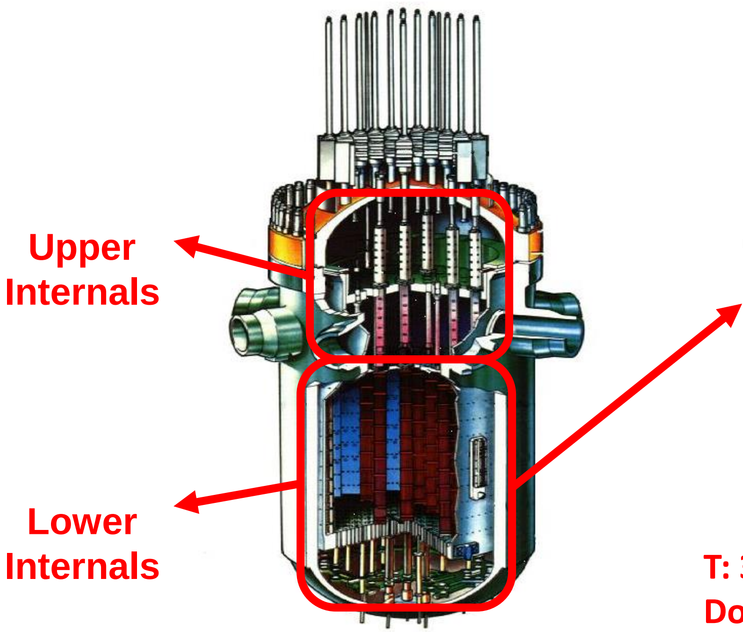
IRRADIATION EFFECTS ON INTERNALS: MICROSTRUCTURE & MECHANICAL PROPERTIES

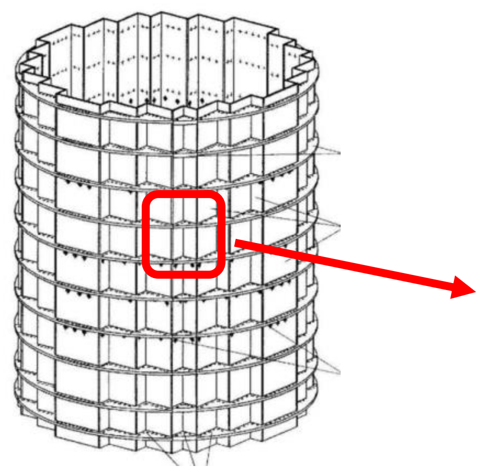
Benoit Tanguy

Section for Research on Irradiated Materials, CEA Saclay

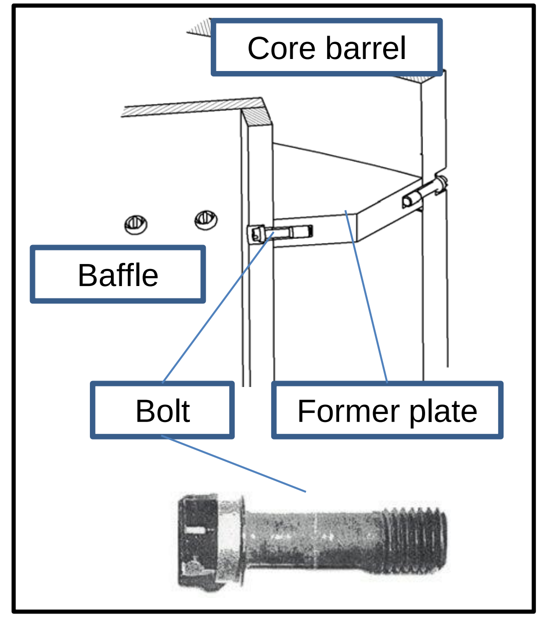
This project received funding under the Euratom research and training programme 2014-2018 under grant agreement N° 661913

Internals in LWRs





T: 300 – 380 ℃
Dose: up to 120 dpa after 60 years
Environment: primary water



Reactor pressure vessel

Lower internals

Design role of the Lower Internals:

- Support the core weight
- Circulation of the primary coolant
- Positioning of the core and fuel assemblies
- Protection of the RPV against irradiation embrittlement

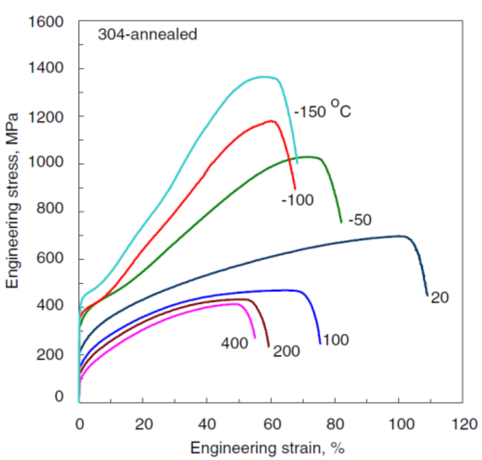
Internals: Materials (1/2)

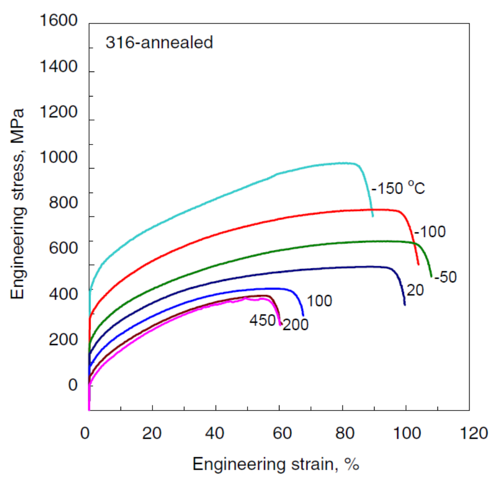
- Chemical composition (% weigth)

Grade	Ni	Cr	Mo	Ti	Nb	Mn	C	N	Si	Fe	Components
Type 304	8-10.5	17.5-19.5	--	--	--	2	0,07	0,1	0,75	Bal.	core barrel, former plate, baffle plate,control rod, top guide, core shroud,bolts
Type 316	10-14	16-18	2-3	--	--	2	0,08	0,1	0,75	Bal.	bolts
Type 347	9-13	17-19	--	--	10xC min,1.0 max	2	0,08	--	0,75	Bal.	bolts
Type 321	9-11	17-19	--	5xCmin-0,7	--	≤2	max 0,08		≤0,8	Bal.	bolts, core barel

  - L: low carbon (C<0,03-0,035); N: high nitrogen
  - Minor elements (ex for 316): P≤0,035; S≤0,030;Si≤1,0;Cu≤0,20;Co≤0,2)
  - Nb and Ti: limit the Chromium depletion during welding process

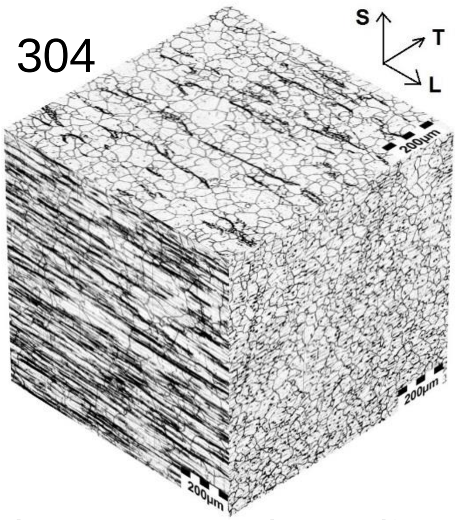
- Initial mechanical properties



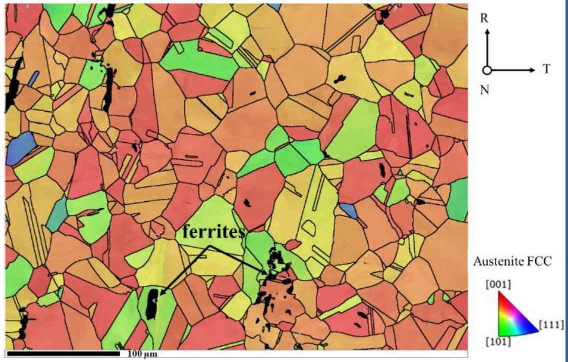


- SA (RT): YS~300MPa, UTS~650 Mpa
- SA (340°C): YS~200MPa, UTS~430 Mpa
- Fracture thougness: ductile (Jic~ 650 KJ/m^2)

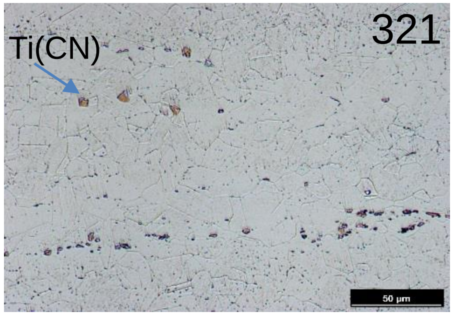
Internals: Materials (2/2)



γ Phase + Feα phase (few %)



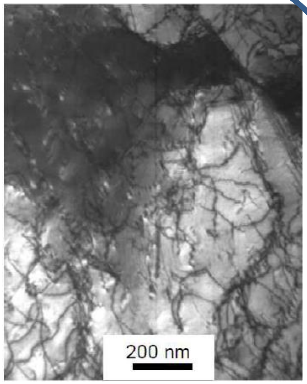
+ few MnS stringers



Inclusions of TiN, Ti(CN)

High density of deformation twins

Solution annealed (SA)
Low dislocations density
(ρ_{ini} = ~10^{10} m^{-2})



Cold Worked (CW)
High dislocations density
(ρ_{ini} = ~10^{14} m^{-2})
Twins

+ few inter- and intragranular precipitates: carbides

Sources: (Panait, Fontevraud 8), (Ernestova, Fontevraud 8), (Paccou, CEA PhD), (Gupta, CEA PhD)

Ageing of PWRs Internals

Internals: Amongst the most harshly irradiated components in a nuclear power plant

Component	Material	Temperature (°C)	Dose at 40 years* (dpa)	Dose at 60 years** (dpa)
Baffle bolts	CW-316 and 316L 17%Cr-11%Ni-2.5%Mo	~300 to 350	up to ~80	max 120
Baffle plate	SA-304L 18%Cr-10%Ni	~ 300 to 350	up to ~80	max 120
Former		~300 to 350	up to ~50	max 75
Core barrel		~ 300	up to ~ 10	max 15
Core barrel longitudinal and circumf. Welds	308L welds (SAW)	~ 300	up to ~ 10	max 15

(*) Calculated after 40 years operation, at 80 % effective full power.

(**) Estimation based on dose at 40 years

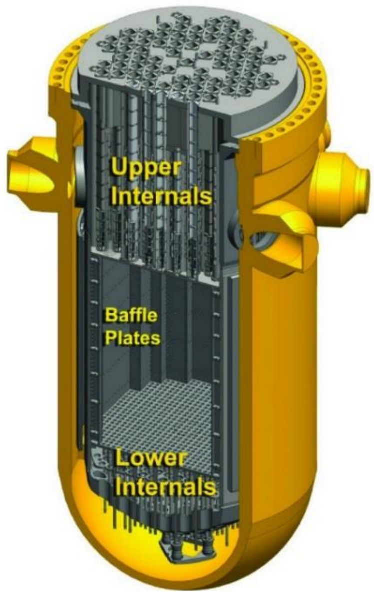
dpa: displacement per atom

1dpa ~6.5 10^{20} n/cm^2(E>1MeV)

- High dose irradiation can induce degradations not anticipated at the design stage
- Interactions with environment add more complex ageing phenomenon
- No surveillance programm (in contrast to RPV)

➢ Necessary to evaluate the potential long term evolution of the material properties

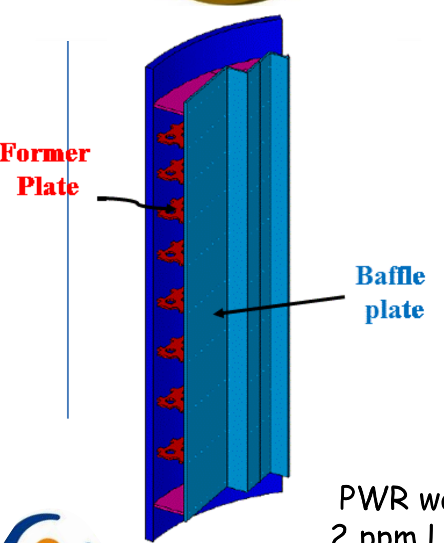
Ageing of PWRs Internals



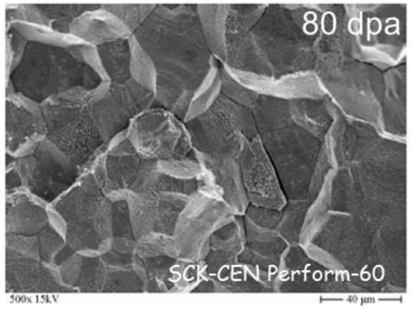
- Ageing issues related to lower Internals
  - Hardening, uniform elongation, and fracture toughness decreases
  - Creep under irradiation
  - Swelling (risk)
  - Radiation induced segregation, precipitation
  - Irradiation assisted stress corrosion cracking (IASCC)
  - Wear

Under irradiation

Environnment



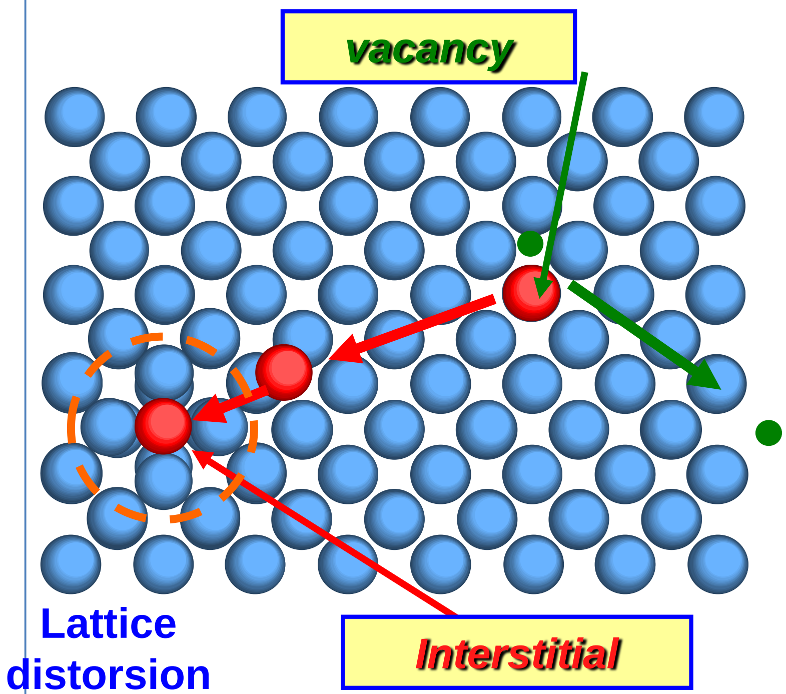
PWR water 155 bars, 288°C, 30ccH2/kg$H_2$0, 2 ppm Li, 1000ppm B, O2 < 5 ppb, pH300°C ≈ 7



