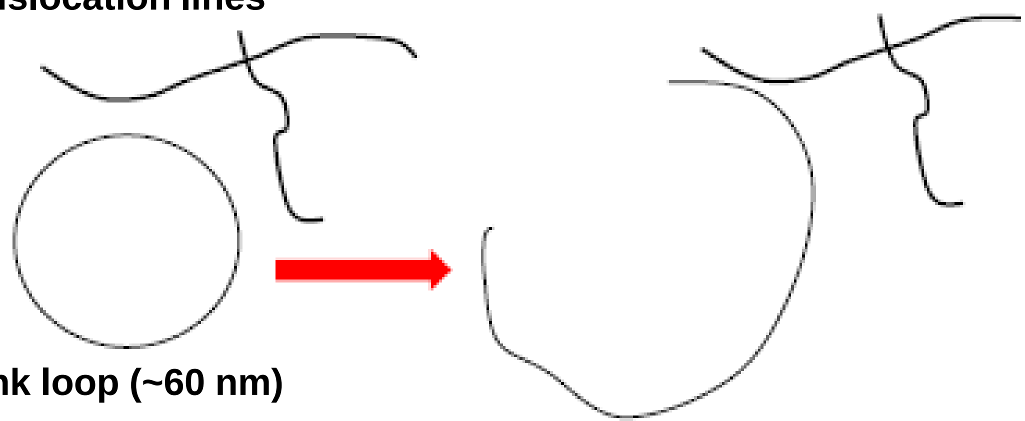
Interstícial clustering -> faulted loop (Frank)



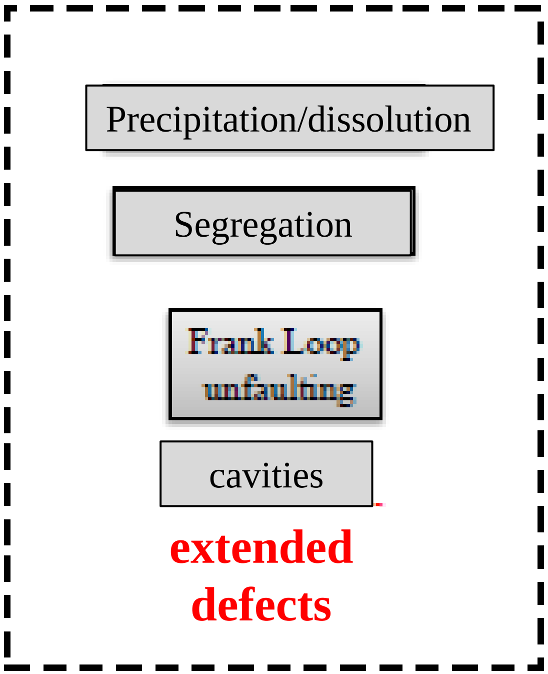
Radiation effects in metals

- Defects can migrate and interact with the microstructure and dislocation network
  - Vacancies can be attracted to cavities
  - Frank loops can unfault into other types of loops or as a dislocation line
  - Defects can drag solutes (coupling). This can accelerate or modify precipitation
- Extended defects: dislocations, cavities, precipitates → change in the microstructure
- with a direct impact on the material properties

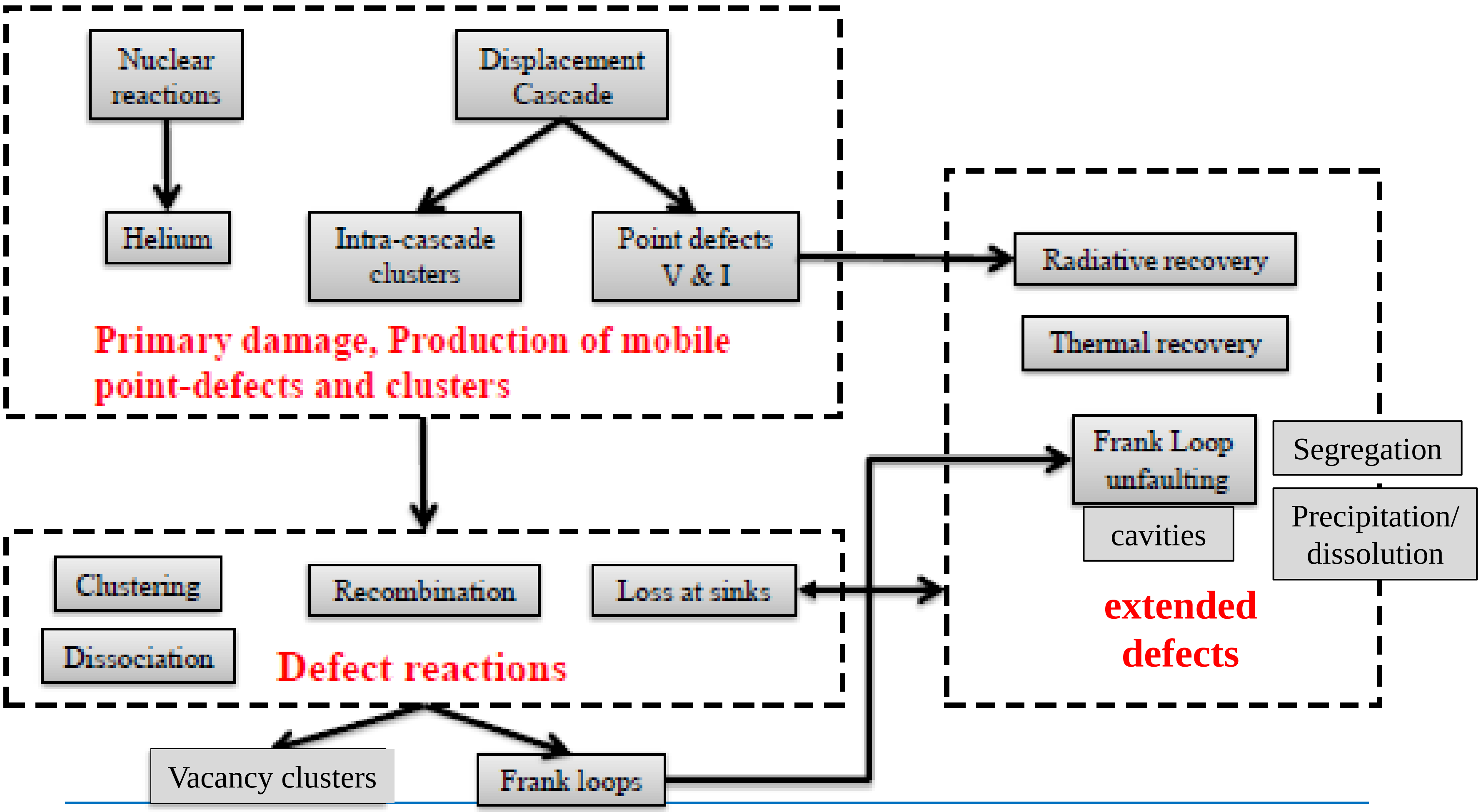
dislocation lines



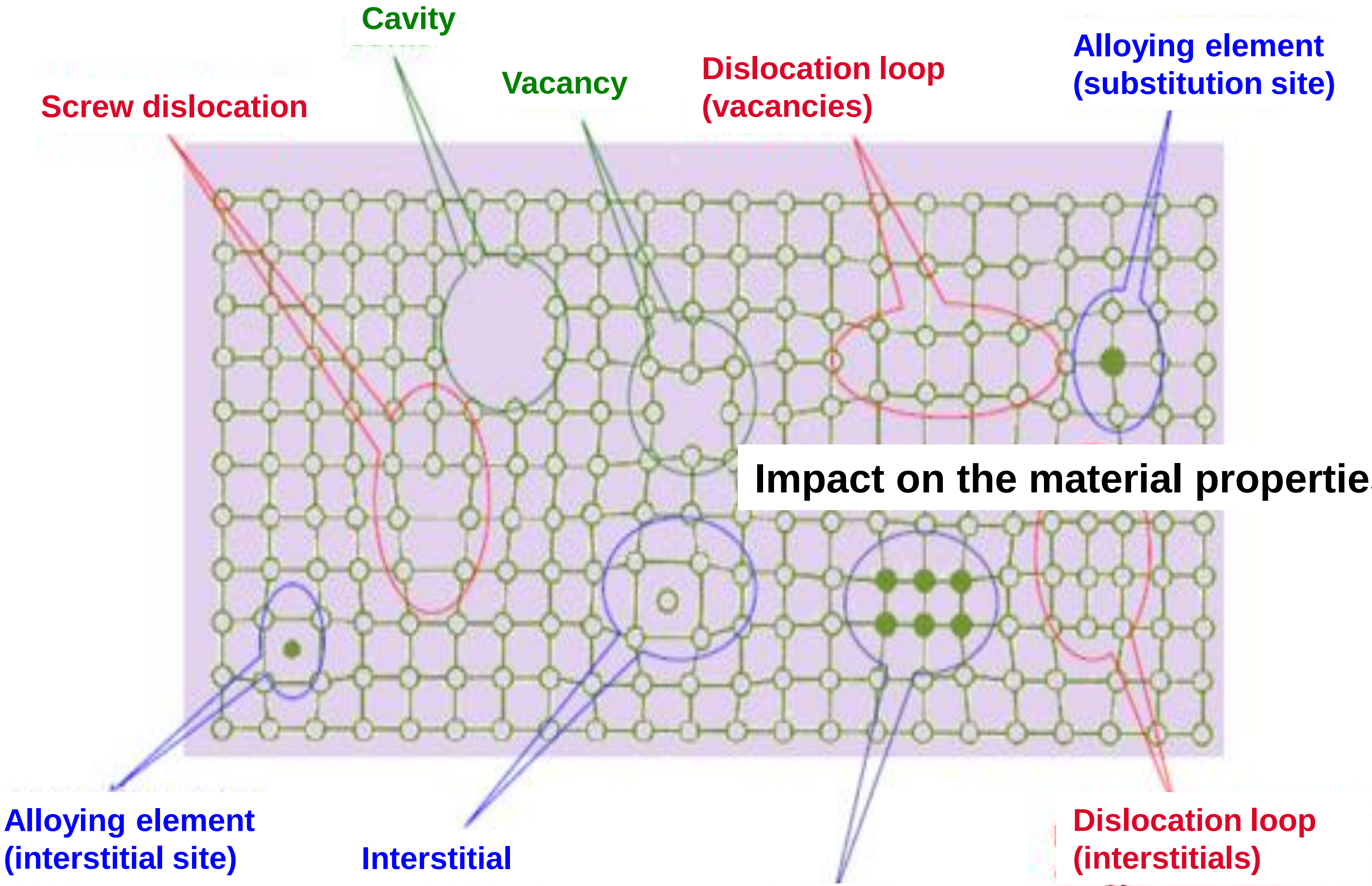
Frank loop (~60 nm)



