TEM INVESTIGATION OF ION AND NEUTRON IRRADIATED AUSTENITIC STAINLESS STEEL

TASK 2.2: FLUX EFFECTS ON INTERNALS

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Outline

- Introduction
- Objective and scope
- TEM examination of neutron irradiated material
- TEM examination of ion irradiated material
- TEM in-situ Post Irradiation Annealing
- Summary

Introduction

- Austenitic stainless steels: Fe based alloys used to fabricate internal components of nuclear LWR in operation.
- These internal components may suffer different problems when operating in the LWR environment, such as IASCC and swelling.
- IASCC is the most common degradation phenomenon of internals

→ It manifests as IG cracking
→ Complex phenomenon: radiation, materials, water environment and stress-strain are simultaneously involved.
→ Materials aspects may have different contributions and a relevant one is radiation damaged microstructure, i. e., the different radiation induced features produced by neutrons on austenitic stainless steels.

Objective and scope

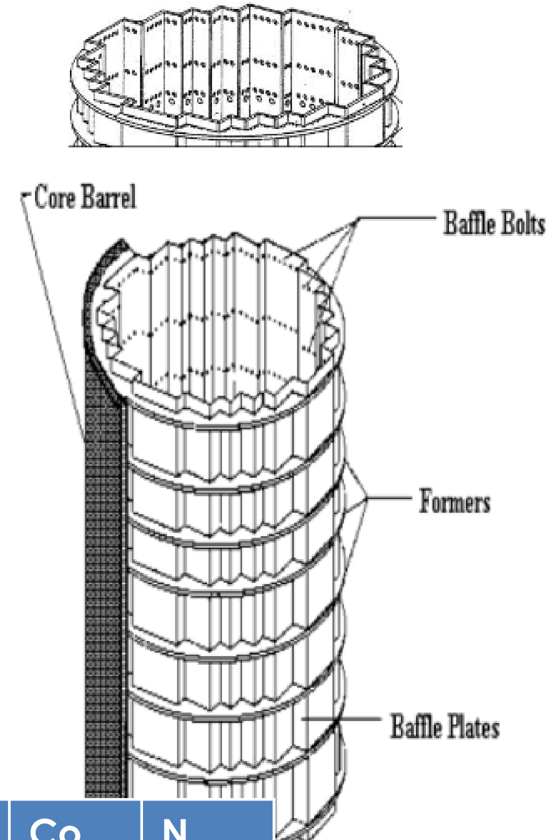
- This work is focused on alterations produced in the microstructure due to irradiation

  - TEM examination of available **neutron** irradiated austenitic stainless steel from a VVER reactor
    - Description of radiation damaged microstructure, i.e., radiation induced features
  - Same material has been ion irradiated at two different **ion** fluxes up to the same dose as the neutron irradiated material and examined by TEM
    - Emulate all or part of radiation induced features created by neutron irradiation
  - In situ TEM PIA experiments have been performed on **ion** irradiated material
    - To obtain information about stability of radiation induced features created by ion irradiation

- TEM examination of available **neutron** irradiated austenitic stainless steel from a VVER reactor
- Same material has been ion irradiated at two different **ion** fluxes up to the same dose as the neutron irradiated material and examined by TEM
- In situ TEM PIA experiments have been performed on **ion** irradiated material

Neutron irradiated material

- Material under study:
  - Materials designated for microscopic studies within the Soteria project are coming from the in-service irradiated internal components of the decommissioned VVER-440 Nord/Greifswald Unit 1.
  - Ti-stabilized austenitic steels - 08Ch18N10T, equivalent of AISI-321SS
  - Specimens for TEM investigation were fabricated from the reactor core internals material (RCI) of the **core barrel** (material AGS I) and from the material of the core basket (material AGS II), located in position near to the active zone center.



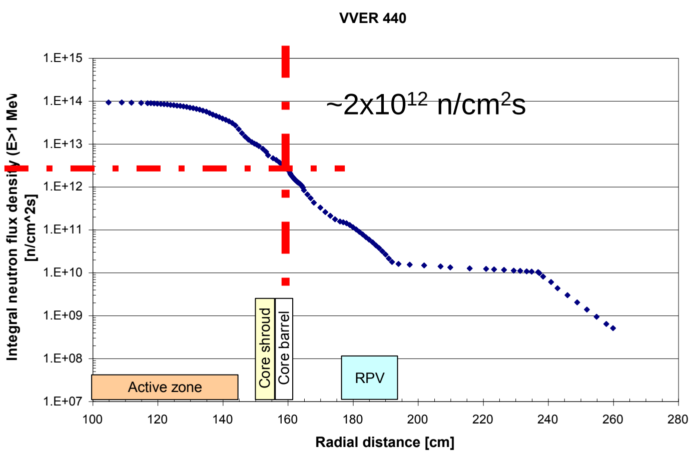
- Composition of 08Ch18N10T, equivalent of AISI-321SS