Main microstructural and micro-chemical changes under irradiation

Irradiation effects on microstructure

- Formation of irradiation-induced defects at the nanoscacle



Interactions of neutrons with crystalline matters

- Nuclear Fission reaction: Emission of ~MeV neutrons
- Collisions between neutrons and material atoms: Elastic scattering, …
- Creation of Frenkel defects: Vacancies / Interstitials
- Agglomeration / recombinaison

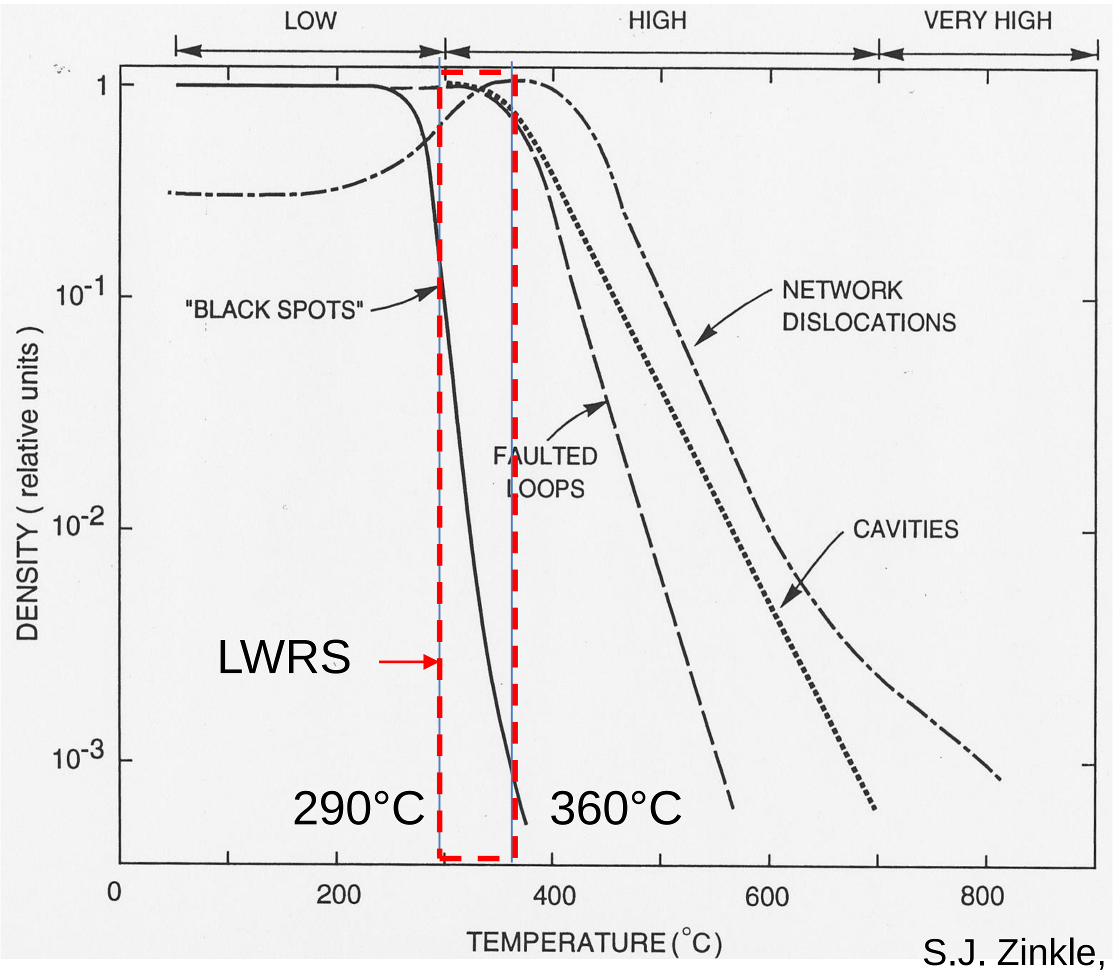
Most of the crystalline defect disappear, but some remains:

- 1D Vacancies, Interstitials
- 2D Dislocations
- 3D Voids, bubbles

Depending on materials composition and thermomechanical treatment /irradiations conditions: T(°C), neutron spectrum, flux

Irradiation effects on microstructure

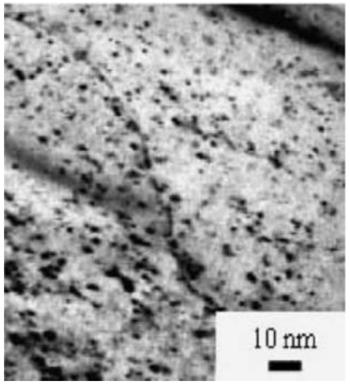
Temperature dependence of microstructural components in neutron-irradiated austenitic stainless steel (overview in 1993)



S.J. Zinkle, P.J. Maziasz & R.E. Stoller, J. Nucl. Mater. 206 (1993) 266

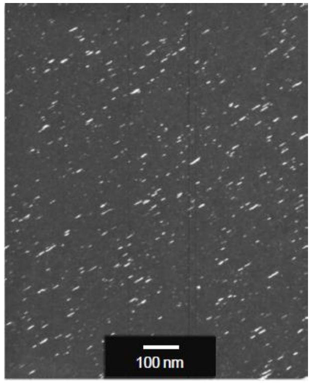
Irradiation effects on microstructure

❑ Formation of irradiation-induced defects at the nanoscacle

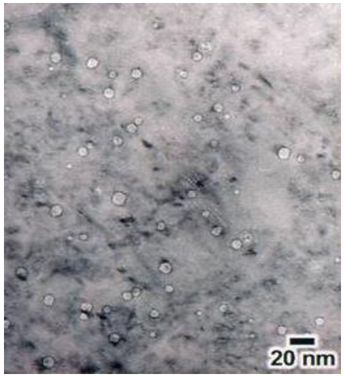


Black dots

PWR bolt (9 dpa, 312°C)

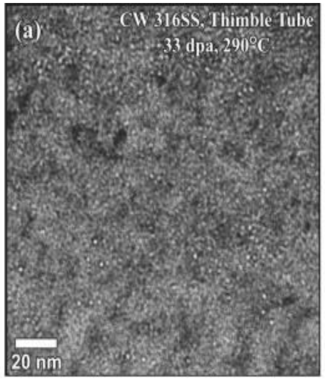


Frank loop (TEM DF image)



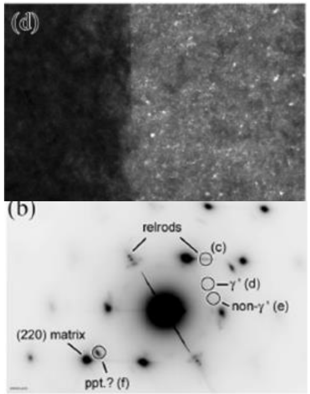
Nano-voids

PWR TT (33 dpa, 290°C)



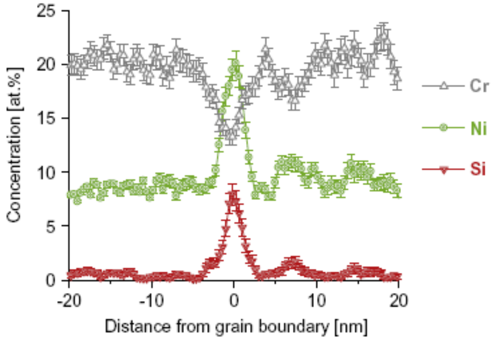
Gas bubbles

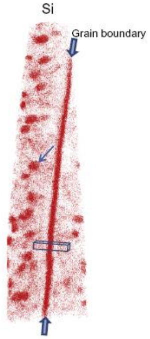
PWR bolt (7 dpa, 299°C)



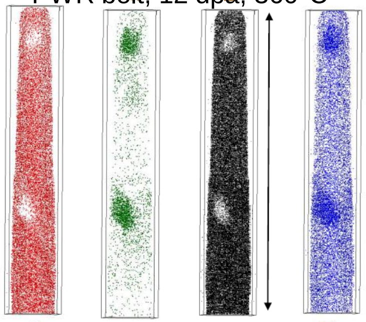
Precipitates (Ni_3Si, $M_{23}X_6$, G, …)

❑ Segregation at grain boundaries and on defects





PWR bolt, 12 dpa, 360°C



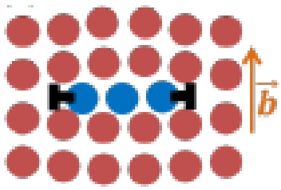
Cr Si Fe Ni

- ➢ Cr (Fe, Mn, Mo) depletion at GB
- ➢ Ni and Si enrichment at GB
- ➢ Segregation of Ni and Si at defects (FL, voids)

Sources: (Panait, Fontevraud 8) (Edwards et al.,2003, Pokor et al.2004) (Toyama, JNM,2011,2012) (Etienne, Fontevraud7), (Pokor,PAMELA)

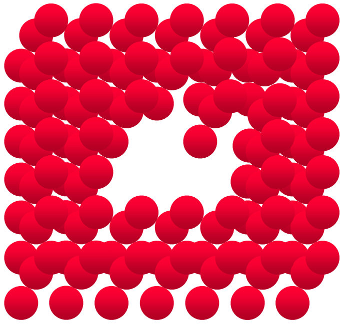
Irradiation effects on microstructure

- Frank loops: dislocation loops γ planes (111), b=a/3<111>, interstitial, faulted and sessile



- Gas bubbles:
  - Condensation of vacancies and helium atoms originating from transmutations reactions
  - Size of few nanometers, preferentially appear on dislocations, at gbs, or at the interface with some precipitate

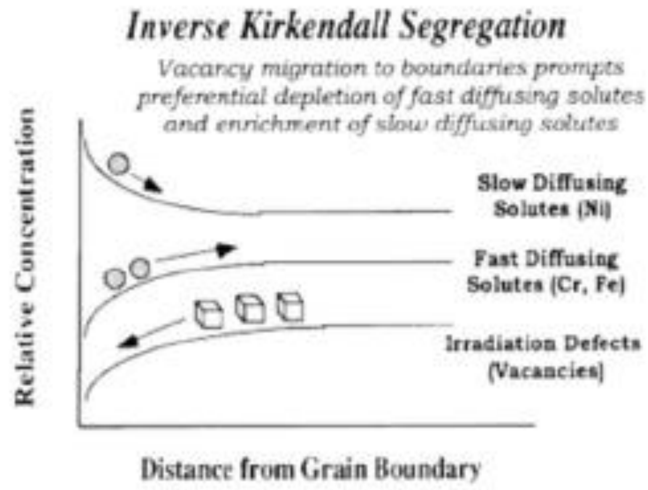
- Cavities:
  - Purely vacancy clusters
  - Grow due to supersaturation of vacancies in the matrix
  - preferentially appear in intragranular positions, possibly associate with some precipitate



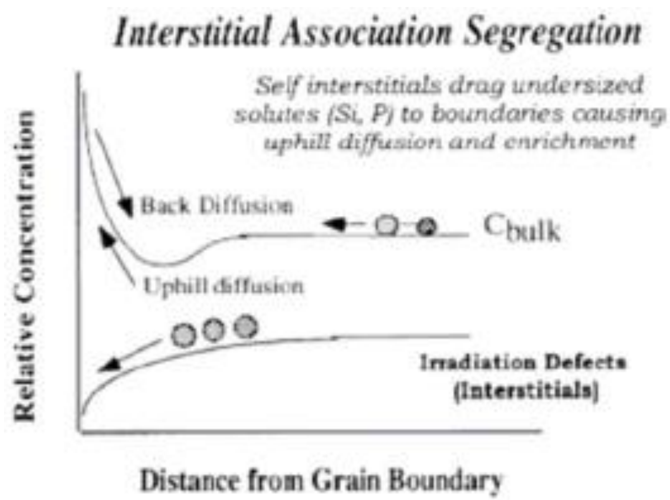
Irradiation effects on microstructure

- Radiation induced segregation (RIS):

  - Reverse Kirkendall effect: diffusion of v towards gb leads the elements that diffuse the most quickly in the reverse of this flow (Cr,Fe are depleted, Ni is enriched)

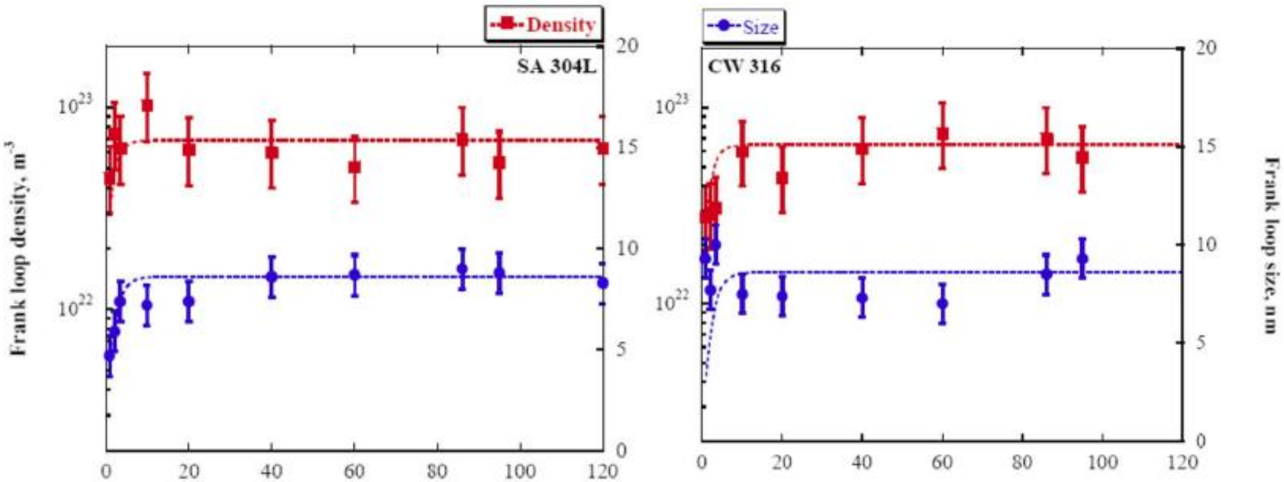


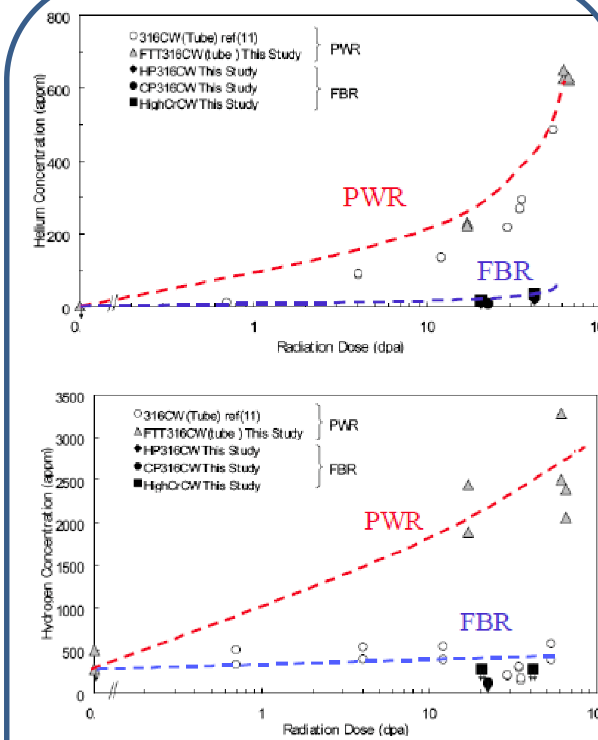
  - Interstitial association: elastic interaction between the flow of I point defects towards the gb and the material's atoms induce a movement of the smallest elements to the gb (enrichment of low size element as Si and P)