Radiation effects in metals

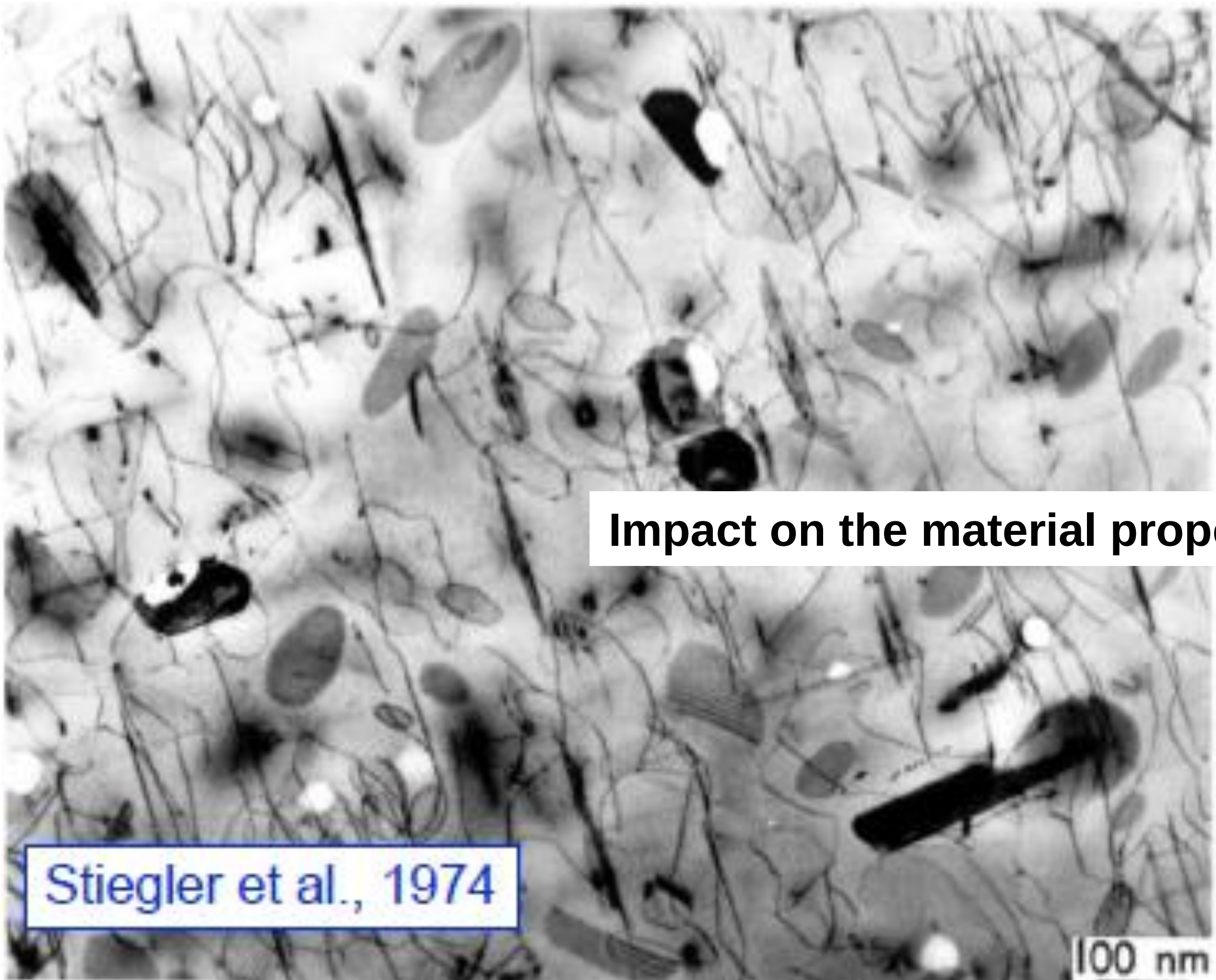


Radiation effects in metals



Impact on the material properties ?

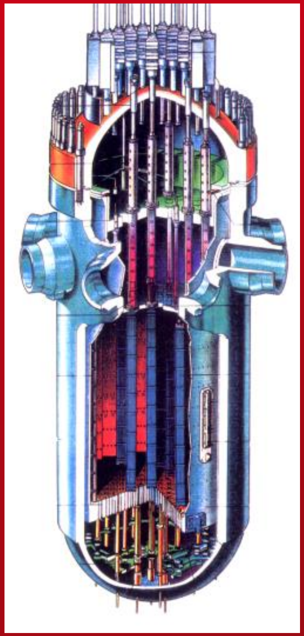
Radiation effects in metals



Impact on the material properties ?

SA 316 irradiated in EBR-II to 1.9×10^{26} n/m^2 at 580 °C

Radiation embrittlement LWR vessel steel

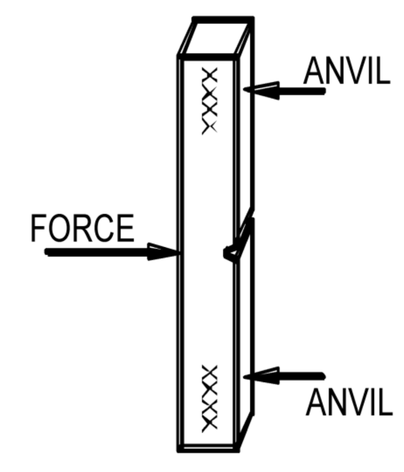


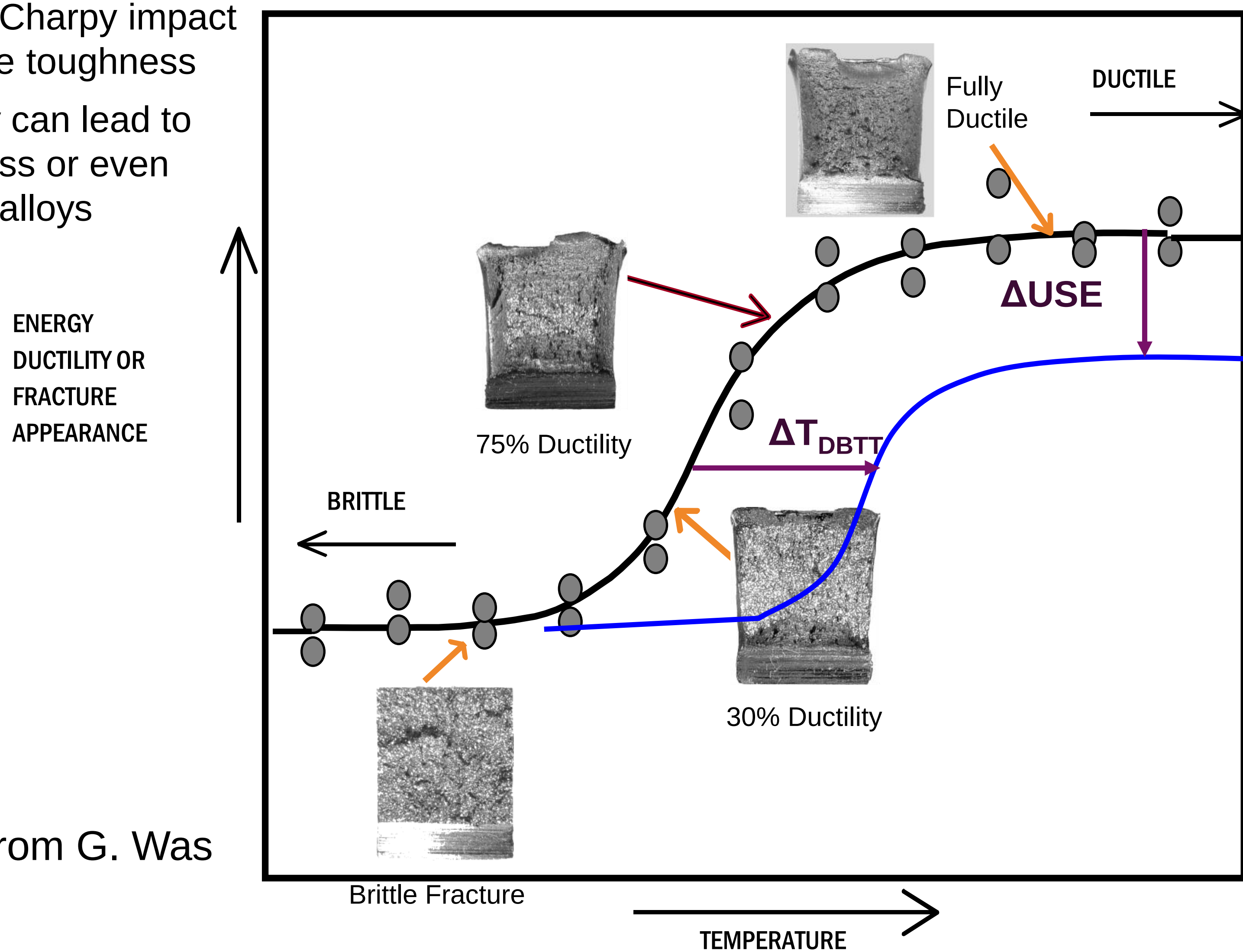
2. Radiation embrittlement – LWR vessel steel

Ductile-Brittle Transition Temperature

bcc bainitic steel with Mn, Ni, Mo…

- Measured with Charpy impact test and fracture toughness
- Loss of ductility can lead to loss of toughness or even failure in some alloys



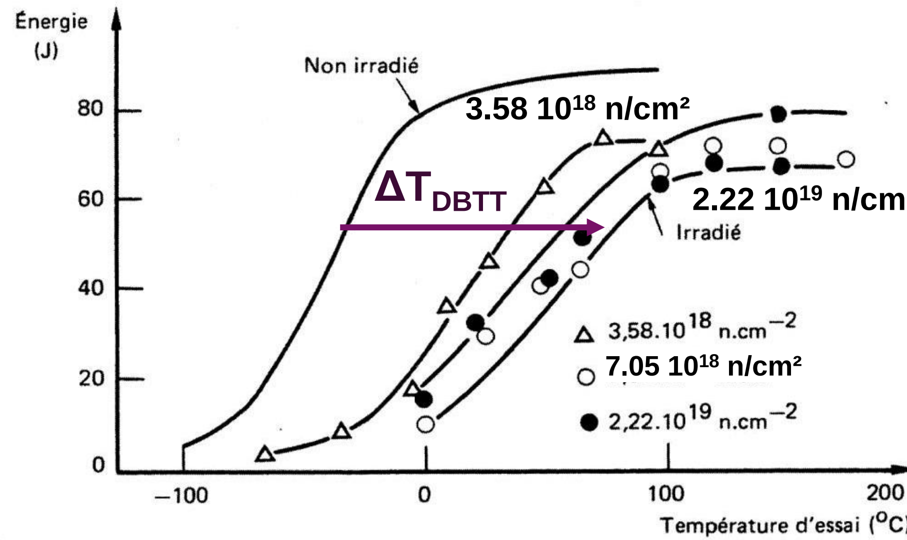


from G. Was

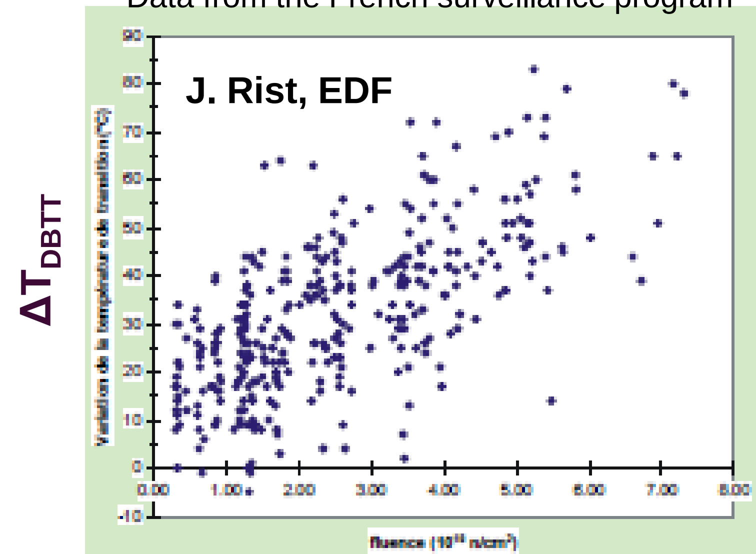
Embrittlement due to hardening

- DBTT of irradiated steel
  - Higher strength increases the probability of failure by cleavage, leading to higher transition temperature
  - DBTT increases with fluence
  - At high dose: occurrence of brittle Mn, S, Ni enriched phases (late blooming phases)
- The trend is not linear and saturates (?)