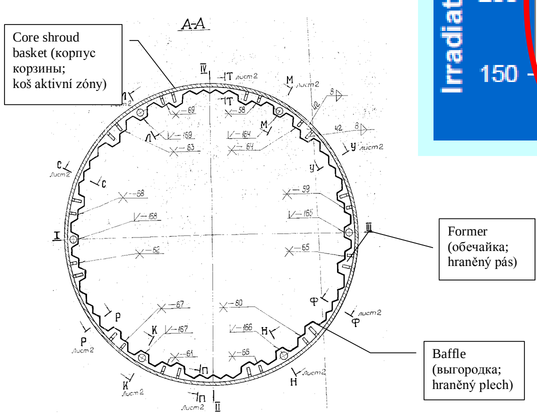
Fe	C	Mn	Si	P	S	Cr	Ni	Ti	Cu	Co	N
Bal.	0.07	1.41	0.46	0.011	0.007	18.18	9.72	0.49	0.03	0.02	0.013

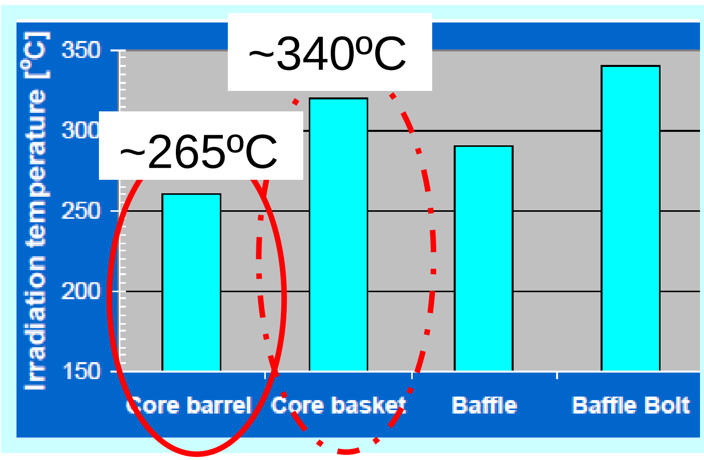
Irradiation conditions, in-service material

- The Greifswald Unit 1 was in service for 15 years
- Temperature at the core barrel: **265°C**
- Based on theoretical calculations the neutron flux ranges (E> 1 MeV) are:
  - 2.6 x 10^{17} n/m^{2}s, i.e. 4.4*10^{-8} dpa/s in position of RCI core baffle,
  - 6-9 x 10^{16} n/m^{2}s, i.e. 1-1.5*10^{-8} dpa/s in position of RCI core shroud / basket,
  - 1.7-4.6 x 10^{16} n/m^{2}s in position of RCI **core barrel (3-7.8x10^{-9} dpa/s)**.

Components 15 FPY in-service irradiated	2.4 dpa	5.2 dpa	11.4 dpa	11.4 dpa
	Core barrel	Core basket	Baffle	Baffle Bolt
	Plate 36 mm	Plate 32 mm	Sheet 8 mm	M12 x 29 mm
Microhardness	285 ± 33	363 ± 9	431 ± 72	431 ± 72







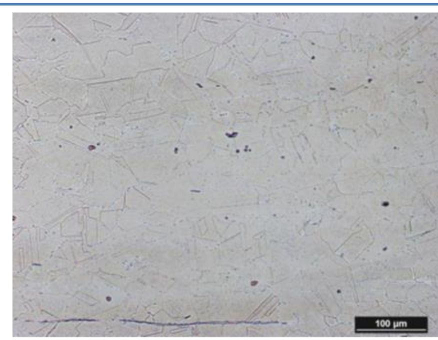
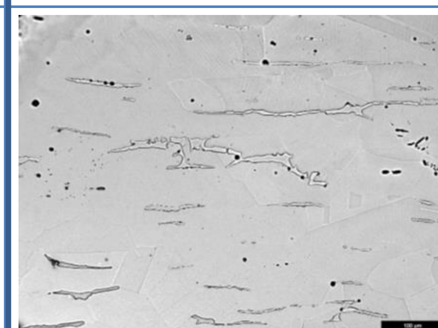
Section of WWER 440 internals and calculated integral neutron flux density in course of reactor core radial dimension.