Irradiation effects on microstructure

❑ Evolution of Frank loops with dose (MTR data)





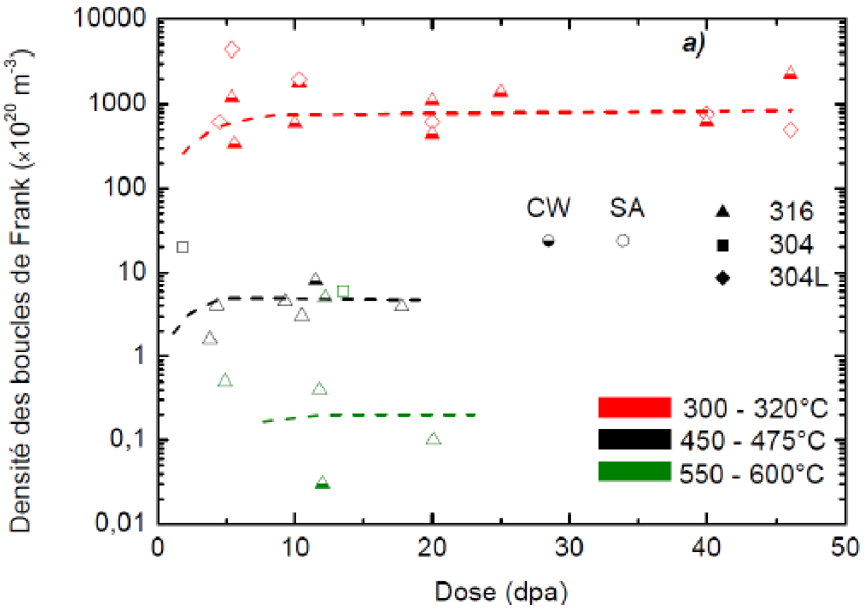
- ➢ Saturation of size and density of FLs around 5-10 dpa
- ➢ Only slight difference between SA 304 and CW316
- ➢ Little effect of the neutron irradiation flux and He rate production on the FL data (within the dose rate investigated) (T<360°C)

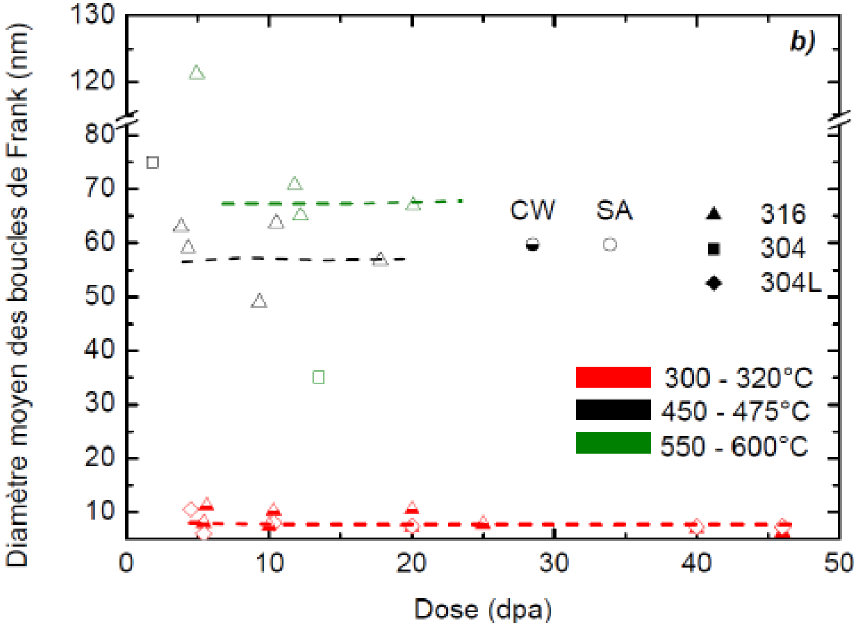
Comparison of He and H production betwen PWR and FBR

Dose rate (dpa/s): 9,410-7 (Bor 60), 2,910-7(Osiris), 1,410-6(EBRII)

Irradiation effects on microstructure

- Evolution of Frank loops with temperature (MTR data)



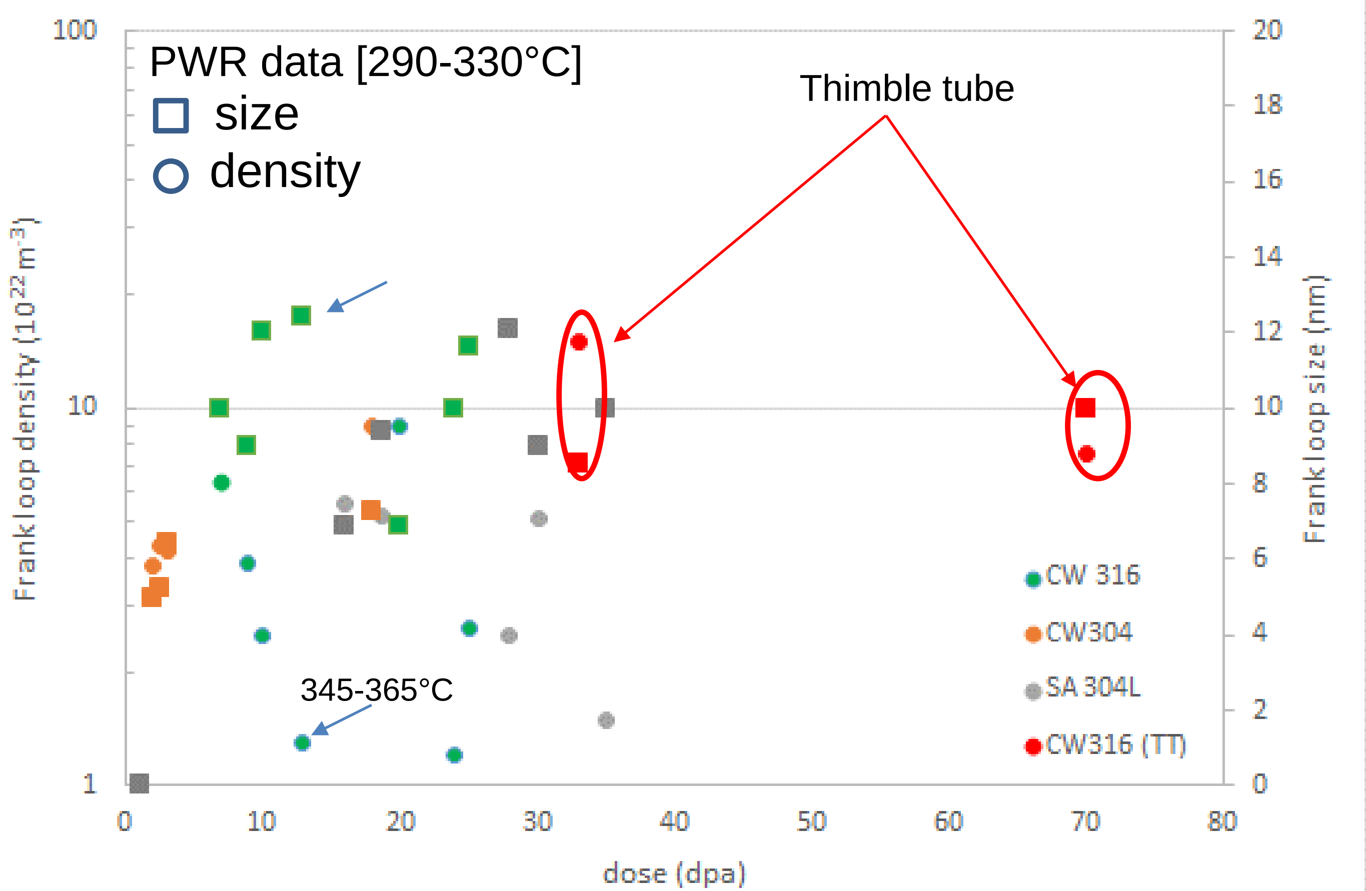


➢ With increasing temperature, FL density decreases, FL diameter increases

Source: (Zouari, PhD, 2012)

Irradiation effects on microstructure

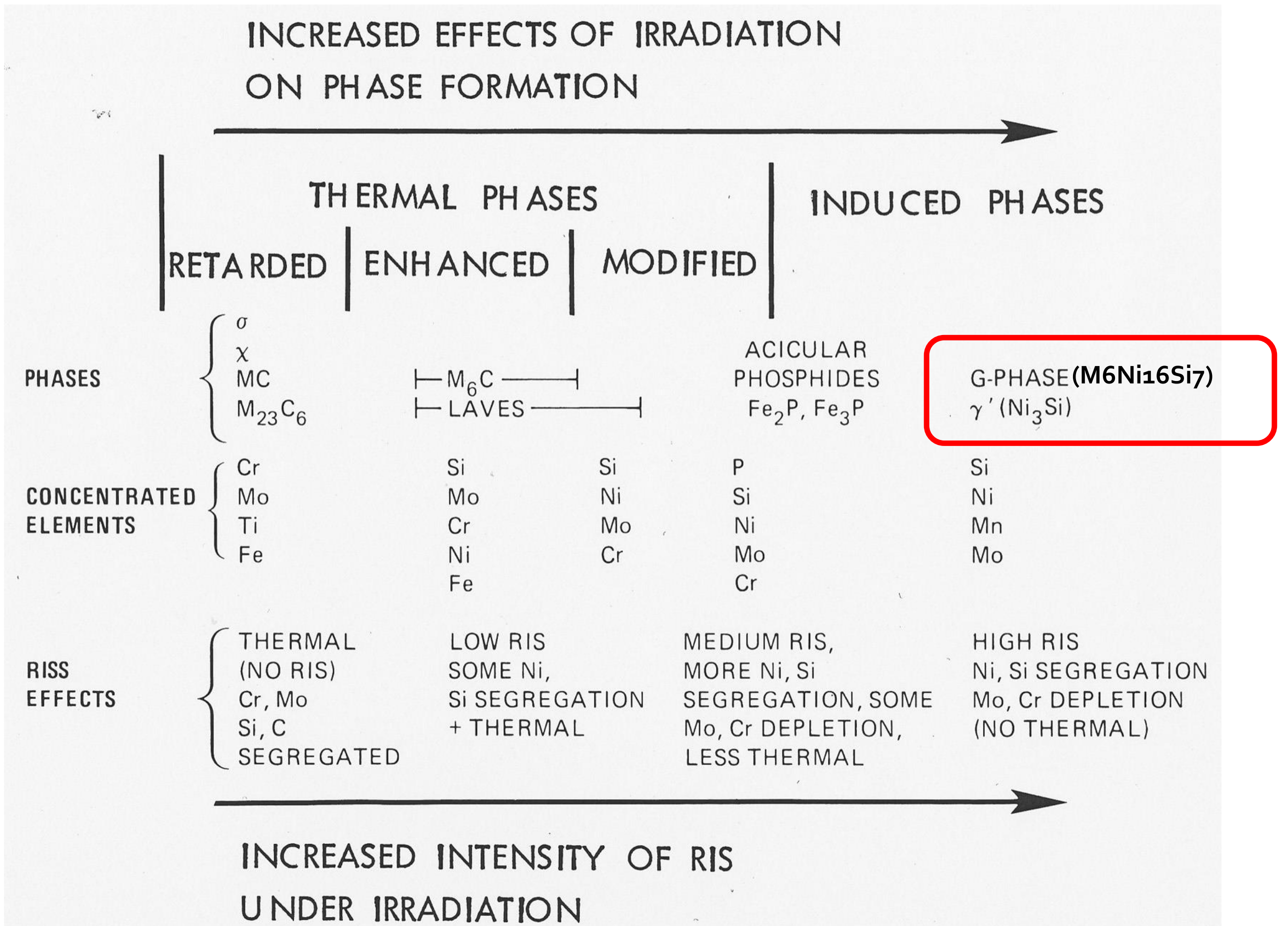
❑ Evolution of Frank loops with dose (PWR data)



- ➢ High density of FLs after few dpa
- ➢ No siginificant difference of metallurgical state (CW/SA)
- ➢ Trends to saturate for size and density
- ➢ In agreement with FBRs data