DBTT of vessel steel before and after irradiation, 290°C



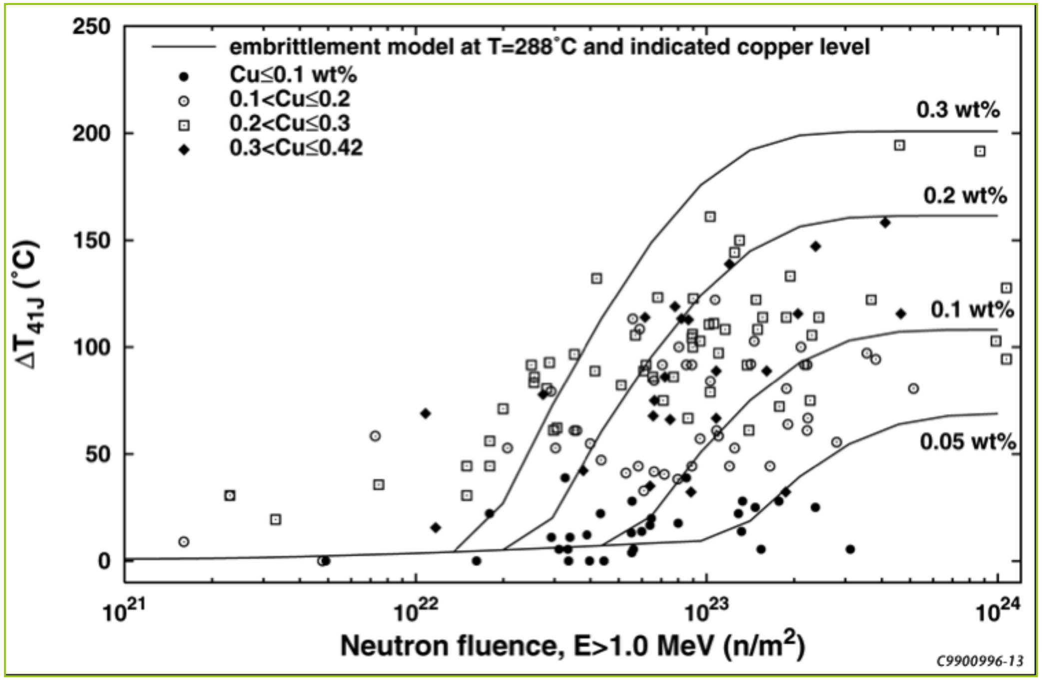
Data from the French surveillance program

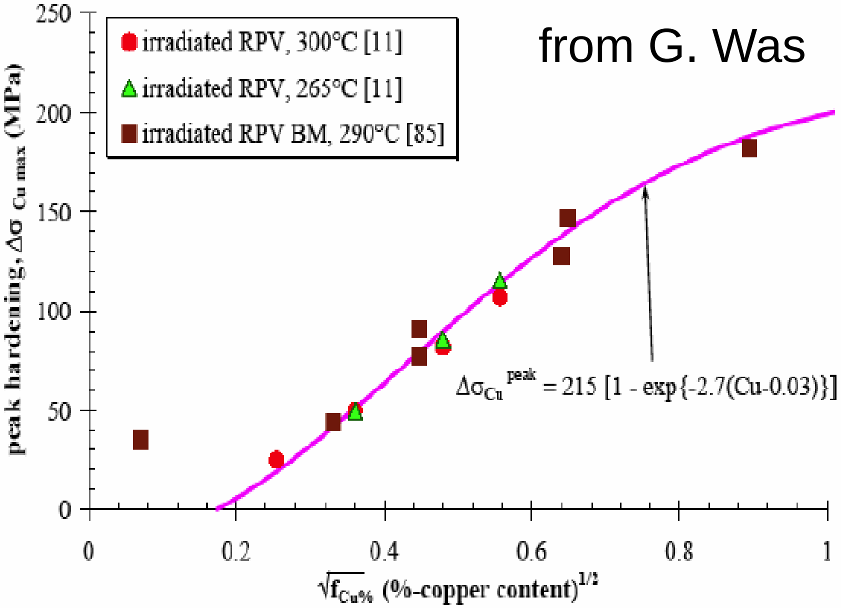


2. Radiation embrittlement – LWR vessel steel

Effect of steel purity on hardening and embrittlement

- Effect of chemical composition through a large body of analytical studies
  - P, S segregate at grain boundaries
  - Cu, Ni clusters inside the grains
- Cu content was shown to have a strong impact





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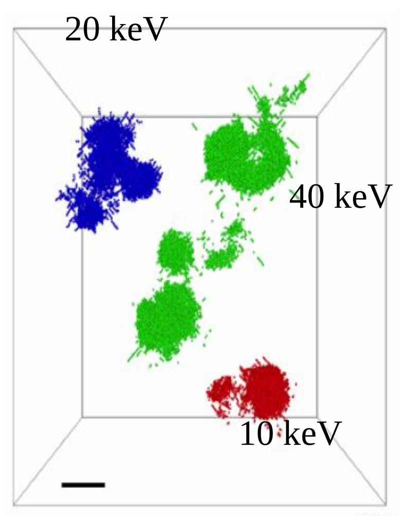
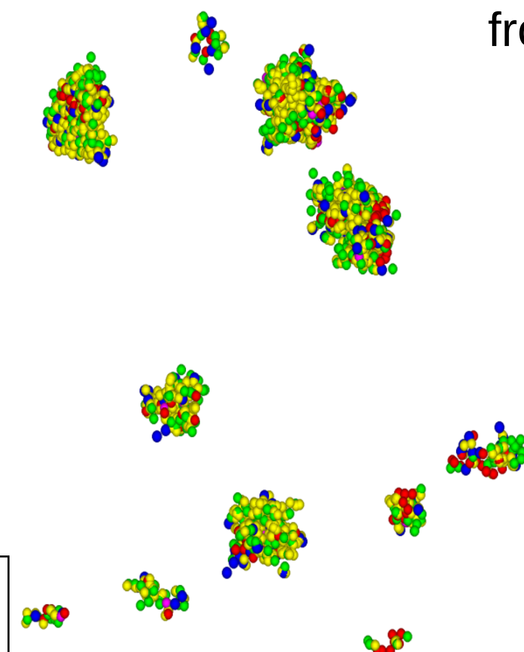
Microstructure origins of embrittlement

- Formation of nanoscale precipitates rich in Cu, Ni, Si, P, Mn
- Composition and size don't seem to change with dose
- Number increases with dose and Cu content



Neutron radiation produces an extremely high number density of nanoscale copper-, manganese-, nickel-, silicon-, and phosphorus-enriched precipitates.

from G. Was



Fe Cu Ni Mn Si P atoms

Combination of experimental, modeling, and microstructural studies leads to advances in predictive capability.

Recommended values for DBTT shift calculation

- Several empirical estimates have been developed to account for the shift in DBTT with dose and chemical composition

F: fast fluence

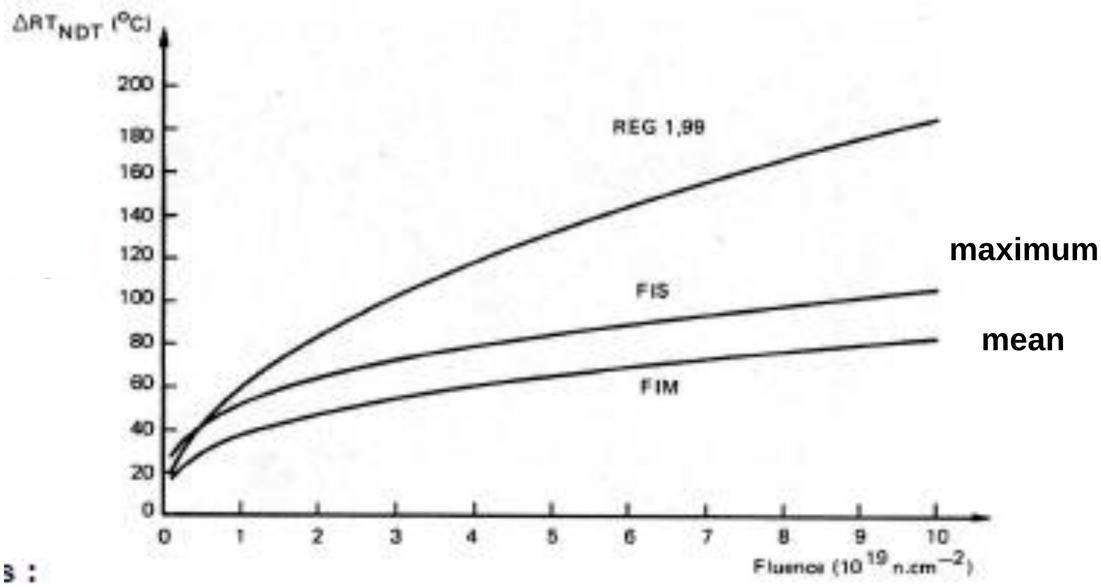
- NUREG (Nuclear Regulation Board, USA)

$$\Delta RT_{NDT} = [22 + 556\,(\%Cu - 0.08) + 2778\,(P - 0.008)]\left({}^{F}/_{10^{19}}\right)^{1/2}$$

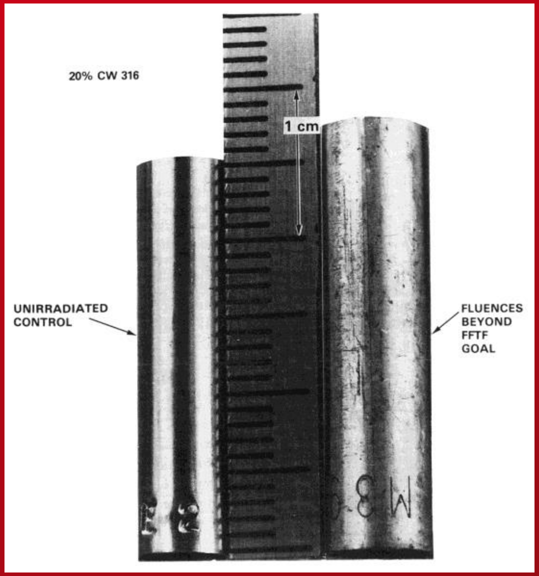
- EDF Framatome CEA

concentration in weigth %

$$\Delta RT_{NDT} = 8 + [24 + 238\,(\%Cu - 0.08) + 1537\,(P - 0.008) + 192(\%Ni^2\%Cu)]\left({}^{F}/_{10^{19}}\right)^{0.35}$$