TEM characterization of neutron irradiated material

- ❑ CIEMAT and CVR collaboration
- ❑ Microscope at CVR facilities:
  - TEM JEOL JEM 2200FS
  - Electron source: Field Emission Gun (FEG)
  - Accelerating voltage: 200 kV
  - In-column Omega filter
  - CTEM, STEM
  - HAADF STEM detector for Z-contrast imaging
  - EDX detector 80 mm^2
  - EELS

Sample preparation

- TEM specimens (foils) were prepared from the Ti-stabilized, type AISI 321SS.
- The steel slices were mechanically grounded and polished to the thickness about 80 μm by standard SiC papers.
- TEM disks, 3 mm in diameter, were punched out from the thin slice.
- Finally, the electro-polishing of thin foils at the voltage of 12 V with use of Fischionne twin-jet polisher was performed in 5 % perchloric acid and methanol solution cooled to temperature about –37°C.

Component	Core Basket	Core Barrel
Material	08Ch18N10T	08Ch18N10T
Grain size	~ 40 μm	~ 100 μm
Delta ferrite	1 %	4 %
Dose	5.2 dpa	2.4 dpa
Irradiation time	15 FPY	15 FPY
Irradiation Temperature	340 °C	264-299 °C
Initial microstructure		

Methodology of TEM examination of radiation induced features (RIF)

- Radiation damaged microstructure in austenitic stainless steels consists of different radiation induced features
- Each of them is detected employing different imaging or analysing methods

Common radiation induced features found in austenitic stainless steels after neutron irradiation

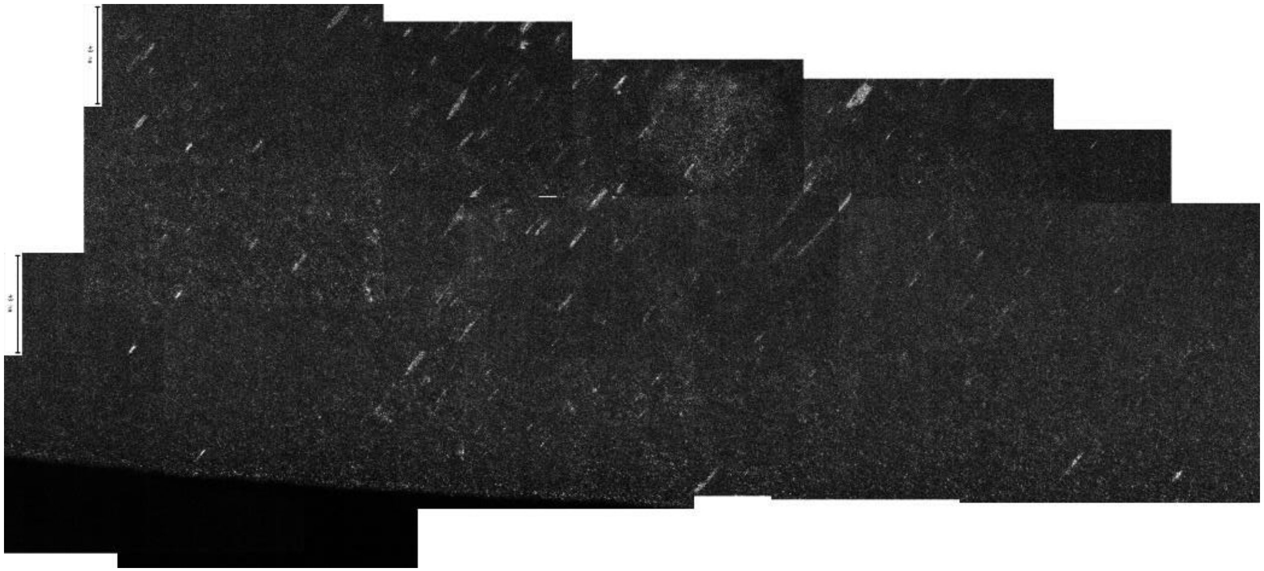
RIF	Detection method
Faulted loops (FL)	CDF rel-rod technique
Black dots and unfaulted, perfect loops	two beam BF and WBDF (CTEM, STEM)
Cavities	over- and under-focus images (CTEM), STEM, EELS analysis
Precipitates	CDF, EDX analysis
RIS to grain boundaries, dislocation lines, dislocation loops	EDX analysis