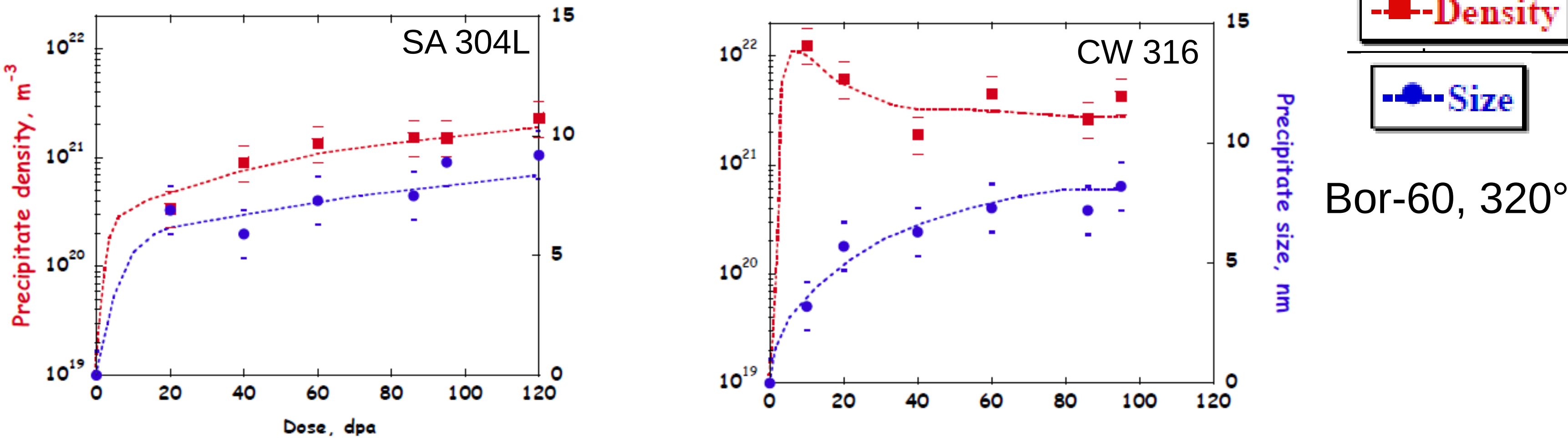
Sources: (Monnet, Fontevraud 4, 1998) (Cauvin, Fontevraud 3, 1994) (Pironet, Fontevraud 4,1998), (Pokor, Fontevraud 5, 2006), Panait, Fontevraud 8, 2014) (Edwards, Font. 6,2006) (Renault, Fontevraud 9,2018) (Goltrant, Fontevraud 3,1994)

❑ Overview of precipitation in irradiated austenitic SS



Irradiation effects on microstructure

❑ Evolution of precipitates with dose (MTR data)



Bor-60, 320°C

- $M_{23}X_6$, M_6X and/or $M_6Ni_{16}Si_7$ (G) with M=Cr, Fe, Ni, … and X = C, Si
- Quite similar size between SA 304L and CW 316
- Density higher for CW 316 than SA 304L; especially at low doses
- Increasing density and size with dose in SA 304L, saturation for CW316

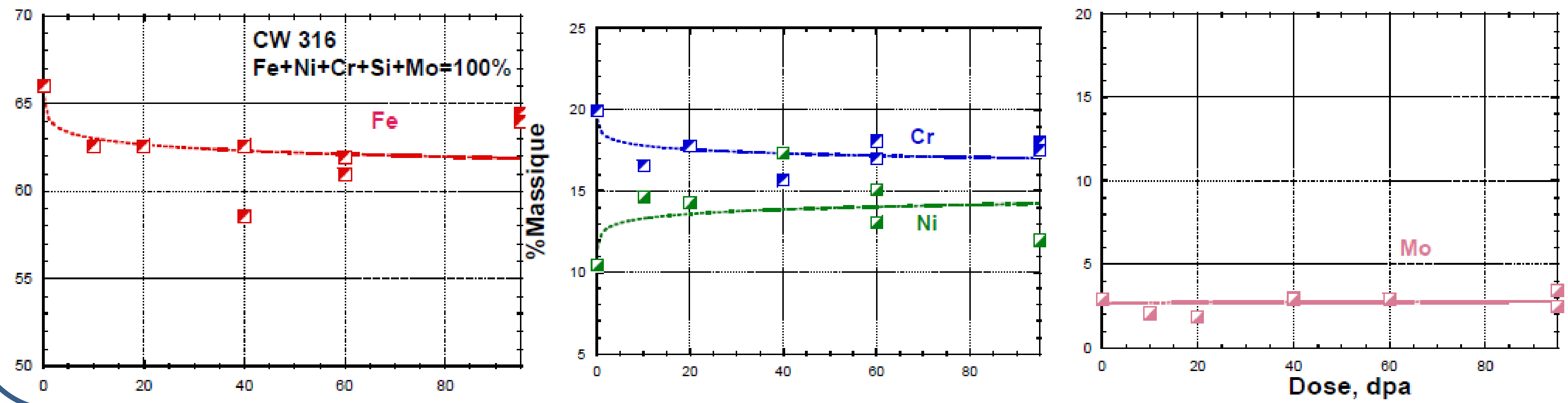
- No significant effect of neutron spectrum (FR/LWR)

Source: (Renault et al.,Env Deg 2009)

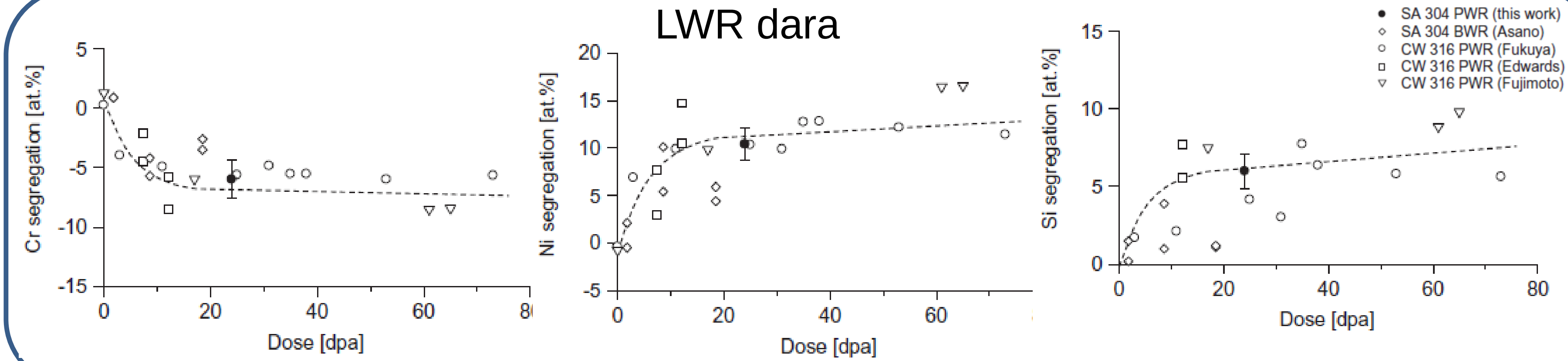
Evolution of RIS with dose (MTR/LWR data)

CW 316, Bor-60, 320°C

same trends for SA 304L



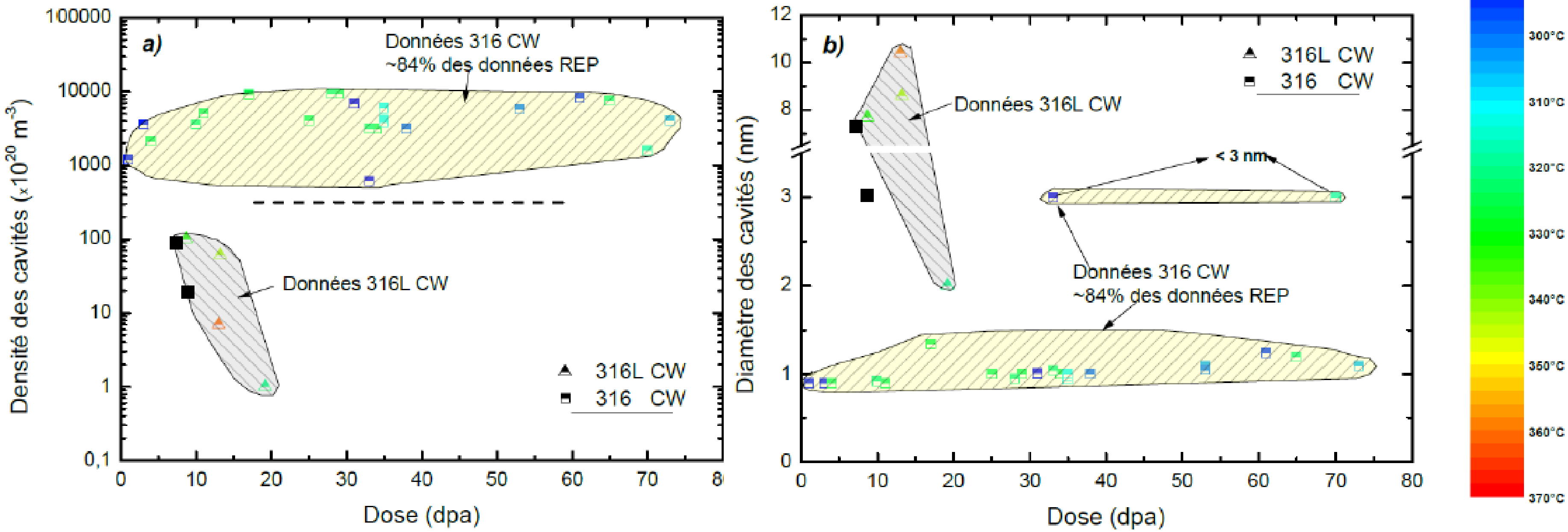
LWR dara



Sources: (Renault, Env Deg, 2009) (Toyama, JNM,2010)

Irradiation effects on microstructure

- Evolution of cavities with dose (PWR data)



- <u>Data only for CW316</u>
- Cavities observed at T<300°C (diameter ~1nm)
- Significant differences between CW 316 and CW 316L (T effect, carbon effect?)

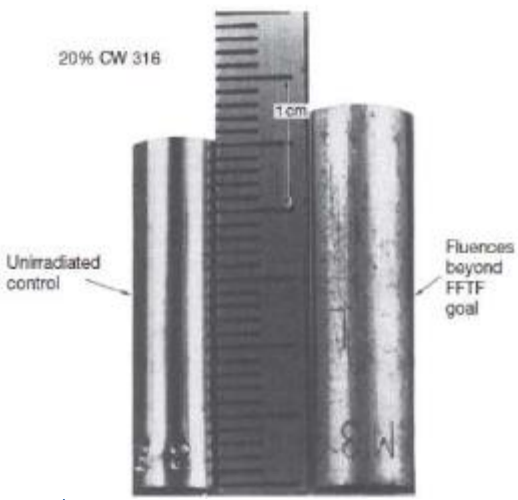
- Strong dependance to flux and spectrum

Source: (Zouari.,PhD 2012) (Panait, Fontevraud8,2014,■)

Swelling as a consequence of voids formation and growth

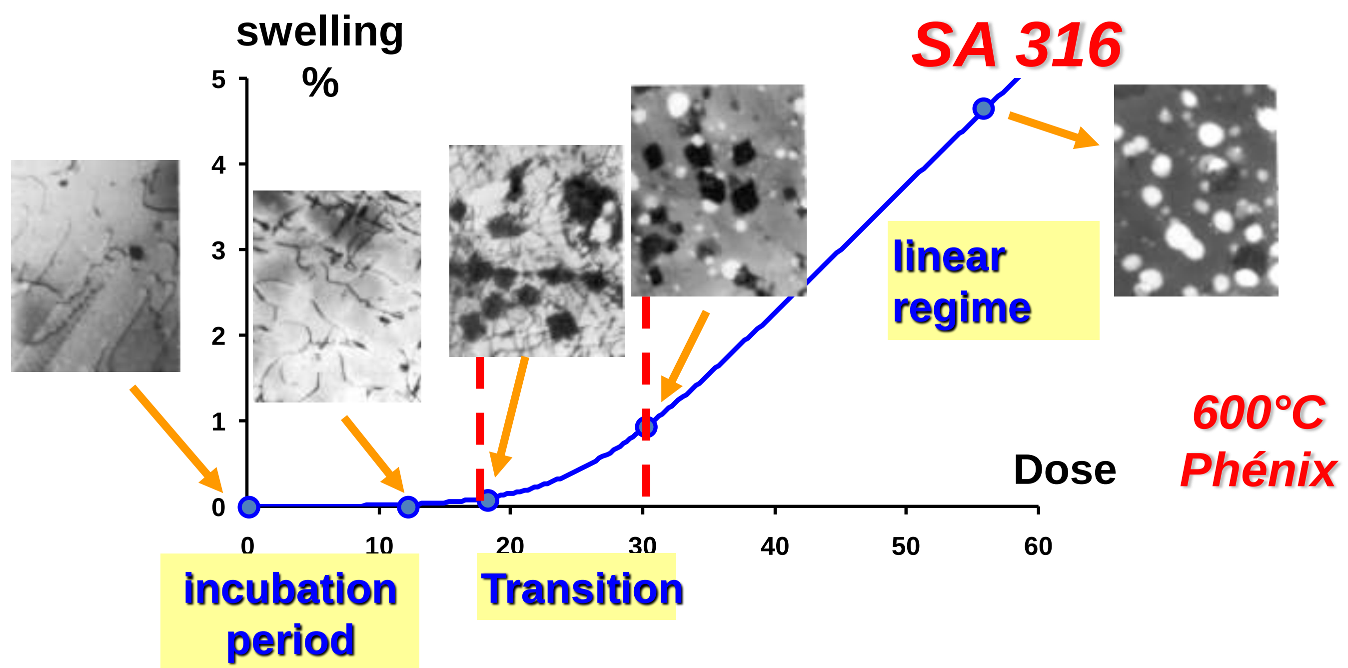
- Evolution of swelling with dose (Fast reactor data)

Swelling: volume increase in a material caused by void formation



CW 316, EBR-II [Straalsund, 1982].