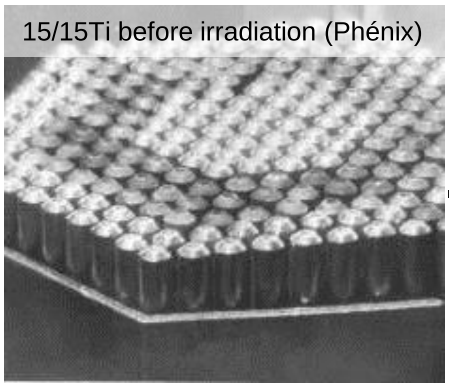


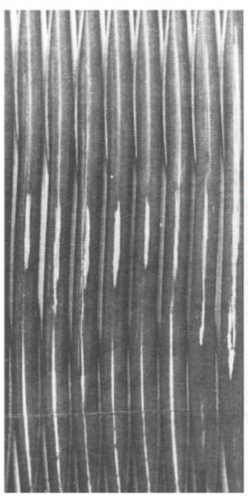
Swelling
SFR cladding



3. Swelling – SFR cladding tubes

Swelling is a critical consequence of irradiation for austenitic steels…





« twist » along the spacer wire

- Diameter increase
- Elongation
- Embrittlement at $\Delta V/V > 6\%$

… and leads to steel embrittlement

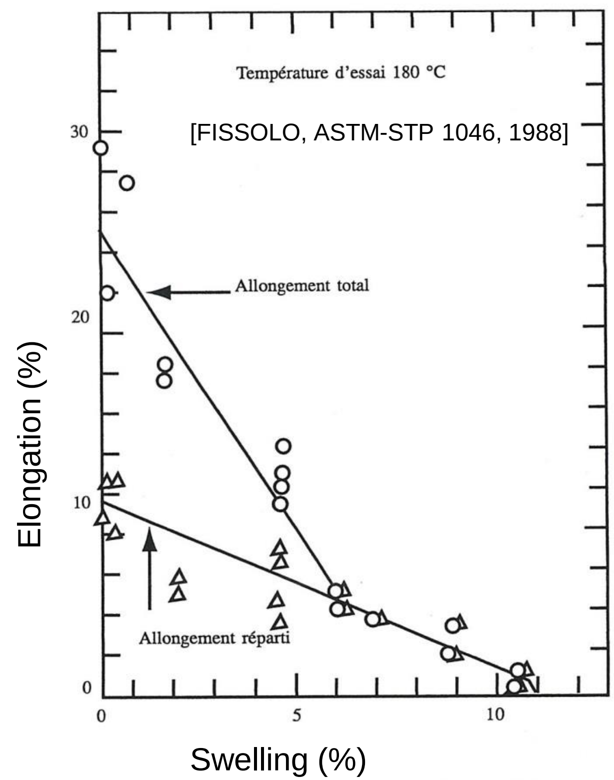


FIG. 4.56. – Influence du gonflement sur la ductilité d'un TH en 316 Tiε irradié dans PHÉNIX [103].

3. Swelling – SFR cladding tubes

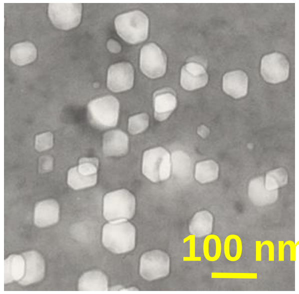
Swelling is principally due to a bias…

- Irradiation → Frenkel pairs are created = 1 interstitial + 1 vacancy
- Defects evolve
  - Formations of clusters, small loops
  - Recombination/annihilation of defects HOWEVER

  Preferential absorption of interstitials at sinks (dislocations)

  ➔ Vacancies are in supersaturation

  ➔ Nucleation and growth of cavities



304 SS
SFR 450°C

This mechanism was observed in the early SFR reactors

Main consequences of swelling :

- Changes in dimensions (elongation, loop deformation, arching/bending)
- Build-up of internal stresses due to inhomogeneous swelling (under dose gradient, temperature gradient) → creep is favored
- Increase in the fuel pellet/clad gap → local heating and promotion of the oxide/clad interaction (internal corrosion)
- Embrittlement due to porosity at high dose

→ swelling must be controlled in SFR cladding tube

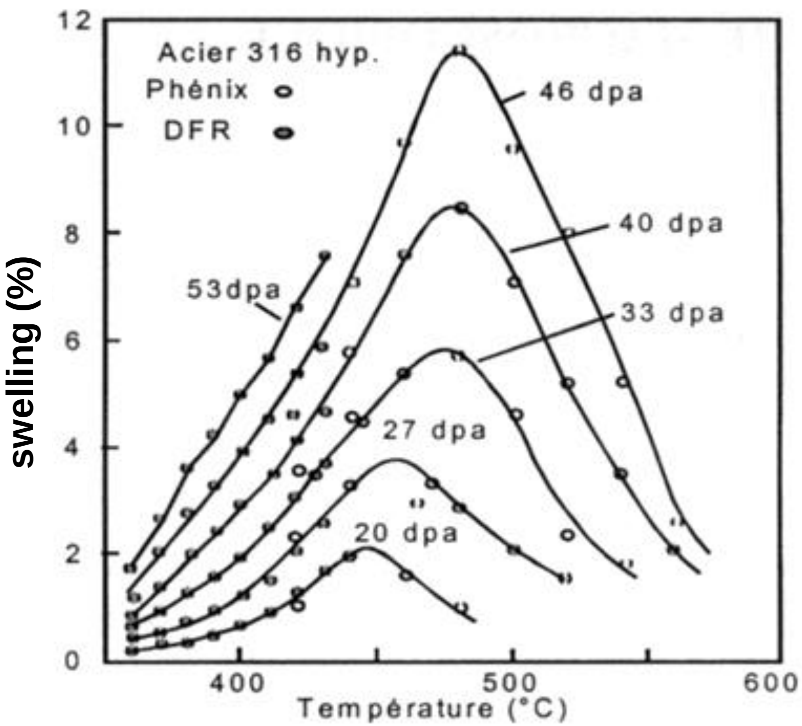
3. Swelling – SFR cladding tubes

Swelling is not linear in a reactor…

- Microstructure evolution (point defects, diffusion, precipitation…) depend on irradiation dose (dpa) and temperature

Swelling follows a bell curve
S = vacancy density × vacancy volume

T< 0,3 T fusion
Low atomic mobility, vacancy supersaturation but cavity growth rate remains low
→ Moderate swelling.



T> 0,5 T fusion
High atomic mobility, high cavity growth rate but no nucleation of cavities
→ Moderate swelling.

Cladding tubes in SA 316 from Phénix and DFR
swelling as a function of temperature

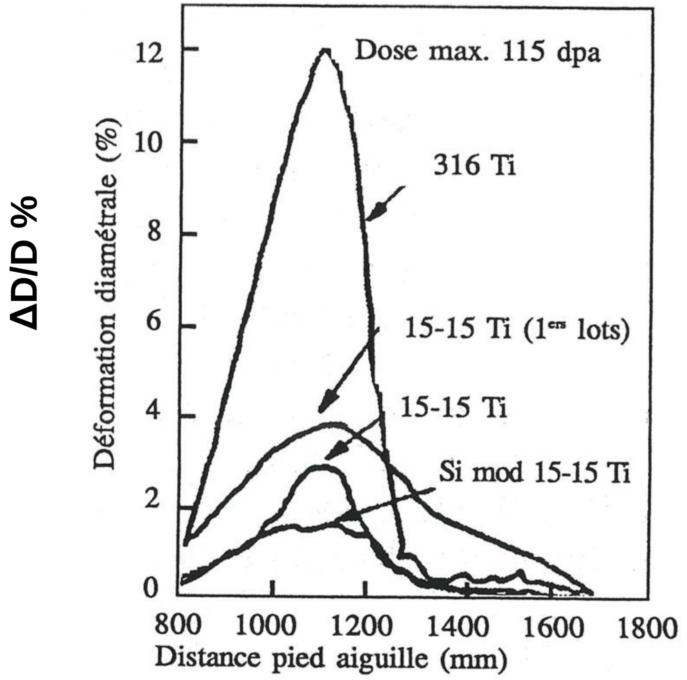
3. Swelling – SFR cladding tubes

Swelling is not linear in a reactor…

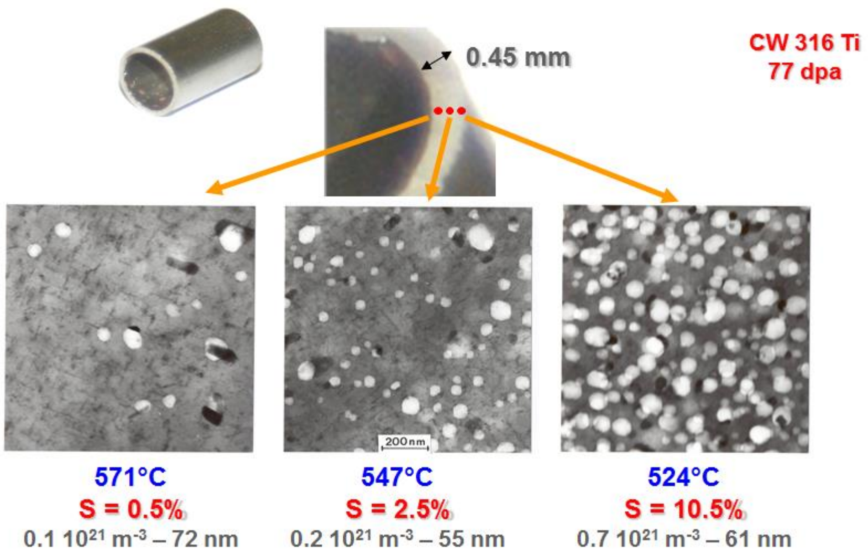
- Microstructure evolution (point defects, diffusion, precipitation…) depends on irradiation dose (dpa) and temperature
- Gradients of temperature and flux in the core induce microstructure and property gradients

Deformation gradient along a clad tube

[MAILLARD, ASTM-STP 1175, 1994]