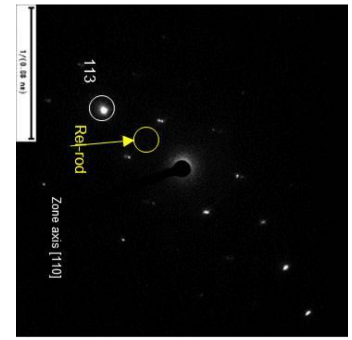
Quantification:

- Image analysis software **JMicrovision**: manual selection of features, counting and sizing

Core barrel 2.4 dpa, 265ºC, $3x10^{-9}$ dpa/s
Dislocation loop component: FL

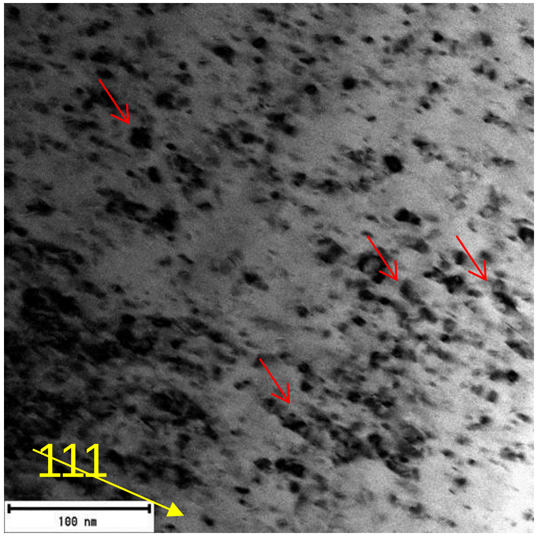
Core barrel 2.4 dpa, 265ºC, $3x10^{-9}$ dpa/s

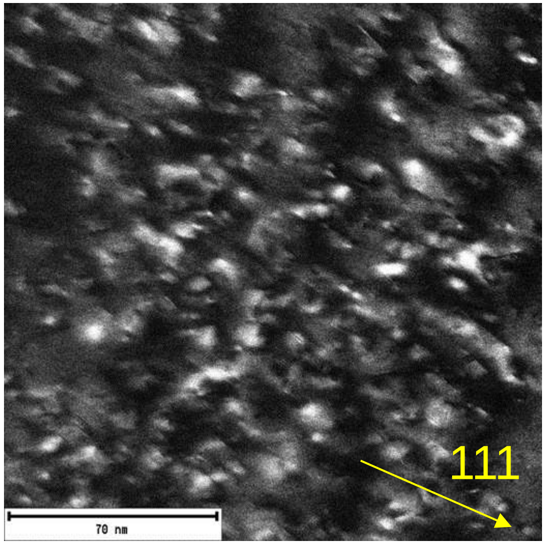




- Rel-rod, centered dark-field (CDF)
- Faulted dislocation loops (Frank loops) on {111} planes with Burgers vector 1/3[111] produce <111> satellite streaks near g=200
- Tilting ~8° away from the [011] zone axis along the [113] reflection allows one variant to be imaged with enhanced contrast in centered dark field,

Core barrel 2.4 dpa, 265°C, 3x10^{-9} dpa/s
Dislocation loop component: black dots and perfect loops



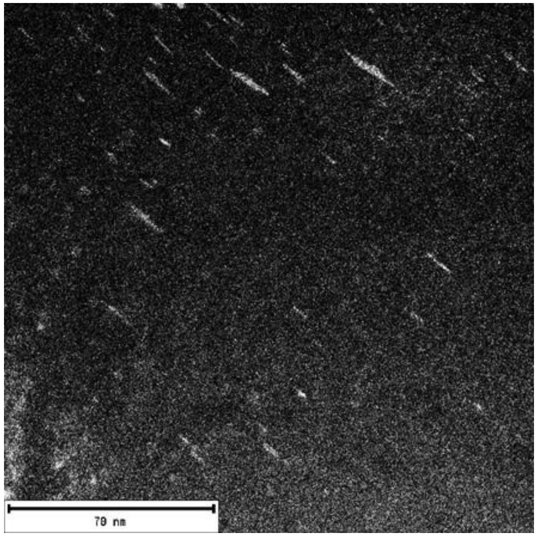


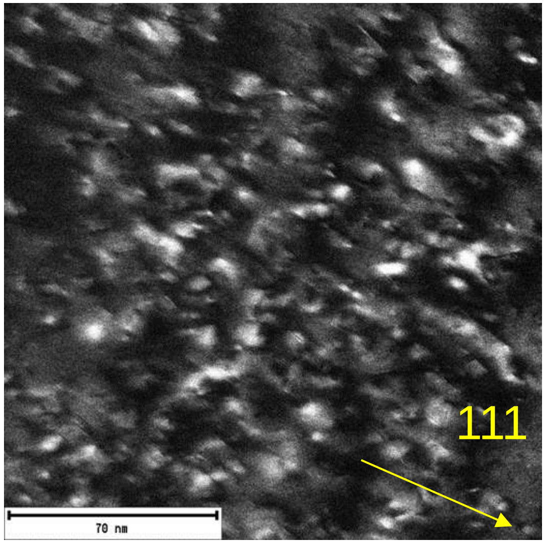
Two-beam conditions (exciting g type 111 or 200, at [110] zone axis)