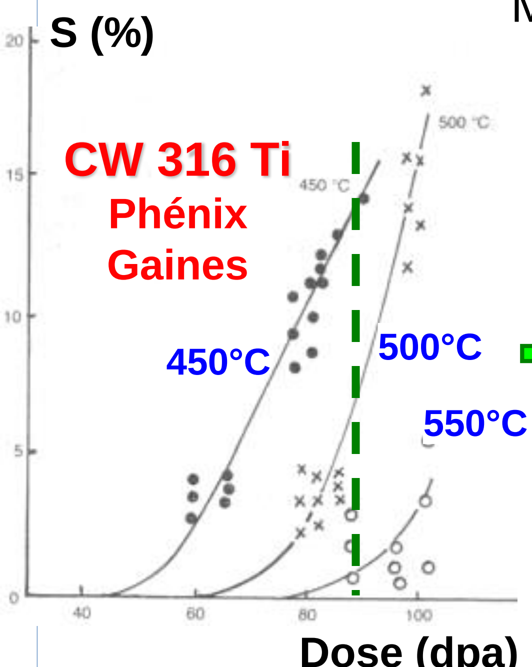
Void formation: nucleation + growth



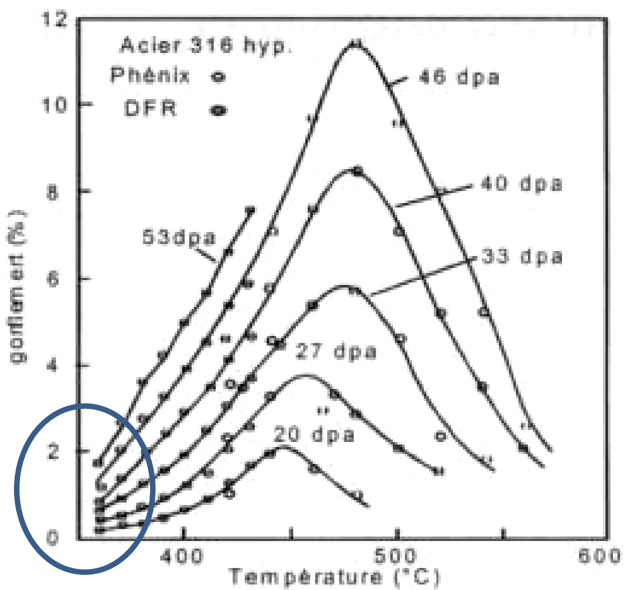
Source: (Dubuisson, 2011)

Swelling as a consequence of voids formation and growth

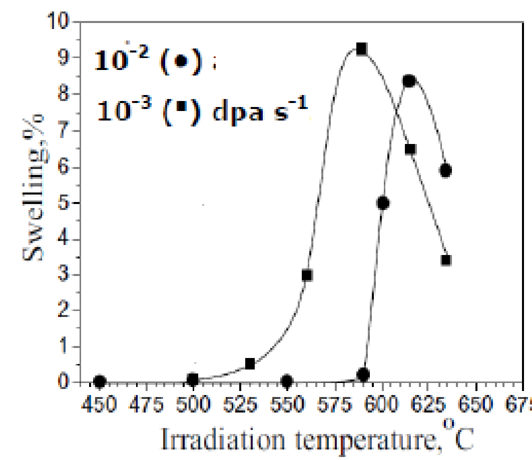
- Evolution of swelling with dose (Fast reactor data)



Maximum swelling = f(temperature)



Temperature maximum swelling decrease with decreasing strain rate



Data from fast reactor can not be directly used to asses swelling in PWRS

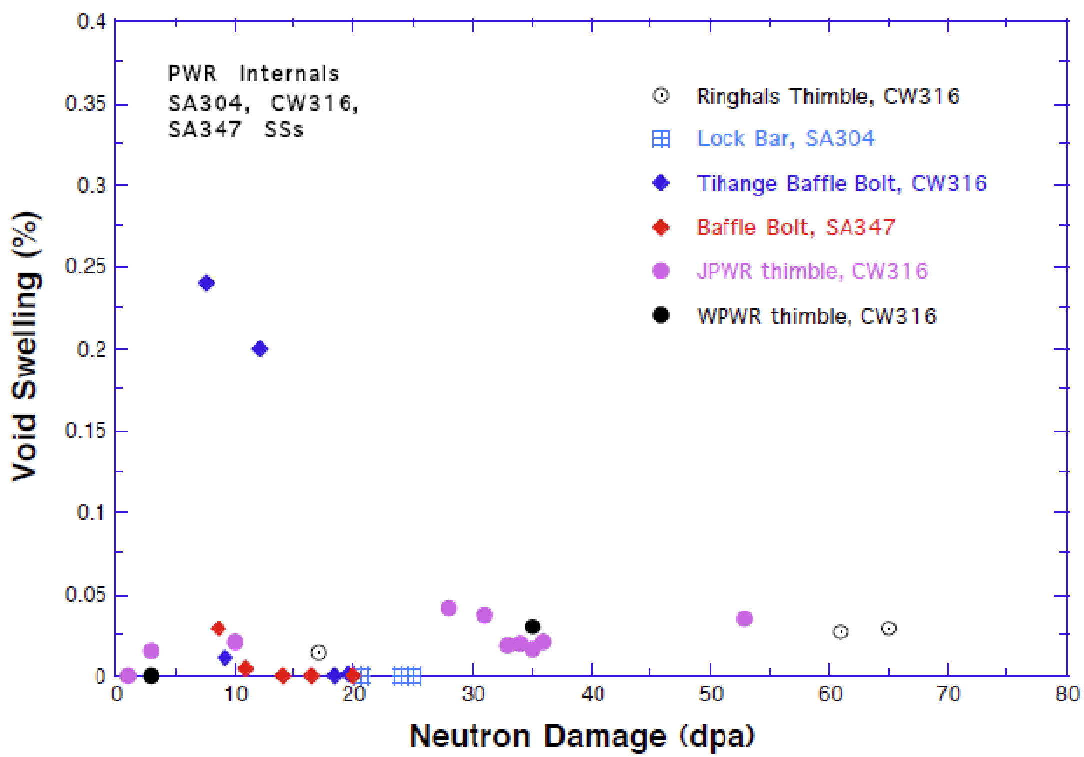
Swelling=f(dose, dose rate, temperature, helium production)

Source: (Dubuisson, 2011) (Borodin and al.,2005)

Swelling as a consequence of voids formation and growth

❑ Evolution of swelling with dose (PWR data)

Based on voids density and size

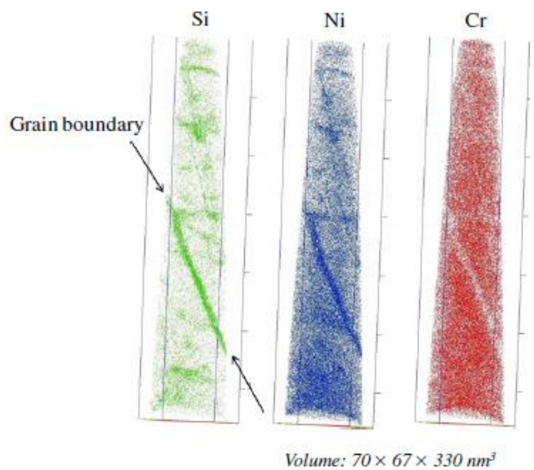


Source: (Chung, NUREG 2006)

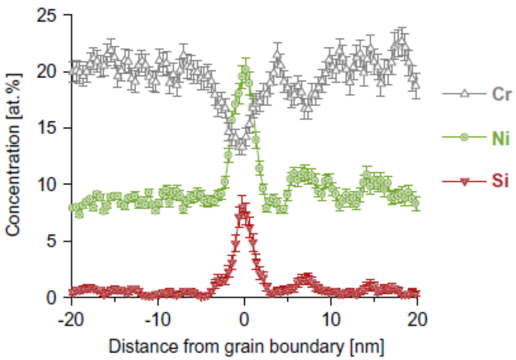
Irradiation effects on microstructure

- Recent insights based on advances in microstructural characterization (APT, HR-TEM): RIS at GB

  - Trends confirmed on Fe(-), Cr(-), Mo(-), Mn(-), Ni(+),(Si(+), P(+)
  - Finer measurements of chemical segregation at interfaces (higher segregation level based on APT)
  - Enrichment of minor elements (B, Cu)



(CW316, 12 dpa, 360°C, Bolt)

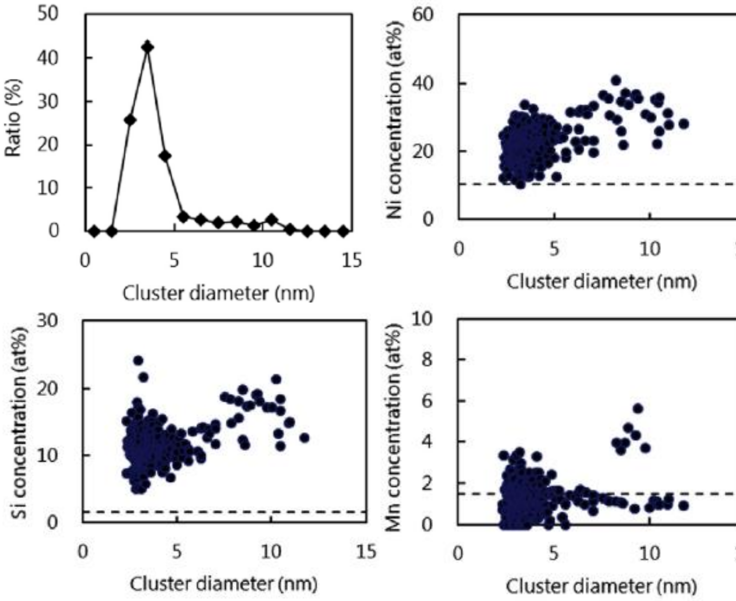


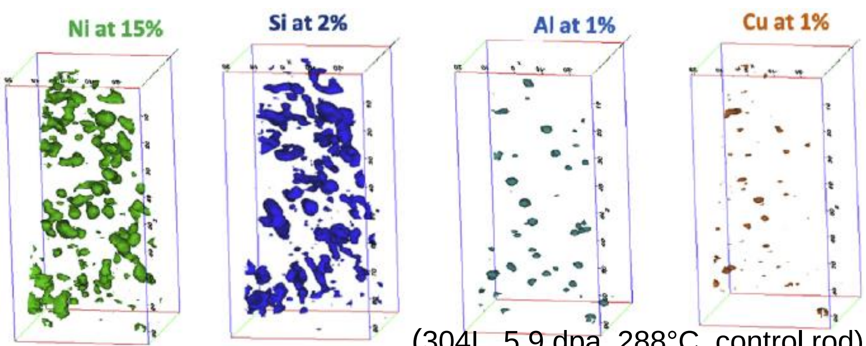
(304, 24 dpa, 300°C, Fuel Wrapper)

Sources: (Etienne and al, JNM,2008) (Fujii and Fukuya, JNM,2016) (Toyama andal., JNM, 2012)

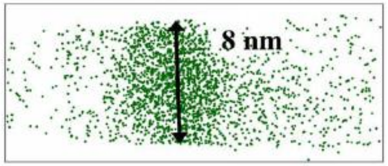
Irradiation effects on microstructure

- Recent insights based on advances in microstructural characterization (APT, HR-TEM): Matrix
  - Formation of intragranular (Ni,Si) (Ni,Si,Mn), (Al-Cu) –rich solute clusters
  - Segregation to dislocation loops and dislocation segments
  - Element's enrichment = f(size cluster)
  - Complementary insights on precipitates formation

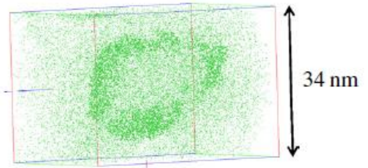




(304L, 5,9 dpa, 288°C, control rod)



(CW316, 12 dpa, 360°C, Bolt)



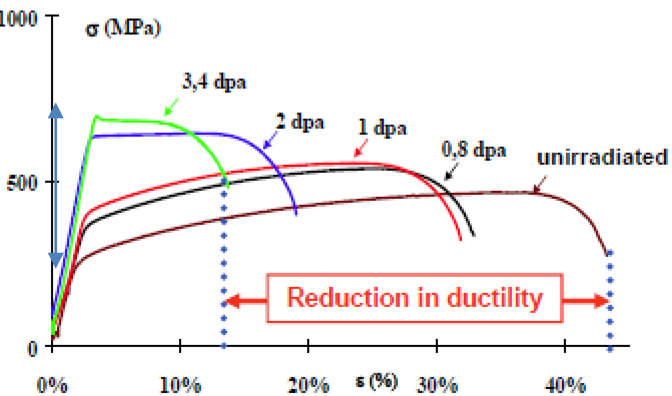
CW316, 10 dpa, 10MeV Fe, 5.6 10-4 dpa/s

Sources: (Etienne and al, JNM,2008,2010) (Fujii and Fukuya, JNM,2016) (Jiao and al, JNM,2018)

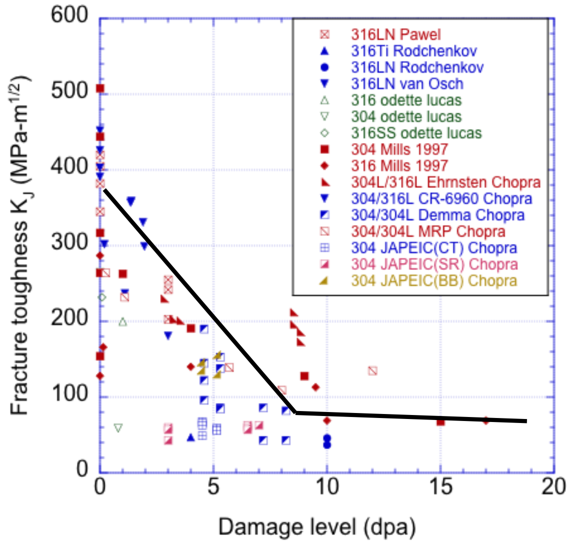
Main mechanical properties changes under irradiation

Irradiation effects on mechanical properties

Increase in yield stress



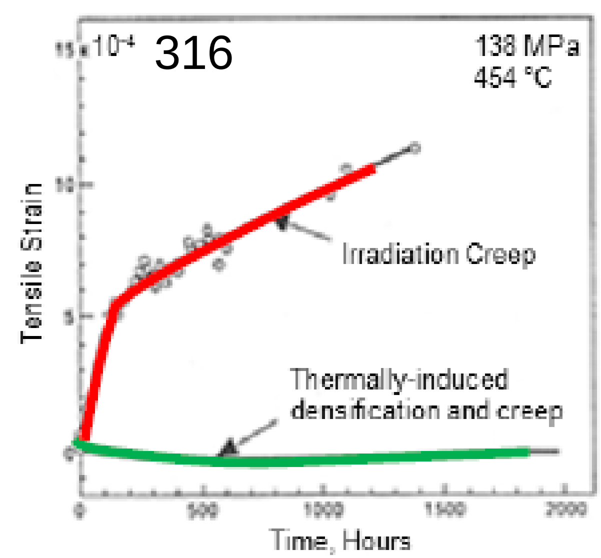
Hardening and loss of ductility



[Zinkle et Was (2013)]