Deformation gradient through tube wall

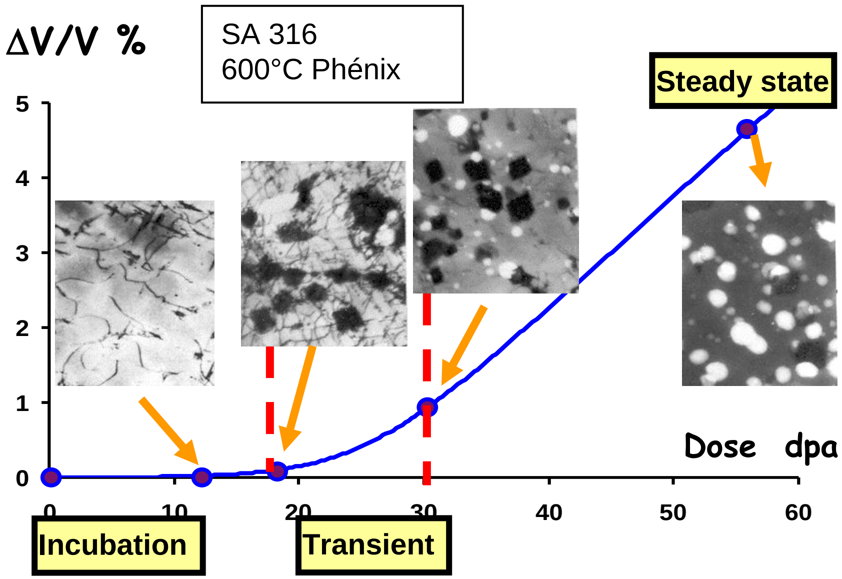


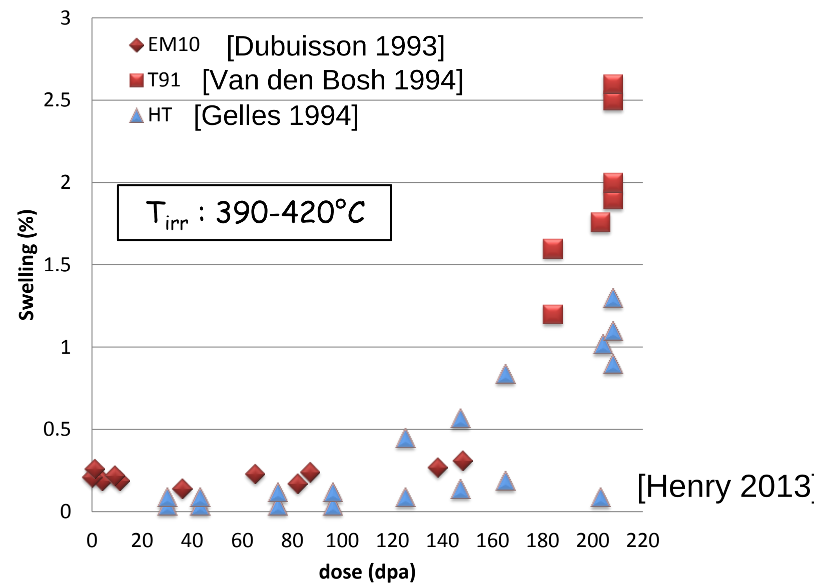
3. Swelling – SFR cladding tubes

Note: irradiation induces other changes in microstructure (precipitation) and behavior

Swelling shows an incubation time

- Steel swelling is a phenomenon with a threshold, which offers an operating widow below a given irradiation dose





- The operating window/maximum dose depend on the steel (structure, chemistry, metallurgical state).
- Swelling is a key issue for austenitic steels but also appears in ferritic steels at high doses

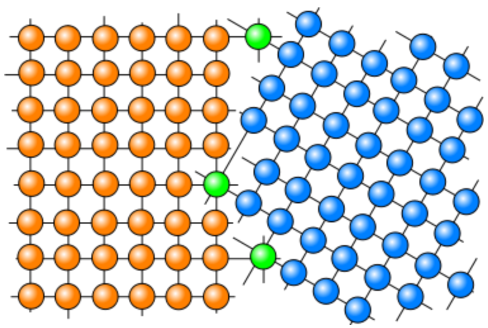
➔ R&D for the last 40 years has allowed to push the threshold further to higher dose (improving defect recombination, increase sink density)

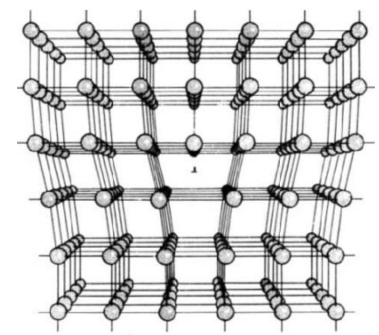
3. Swelling – advanced austenitic steels for SFR

How can we improve swelling resistance?

- In helping recombination between defects and defects annihilation

  Any « fault » in the microstructure is a potential sink for irradiation defects
  - Interface with precipitates
  - Grain boundaries
  - Dislocations (cold work)
  - …





- In adding swelling inhibitors in solid solution : C, Si, Ti, P, N which can surround the dislocations and thus increase sink efficiency for vacancies

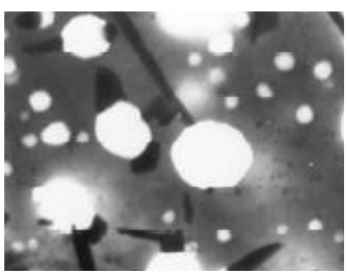
3. Swelling – advanced austenitic steels for SFR

Austenitic steels – improving metallurgy and chemistry

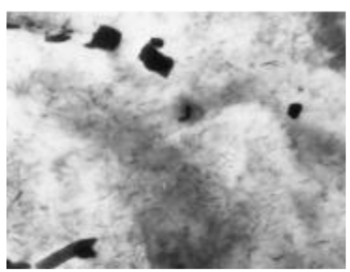
40 years of R&D

Solution annealed 316 (< 50 dpa)	• 1^{st} reference for Rapsodie-1968 and Phénix-1973 • Poor resistance to swelling (~50dpa)
Cold work 316	• Cladding tube and HT in Rapsodie et Phénix → 1980 • Beneficial effect of cold work (dislocation = trap) • Dislocation network instability at T >550°C → swelling
Cold work 316Ti 17%Cr 14%Ni (< 90 dpa)	• cladding and hex tubes • PX 1976→86, SPX 1986→98 • Increase of the incubation dose

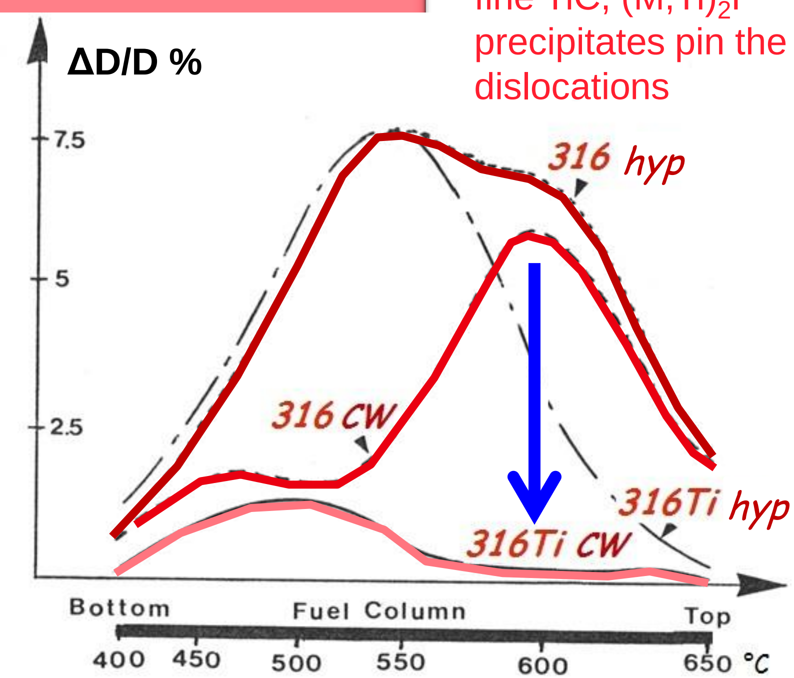
fine TiC, $(M,Ti)_2P$ precipitates pin the dislocations



SA 316 ΔV/V=10%



316Ti CW ΔV/V=0%



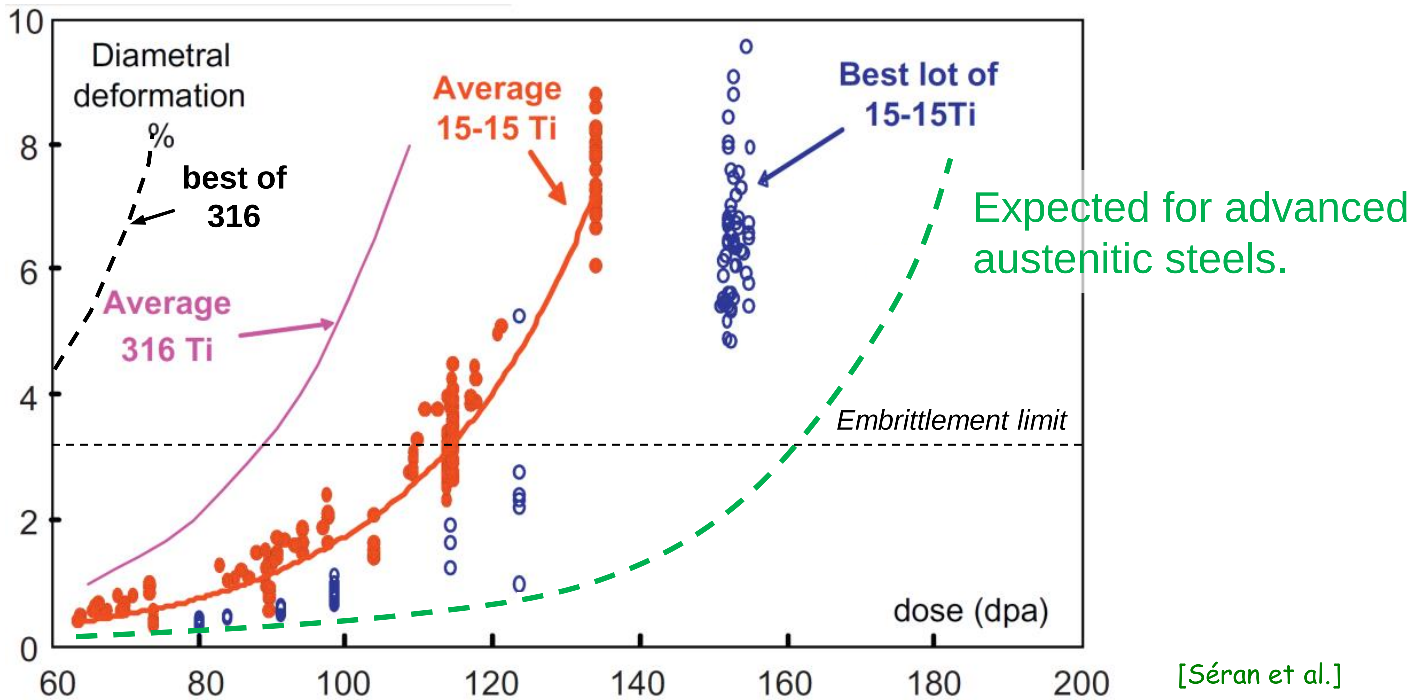
3. Swelling – advanced austenitic steels for SFR

Austenitic steels – improving metallurgy and chemistry

40 years of R&D

Dose	Steel	Features
< 90 dpa	CW 316Ti 17%Cr 14%Ni	• 2nd reference for cladding and hex tubes • PX 1976→86, SPX 1986→98 • Increase of the incubation dose by Ti addition
< 115 dpa	CW 15/15-Ti 15%Cr 15%Ni	• 3rd reference for PX cladding tubes (1982) • Beneficial effect of Cr/Ni = 1
< 130 dpa	Best of	• Minor alloying element optimisation (P, Si…) → French specification AIM1
<< 130 dpa	Very Best of ?	• Further optimisation toward AIM2 ?