Fracture toughness decrease

Irradiation creep

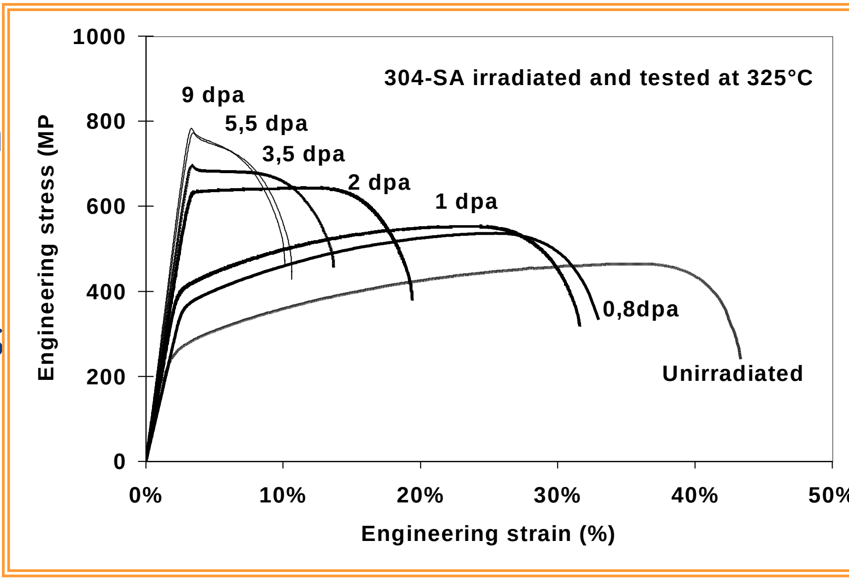


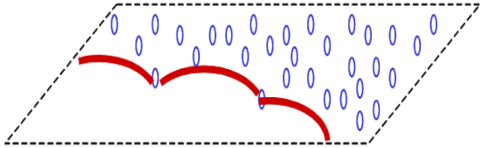
[Gilbert and al. (1972)]

Irradiation effects on mechanical properties

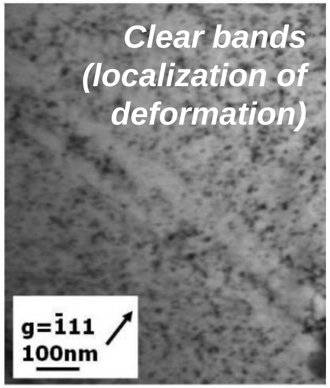
❑ Radiation hardening

- Point defect clusters and precipitates produced by irradiation act, to varying extent, as obstacles to dislocation motion.
- Barrier strength of obtacles depends on their nature (cavities> largeFL> small FIs, bubbles)
- Large increase of yield stress and UTS (up to 5 times) and decrease of UE and TE
- Decrease of strain hardening capacity
- Enhanced localization of deformation at microscale





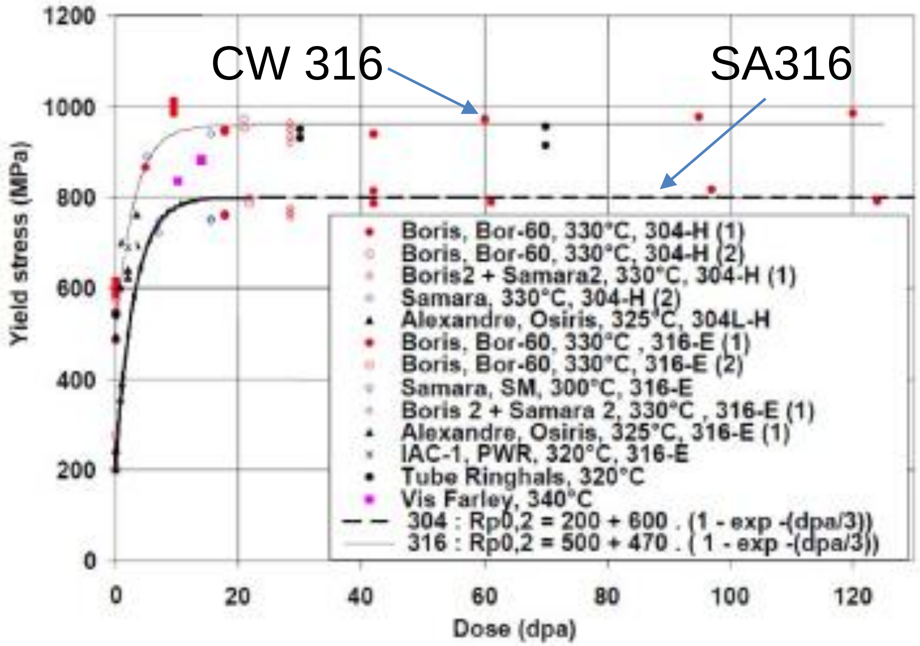
Frank loops acts as forest obstacles to mobile dislocations

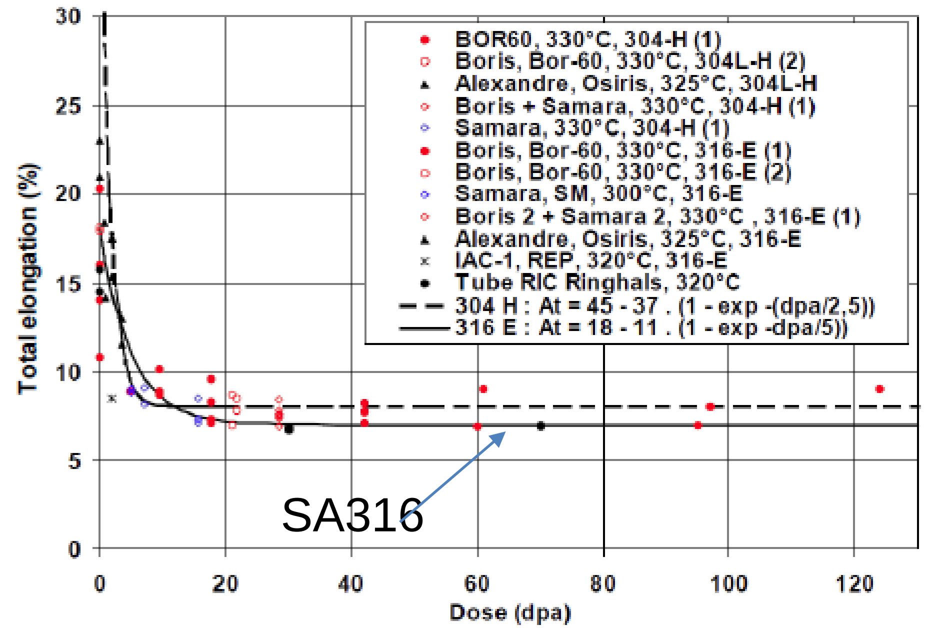


Source: (Pokor, JNM,2004)

Irradiation effects on mechanical properties

- Radiation hardening evolution with dose (MTR/LWR data)





- Strong yield stress increase up to 5-10 dpa, saturation above 8-10 dpa
- Marked decrease of uniform (-~0%) and total elongation
- No significant effect of flux, spectrum on yield stress evolution

Irradiation effects on mechanical properties

❑ Modeling Irradiation hardening

- Primarily irradiation hardening features include: Black dots, dislocation loops, network dislocations, cavities – void and helium bubbles, precipitates, solutes often associated with defect clusters
- In the simple case, modelling radiation hardening requires treating:
  - Dislocations – obstacles strength interaction: α_{obs} (d)
  - Superposition of strength contributions from various obtacles – pre and post irradiation

Irradiation effects on mechanical properties

❑ Modeling Irradiation hardening - Dispersed obstacle Hardening

Taylor factor~3,06 (FCC)

Shear modulus

$$\Delta\sigma_k = \alpha_k M \mu b (N_k d_k)^{1/2}$$

Obstacle strength

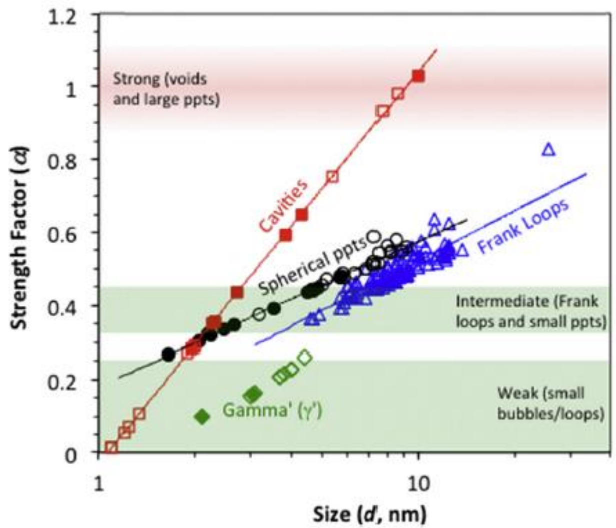
Magnitude of the Burgers vector

$(N_k d_k)^{-1/2}$: average spacing for rigid non-shearable obstacles

α_k depends on the nature of the obstacle and the mechanism of interaction
->calibration based on experimental data

Also $\Delta\sigma_k = \alpha_k M \mu b d_k (N_k)^{2/3}$ (weak obstacles) Friedel Kroupa Hirsch

Dependence on dose (=f(solute segregation at defects,…) ?
-> Required Physically-based modelling



Source: (Tan and Busby, JNM, 2015)

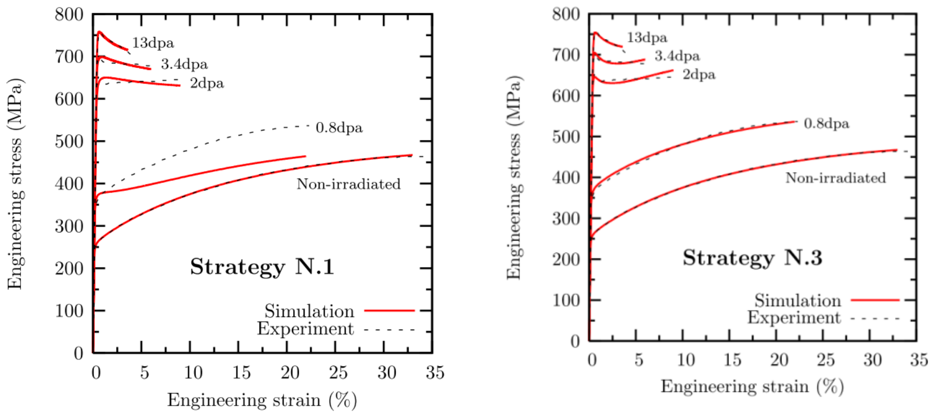
Irradiation effects on mechanical properties

- Modeling Irradiation hardening - Dispersed obstacle Hardening
  - Superposition laws:

Root-sum square law $\Delta\sigma = \sqrt{\sum_k (\Delta\sigma_k)^2}$ [Obstacles with similar strengths]

Linear law $\Delta\sigma = \sum_k \Delta\sigma_k$ [Obstacles with dissimilar strengths]

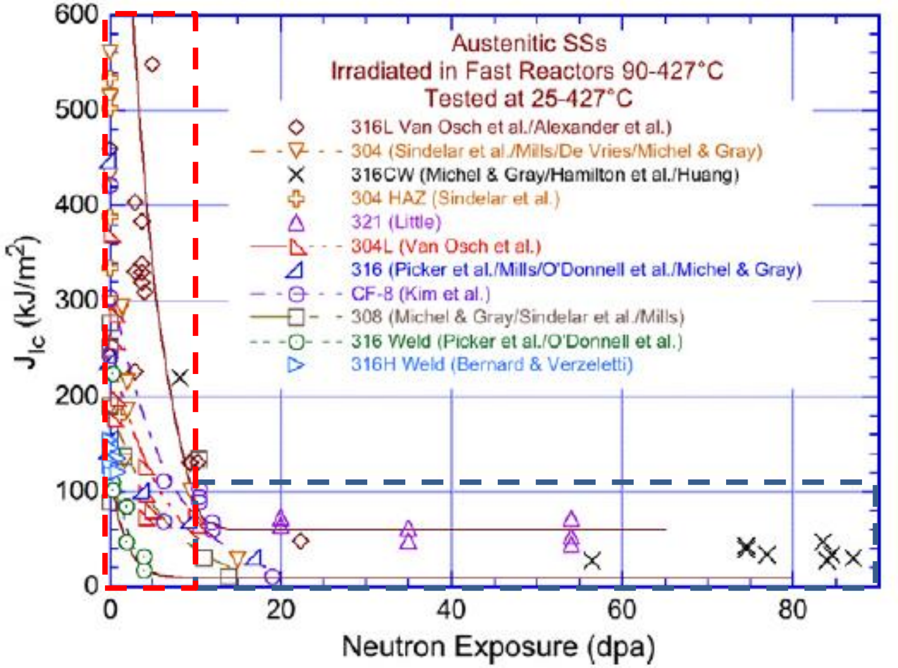
  - Appropriate choice not obvious, requires physically-based modelling



e.g.: cristal plasticity modeling (Thèse X. Han, CEA-EDF)

Irradiation effects on mechanical properties

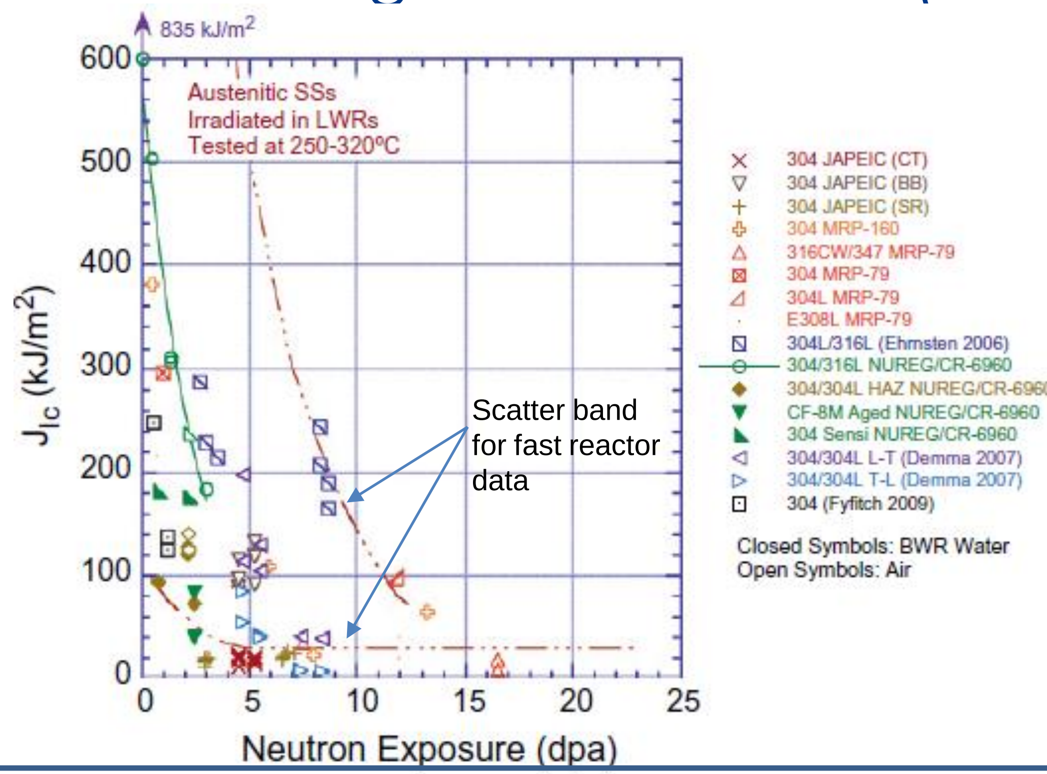
- Effect of irradiation on Fracture toughness
  - Irradiation also reduces the fracture toughness of stainless steels
  - Relevant and valid data on the properties of the structural materials are needed for assessment of the structural integrity and remaining lifetime of NPP reactor internals
  - Most available data on fracture toughness are from materials irradiated in fast reactors and data on material from LWRs are still very scarce



Source: (Chopra and Rao, JNM, 2011)

Irradiation effects on mechanical properties

❑ Evolution of Fracture toughness with dose (LWRs)



- ➢ Trends in agreement with fast reactor data (few data on CW316)
- ➢ Marked effect of specimen orientation (linked to metallurgical features)
- ➢ Same trend but large difference between similar grades -> link with microstructural features ?