3. Swelling – advanced austenitic steels for SFR

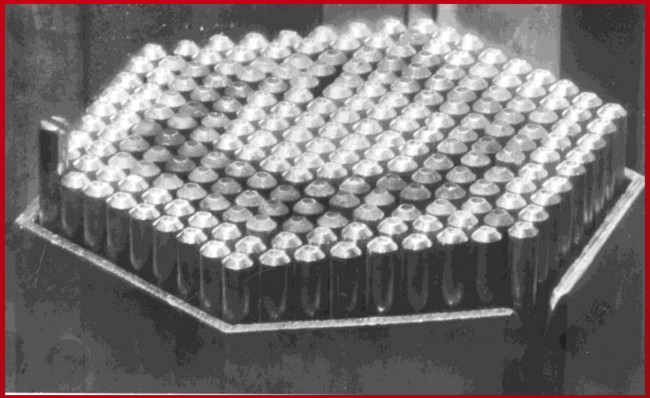


[Séran et al.]

Main features of swelling (austenitic steels):

- Swelling is due to vacancy supersaturation
- Cavities are formed that embrittle the steel
- Swelling varies with dose, dose rate, temperature and steel
- Swelling resistance can be greatly improved by tuning the chemical composition and thermomechanical treatments

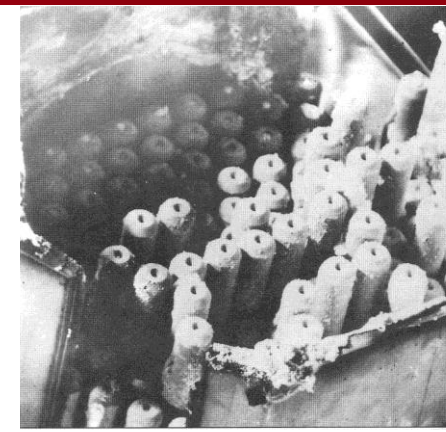
Irradiation creep
SFR cladding

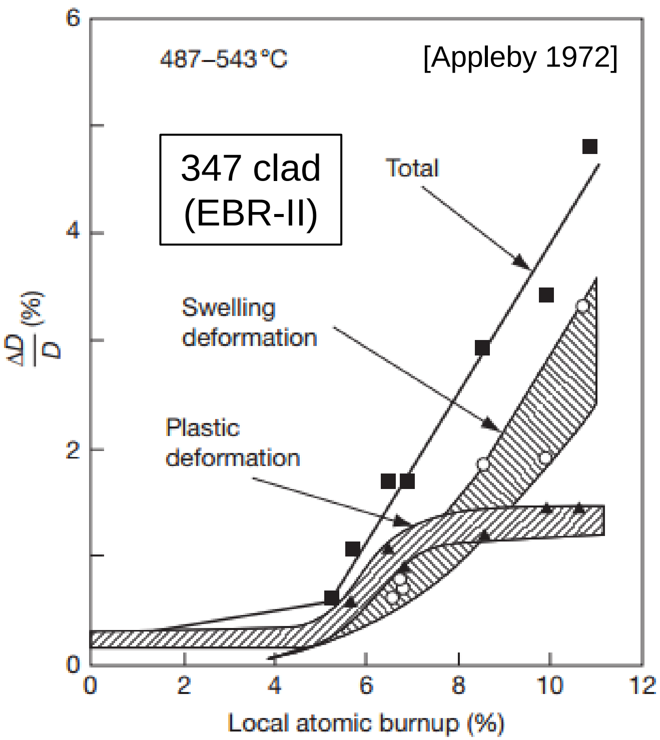


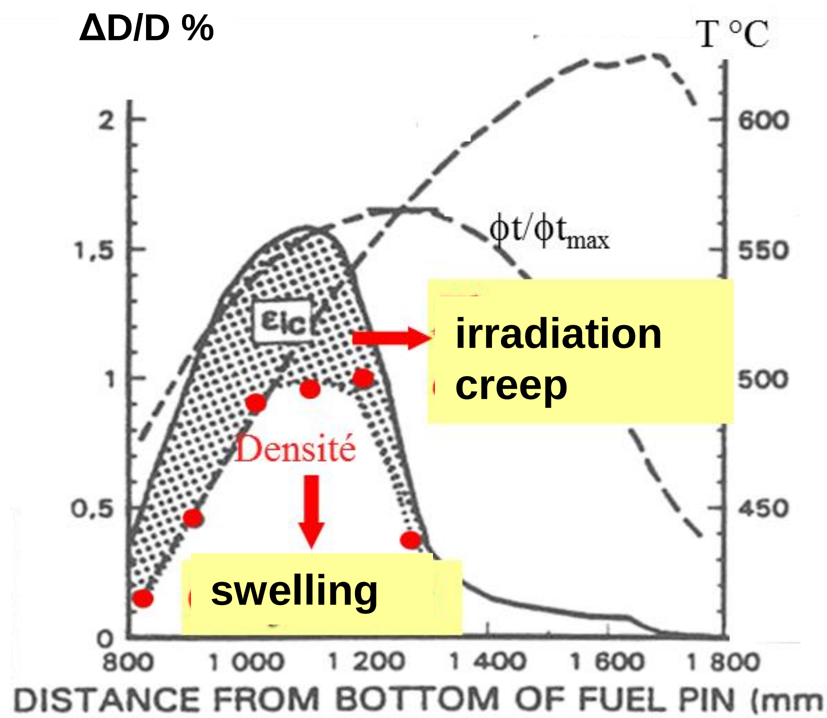
4. Irradiation creep – SFR cladding tubes

Tube deformation results from several contributions

- Swelling (= cavities)
- Thermal creep: plastic deformation under load from fission product pressure or fuel contact
- Irradiation creep: plastic deformation under load increased by irradiation



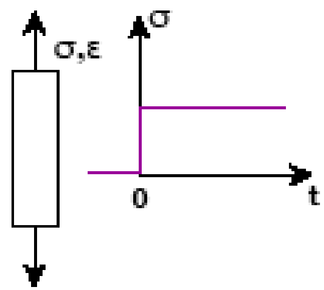


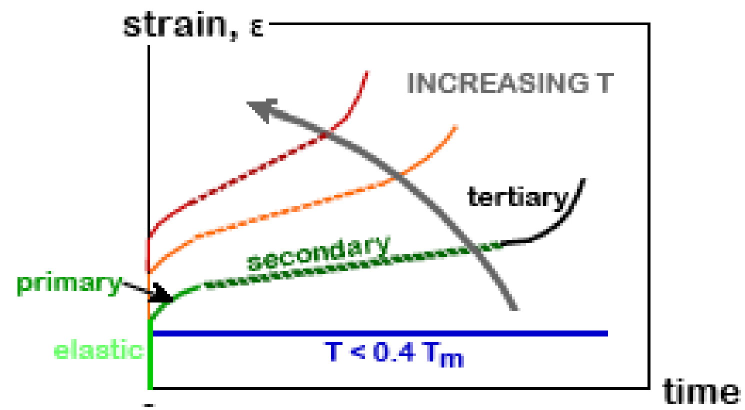


4. Irradiation creep – SFR cladding tubes

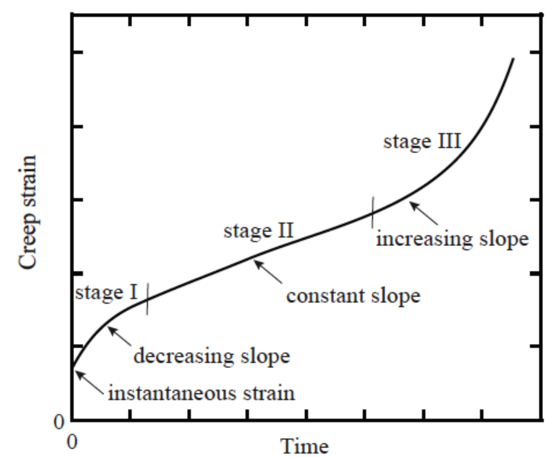
Creep in a nutshell

- Time-dependent plastic deformation of an alloy under constant load and at high temperature





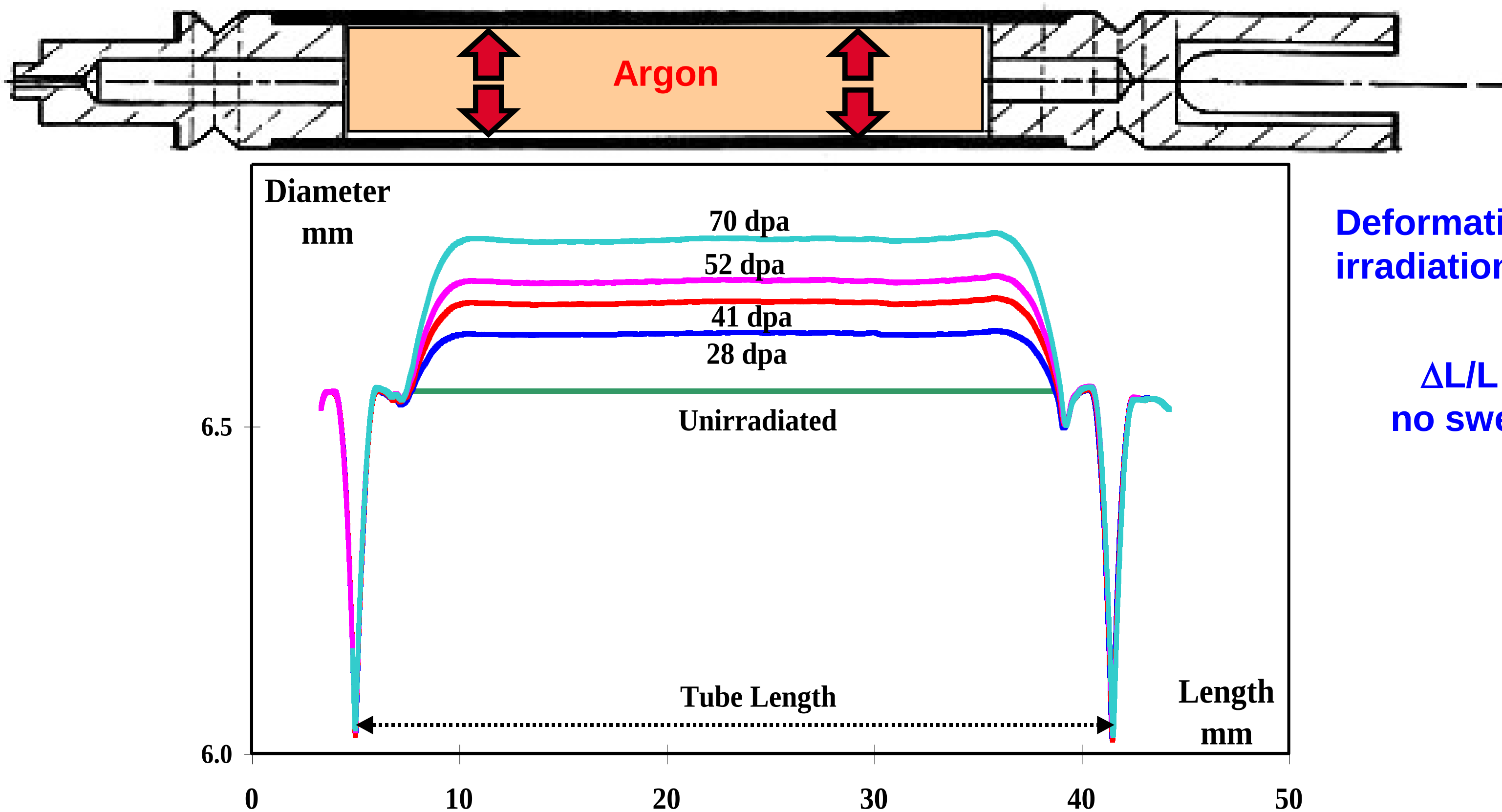
- Creep curve (ideally) shows 3 stages: transient / steady state / tertiary
- Temperature provides the energy for deformation through the creation and diffusion of defects
- In the steady state stage, creep can occur through different mechanisms depending on stress and temperature (Ashby maps) like diffusion (at boundaries or in the grain) and dislocation glide or climb



4. Irradiation creep – SFR cladding tubes

Irradiation creep is athermal

- Occurs under load and irradiation (+ T)
- Radiation produced point defects increase diffusion and allow creep at lower temperature → radiation induced creep or at a higher rate → radiation enhanced creep



Deformation only under irradiation

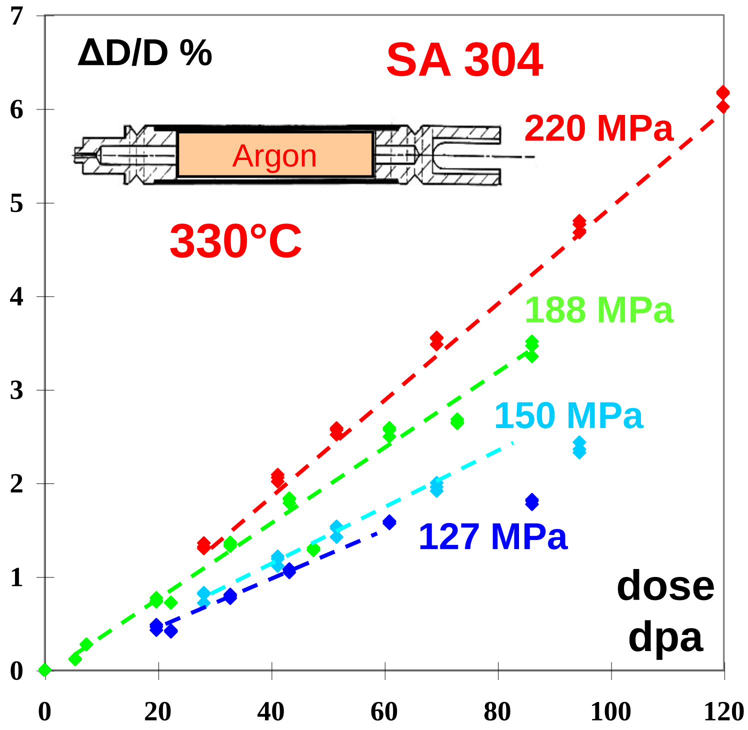
ΔL/L = 0
no swelling

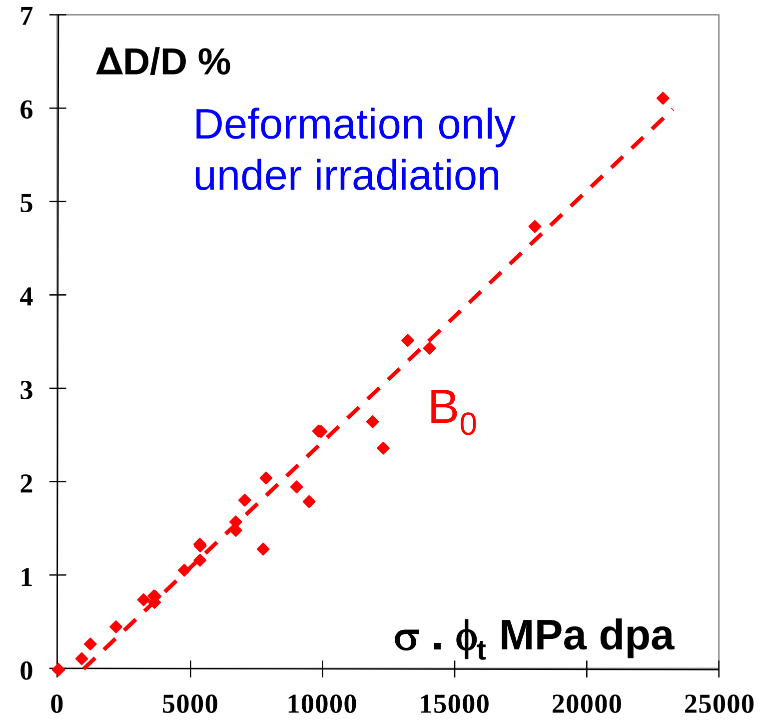
Irradiation creep is athermal

- Linear deformation versus dose (dpa) and versus load (MPa)
- In steady state domain, irradiation creep deformation

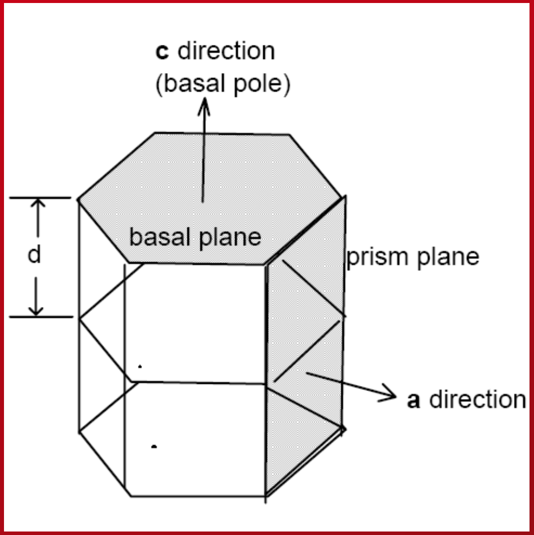
$$\varepsilon = B_0 (\sigma^n \phi t - B_1)$$

with B_0 athermal coefficient





Radiation growth
LWR cladding tubes



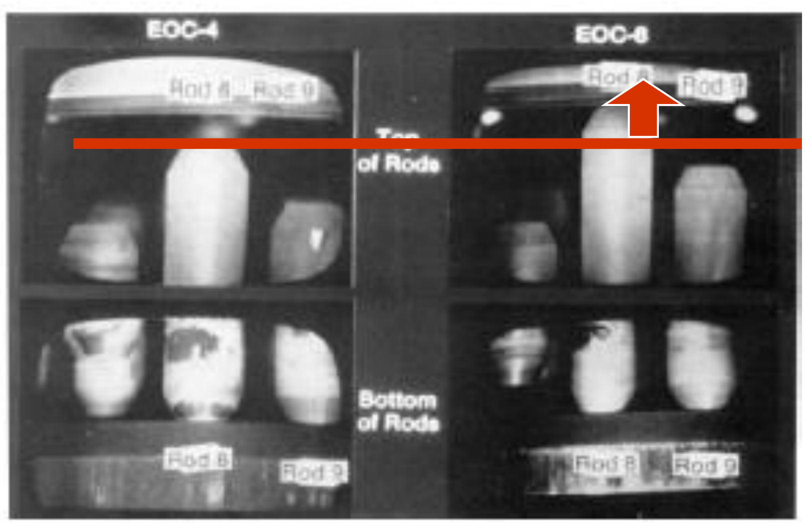
Growth of Zr4-cladding under irradiation

- Deformation free of stress
- Elongation of the cladding can reach several cm
- Growth depends on temperature and neutron flux
- Similar rods have different behavior (see picture). During cycle 5, rod #8 contacted both assembly end-plates. It was somewhat twisted which may affect the thermal hydraulic conditions of the rod.

➢ Margin is taken for free elongation of the PWR cladding

- Other components are concerned : grid in PWR and channel box in BWR whose growth can cause difficulty in fuel handling

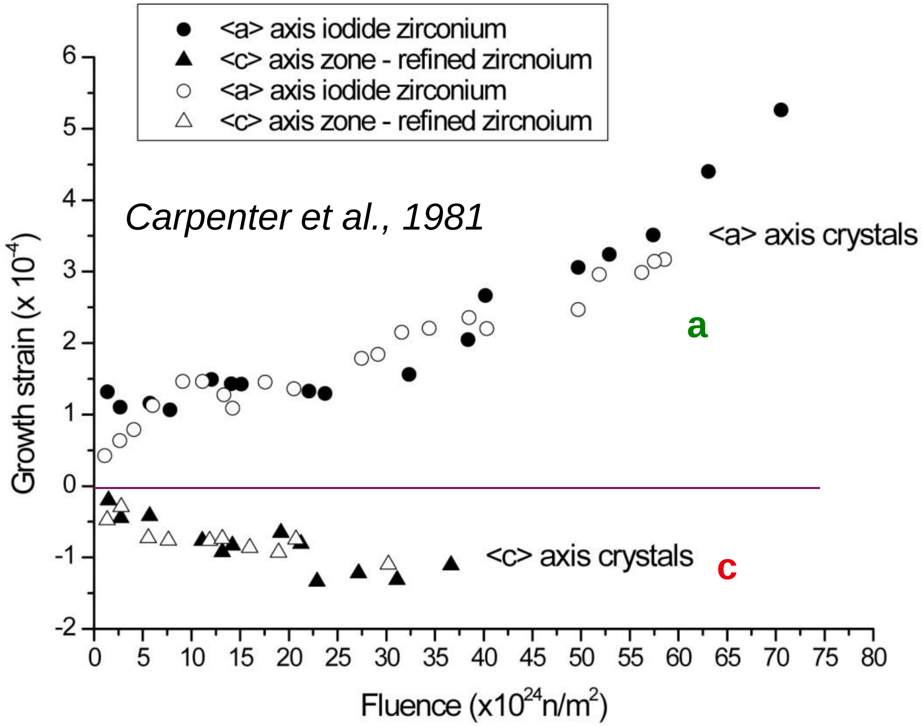
Cycle 4 **Cycle 5**

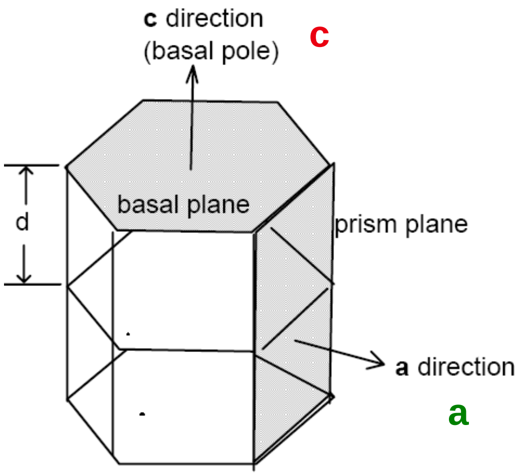


5. Irradiation growth – LWR cladding

Growth of zirconium under irradiation

- Specific to textured materials with anisotropic behavior like Zr or U
- Elongation in the <a> direction and shortening in the <c> directions at constant volume
- Acceleration of the growth at high doses (related to <c> loop formation)