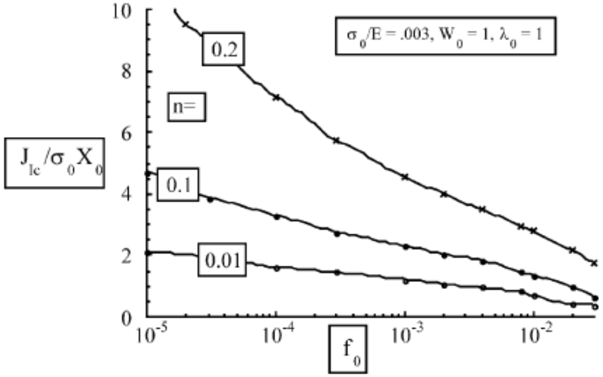
Source: (Chopra and Rao, JNM, 2011)

Irradiation effects on mechanical properties

❑ Open questions for irradiated materials

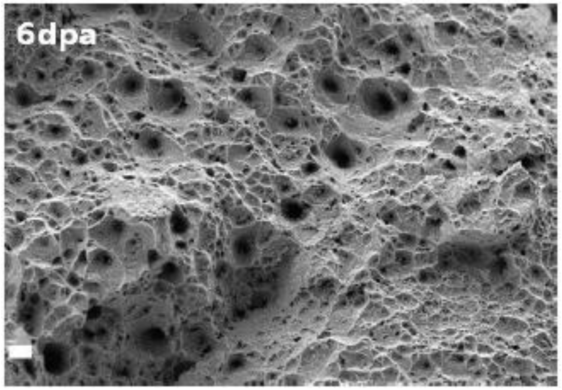
- Decrease of toughness with irradiation seems stronger than expected

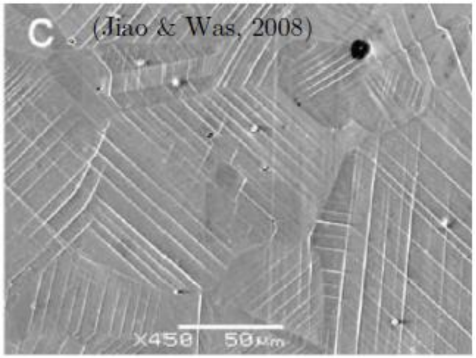
  - ✓ Hardening -> Jc x(2-4) [Jc~ $\alpha\sigma_y \times \lambda$]
  - ✓ Loss of strain hardening -> Jc/(5-10)

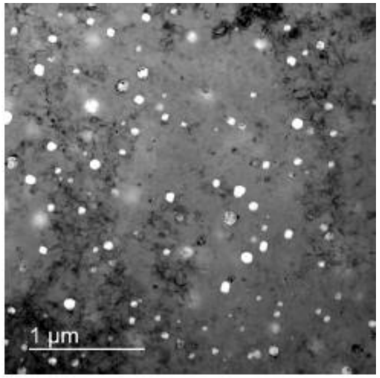


[Pardoen,Acta Mater.2003]

Physical mechanisms of voids growth in irradiated materials?



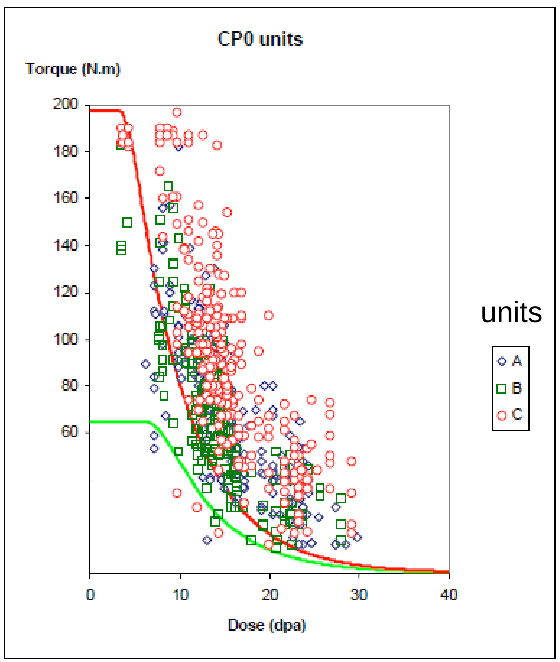




Irradiation-creep

Data from components

Measurement of residual torques during baffle-bolts removal

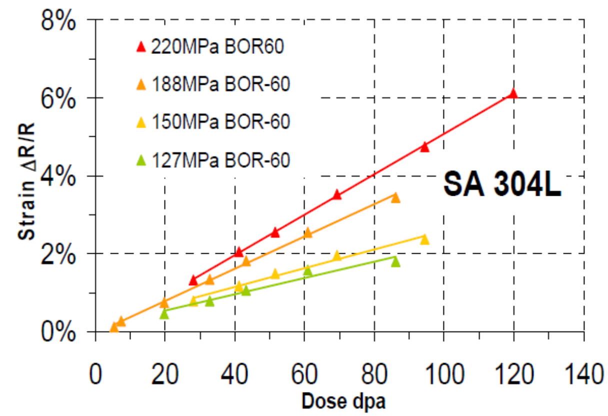


[Lemaire et al; Fontevraud 7]

Assessment of irradiation-creep law

-> Relaxation experiments under irradiation in MTR
-> assessment of the stress effect on the kinetics of relaxation

Pressurised tube

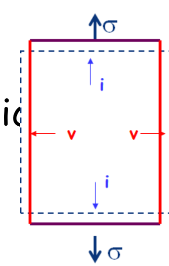


$$\dot{\varepsilon} = A\,\varphi\,\sigma^n \exp(-\frac{Q}{kT}).$$

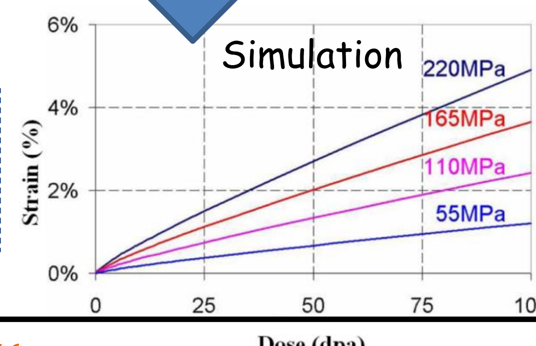
[Garnier et al; Fontevraud 7]

Mechanisms understanding

Several types of potential Mechanisms (clusters dynamic modelling))



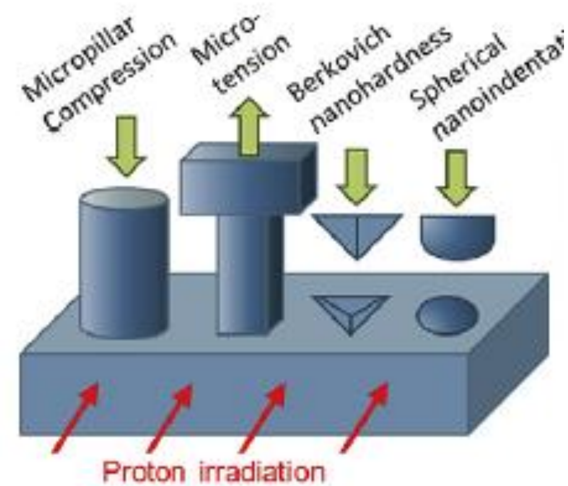
Simulation

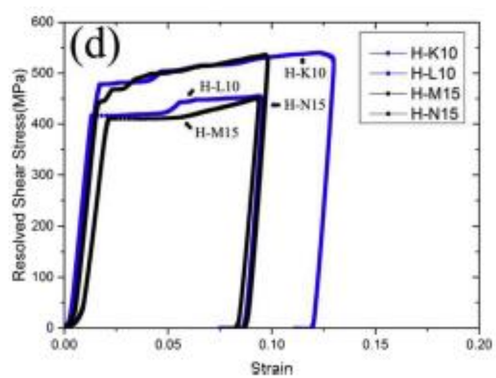


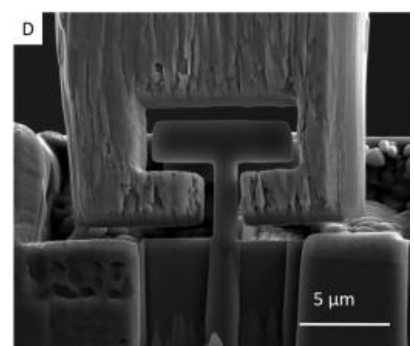
[Garnier et al; Fontevraud 7]

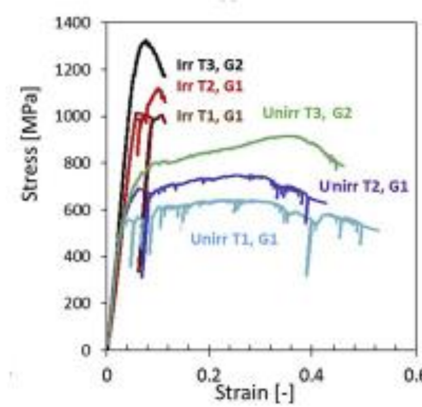
Irradiation effects on mechanical properties

- Recent insights based on small scale mechanical tests techniques
  - Assessment of the mechanical properties at the crystal scale: mechanical anisotropy and heterogeneities of the materials
  - Micro-compression: assess to the CRSS as a function of irradiation and crystallographic orientations
  - Micro-tension: assess to strain to failure





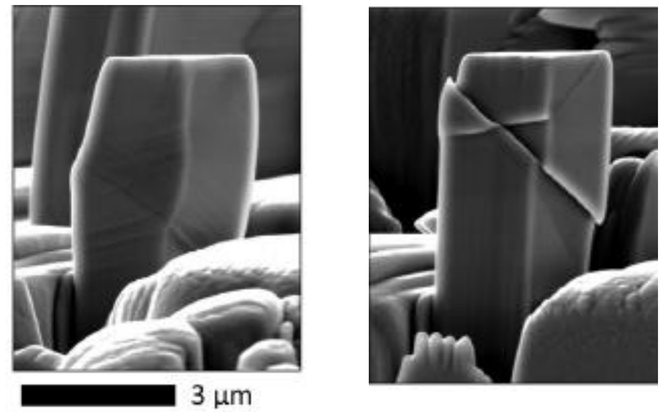


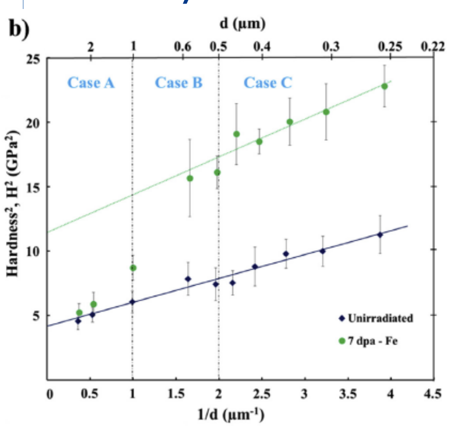


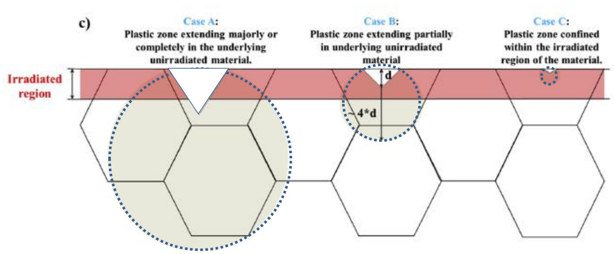
Source: (Vo and al., JNM,2017)(Weaver,2017, JNM) (Jin et al;, JNM, 2016)

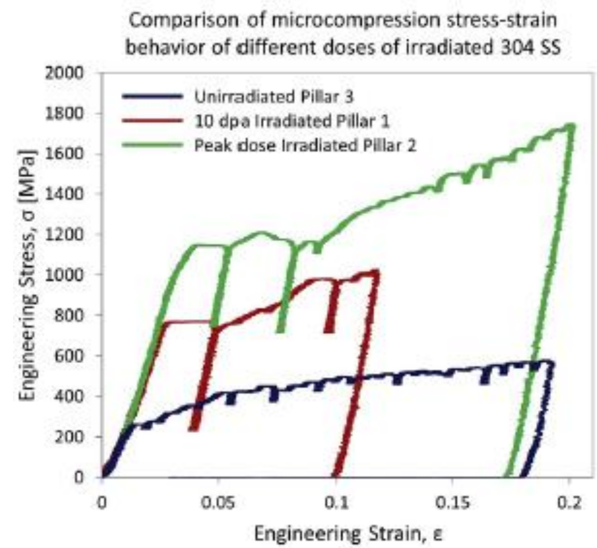
Irradiation effects on mechanical properties

- Recent insights based on small scale mechanical tests techniques
  - Nanoindentation: information on hardening induced by irradiation
  - Tension/compression: information on localized deformation induced by irradiation





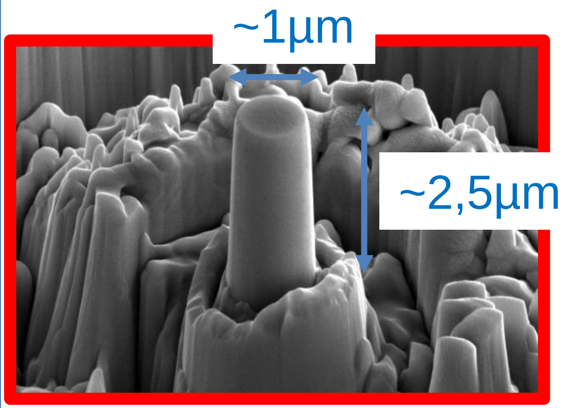




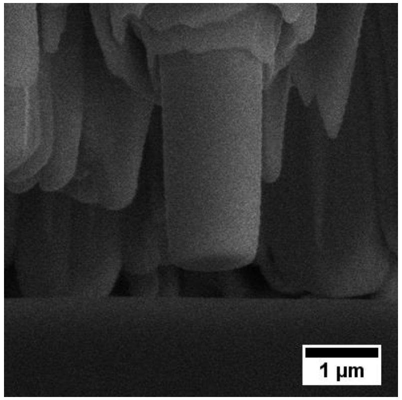
Source: (Gupta et al., JNM, 2017) (Reichart and al., JNM,2017)