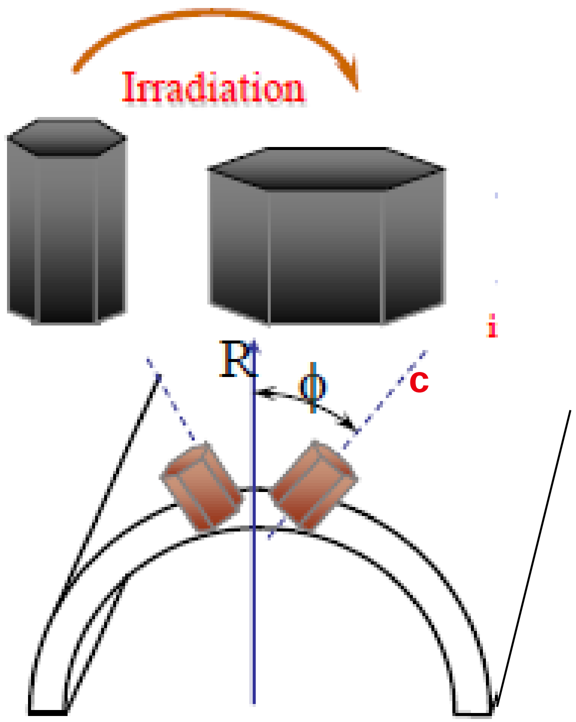


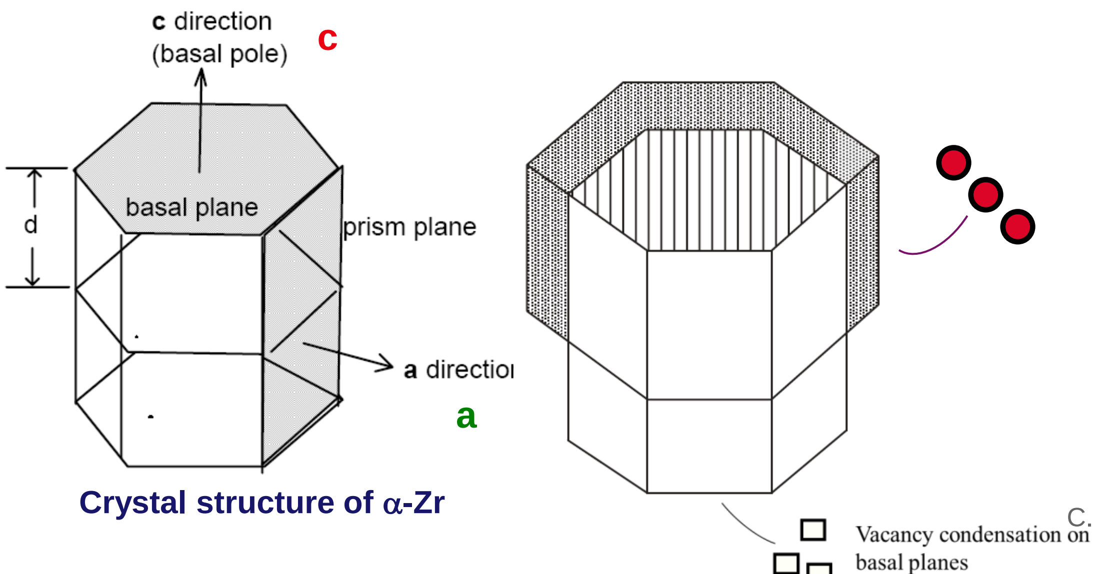
Irradiation Growth Strain at 553K in Annealed Iodide and Zone-refined Zirconium Single Crystals

Mechanism for growth under irradiation

- Cladding tubes have a strong texture with basal pole <c> along the radial direction (R) and <a> direction along the z-axis

  ➔ Growth in the <a> direction with tube elongation
- Interstitial preferential migration parallel to basal planes and condensation on prism planes
- Vacancy preferential condensation on basal planes



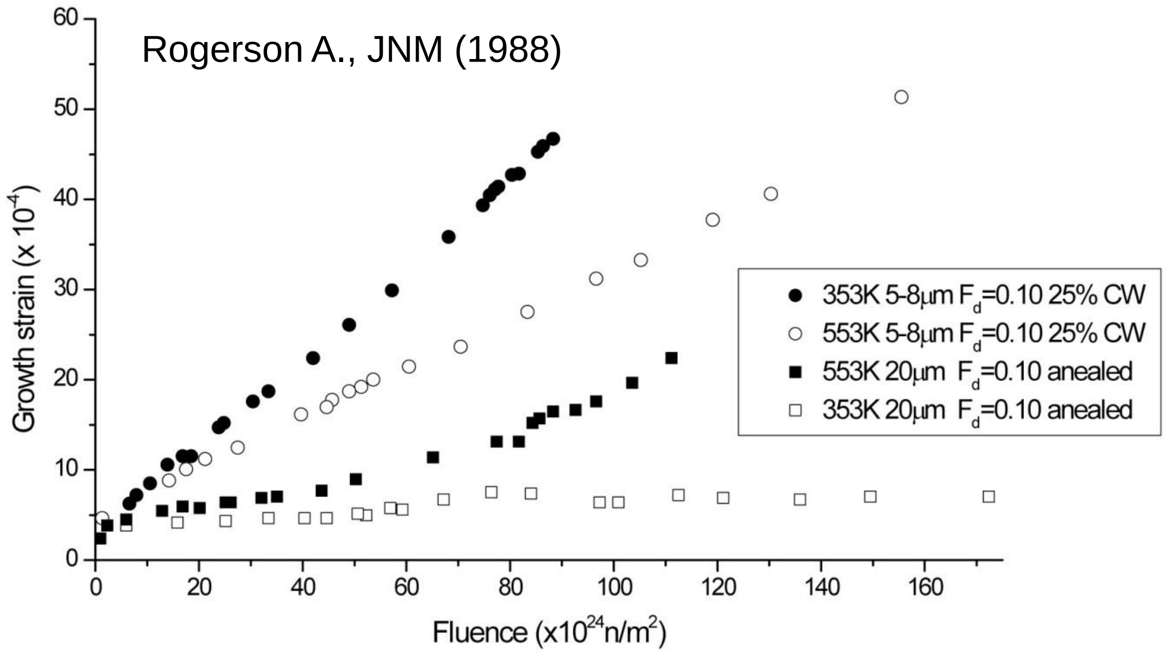


Crystal structure of α-Zr

5. Irradiation growth – LWR cladding

Limitation of irradiation growth

- Increasing isotropy helps in limiting irradiation growth of Zr-alloys
- In the annealed state, unit cells of Zr-alloy are more randomly distributed. Quench also improves the behavior by keeping the high-temperature bcc structure
- Cold work which favors recrystallization with a high texture appears to be negative

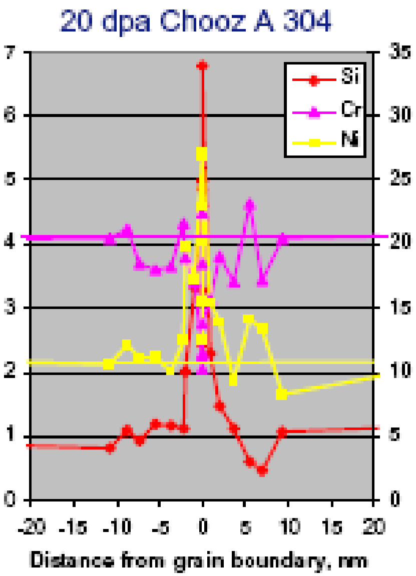


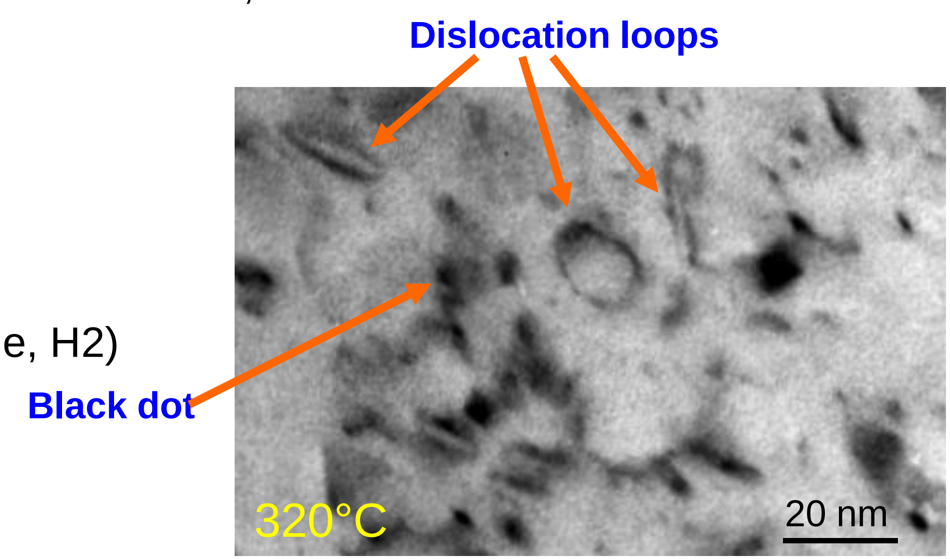
Irradiation Growth Strain at 353 and 553 K in Annealed and 25% Cold-worked Zircaloy-2

Radiation damage in Materials conclusion

Conclusions

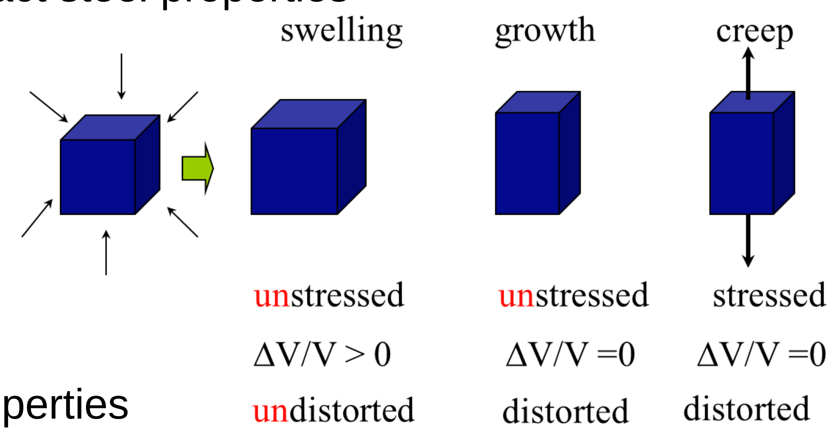
- Irradiation produces point defects in steels. Some interstitials and vacancies evolve in clusters and extended defects like Frank loops, cavities. These features interact with the steel microstructure and change its properties.
- Different mechanisms are induced or enhanced under irradiation depending on environment (temperature, dose/fluence, dose rate, spectrum, load, fluid chemistry…) and materials (composition, thermomechanical treatments, history…)
  - Chemical changes
    - Radiation enhanced diffusion
    - Segregation at surface, grain boundaries, dislocations, interfaces…
    - Precipitation / dissolution
  - Interstitial clustering
    - Dislocation loops
    - Cavities, voids
    - Bubbles (pressurized with He, H2)
  - Change in dislocation network





Conclusions

- Irradiation produces point defects in steels. Some interstitials and vacancies evolve in clusters and extended defects like Frank loops, cavities. These features interact with the steel microstructure and change its properties.
- Changes in microstructure impact steel properties
  - ✓ Change of dimension
    - Swelling
    - Growth
    - Irradiation creep



from G. Was

  - ✓ Change in mechanical properties
    - Hardening
    - Embrittlement due to hardening, induced by segregation, due to cavities…
  - ✓ Specific interactions with environment
    - Irradiation assisted stress corrosion cracking

In fact, all these aging processes under irradiation may be correlated !

- Modelling coupled to fine characterizations and well controlled experiments is a powerful tool to go further into the understanding of this complex metallurgy

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