Irradiation effects on mechanical properties

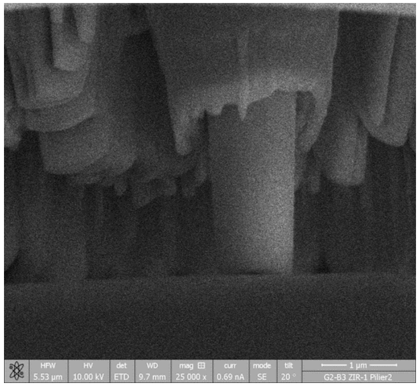
❑ Micro-compression on ion-irradiated 304L

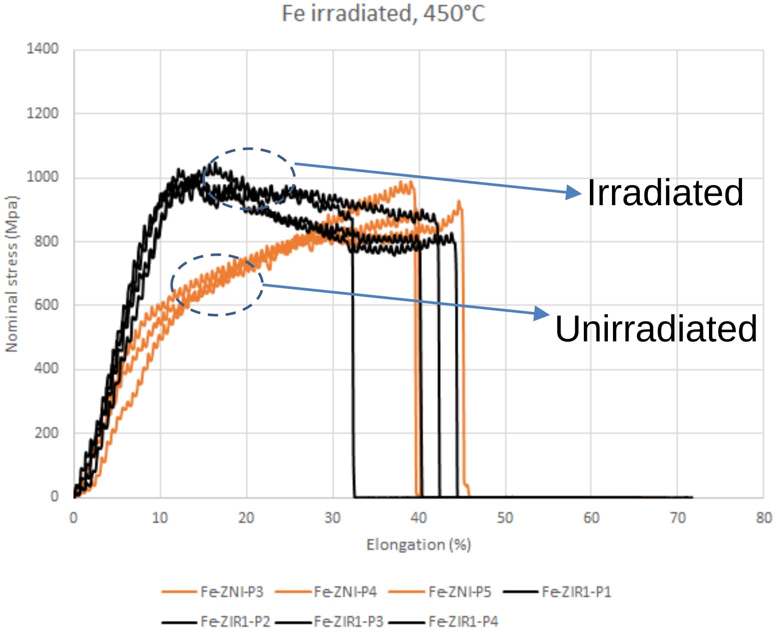


Un-irradiated



Irradiated



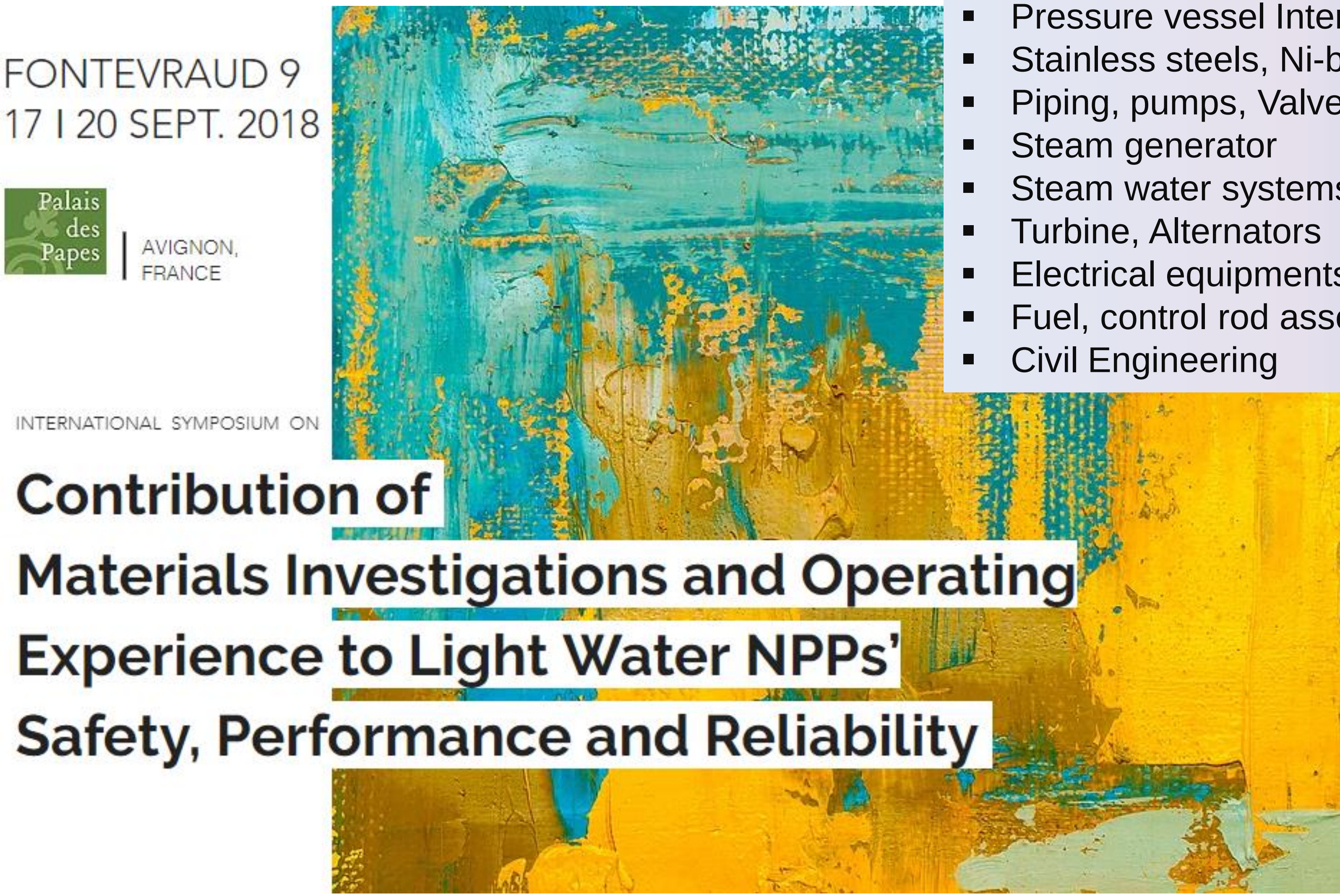


Source: (courtesy of Elie Paccou, PhD, CEA, collaboration with CNRS CEMES) (On-going study)

Conclusions and perspectives

- Several R&D national and international programs related to ageing of Internals since 80-90s
  - Large amount of characterizations on 304-316 industrial alloys
  - General trends of mechanical properties evolution with dose well-established
  - Larger scatter can be observed for a same grade (fracture toughness, IASCC)
  - New insigths with recent characterisation at the local scale (atom-scale characterizations (APT, ….) on precipitates, segregation at interfaces,…), small specimens, in-situ measurements

- More recently, a large effort put on the development of predictive modeling as a support tool to engineening approaches
  - Refining the understanding and modelling of plasticity and fracture mechanisms and their evolution with irradiation
  - Building the link between microscale and mechanical properties



- Pressure vessel components
- Pressure vessel Internals
- Stainless steels, Ni-based alloys
- Piping, pumps, Valves
- Steam generator
- Steam water systems
- Turbine, Alternators
- Electrical equipments
- Fuel, control rod assembly
- Civil Engineering

THANK YOU FOR YOUR ATTENTION!

benoit.tanguy@cea.fr

Irradiation effects on microstructure

❑ Evolution of RIS with dose (MTR vs PWR)

- MTR: Saturation of segregation of around 5-10 dpa , Cr ~ 12%, Ni ~ 25%

- PWR: measurements on a Flux Thimble tube from PWR irradiated to 70 dpa
  → Segregation continues to change above 10 dpa with Cr ~ 8.5%, Ni ~ 30%

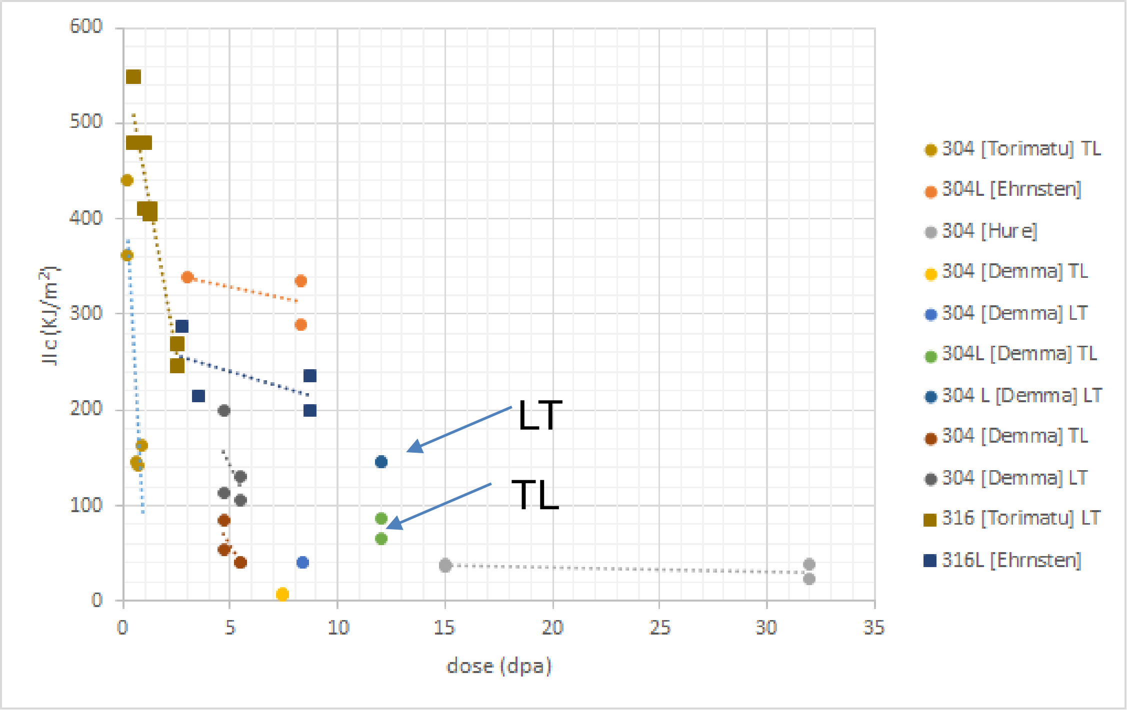
- Doubt on the representativity of irradiation in MTR versus irradiation in PWR ?
  → need of more data on steels irradiated at high dose in PWR
  → need to know the composition of the boundary before irradiation
    - ➢ RIS combine the segregation effects originating from manufacture (boundary Cr depletion in the HAZ, Cr or P enrichment due to quality treatment)
    - ➢ The original composition of the boundary is not generally known for materials irradiated in PWR

  → Difficult to provide a precise quantitative description of RIS between laboratories:
    - ❖ Segregation is inhomogeneous between boundaries or along a single boundary
    - ❖ Profile is extremely fine (+/- 5 nm)
    - ❖ Activity of the samples could lead to artefacts
    - ❖ Thin foil can be contaminated by Si after irradiation and specimen preparation

RIS behavior of minor elements (P, C, N, and B), all of which segregate at GB, is not well established because they are difficult to measure.

Irradiation effects on mechanical properties

❑ Evolution of Fracture toughness with dose (LWRs)



- ➢ Marked effect of specimen orientation (linked to metallurgical features)
- ➢ Same trend but large difference between similar grades -> link with microstructural features ?

Irradiation-creep

- **Primary state**: short-term transient state, generally poorly characterised

- **Secondary state**: stationary state that is retained until swelling appears on the material, and with plastic deformation proportional to the dose and the stress;

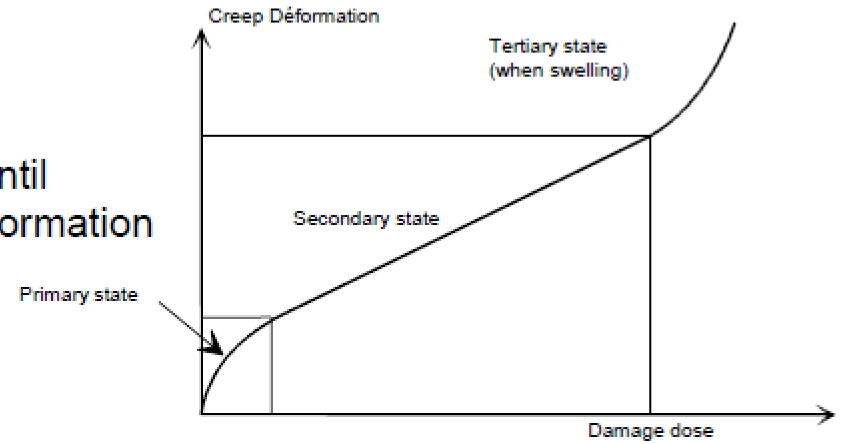
$$d\varepsilon_F = B_0 \cdot \sigma \cdot d\phi$$

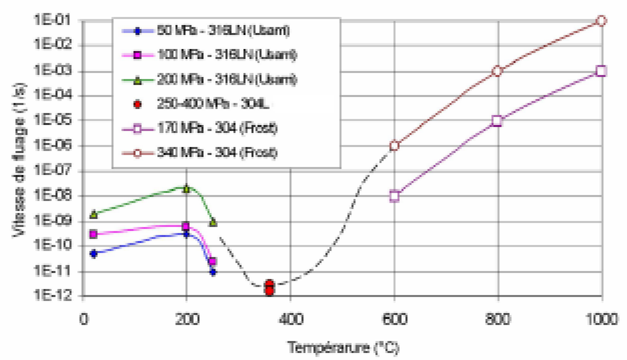
ε_F : creep deformation
ϕ : fluence
B_o : creep compliance

-**Tertiary state**: state for which there is a creep rate acceleration associated with the development of material swelling

$$d\varepsilon_F = (B_0 + D \cdot dV/d\phi) \cdot \sigma \cdot d\phi ,$$

$dV/d\phi$: instantaneous swelling rate





very low thermal creep for austenitic steels

Conclusions and perspectives

Future fields of investigation: Continue to obtain the building blocks

- e.g. Characterisation at the local scale: small specimens, in-situ measurements
- Refining the understanding and modelling of plasticity mechanisms and their evolution with irraddiation
- Building the link between microscale and mechanical properties

To gather building blocks…….