- BF and WBDF modes reveal the presence of other types of defects:
  - → a finer microstructure (black dots),
  - → large loops (perfect and faulted loops).
- Smaller defects (black dots) are more difficult to detect in BF images
- → WBDF images for counting and sizing fine microstructure

- There is still the question about the nature of black dots:
  - Small FL?
  - Vacancy loops?

Dislocation components: Nature of black dots?

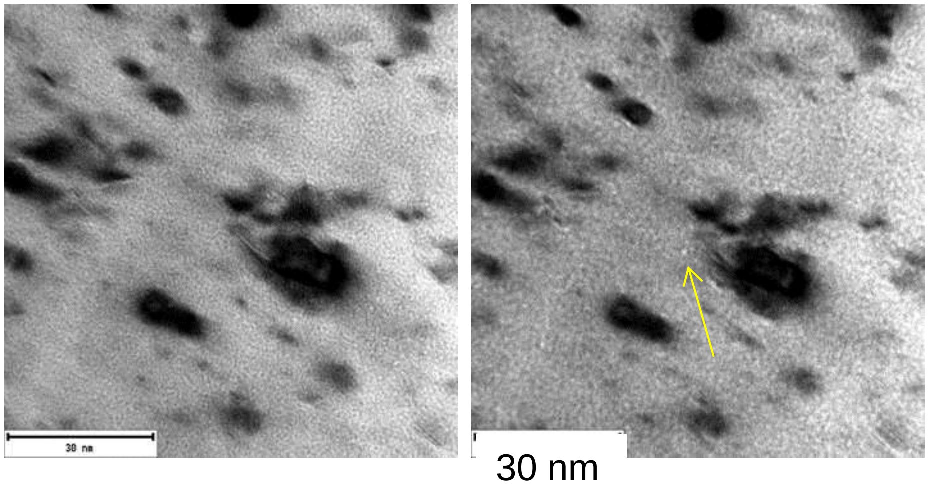




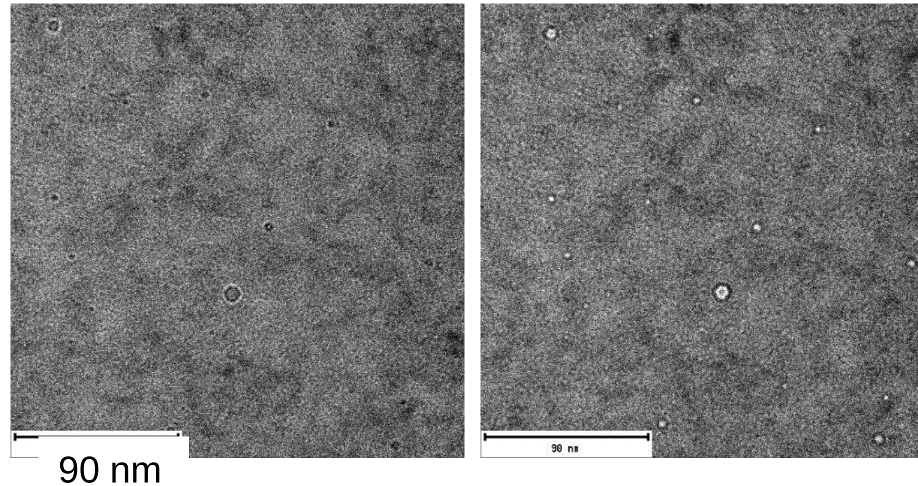
- Black dots=small FL?
  - Comparing WBDF images and CDF rel-rod images where only FL are observed
    - Coincidence is not found: positions in both types of images did not match
    - Need to have images of all the FL families to draw a conclusion
  - Comparing size distributions: population of objects of small size

Core barrel 2.4 dpa, 265ºC, ~10^{-9} dpa/s
Cavities

Core barrel 2.4 dpa, 265ºC, ~10^{-9} dpa/s



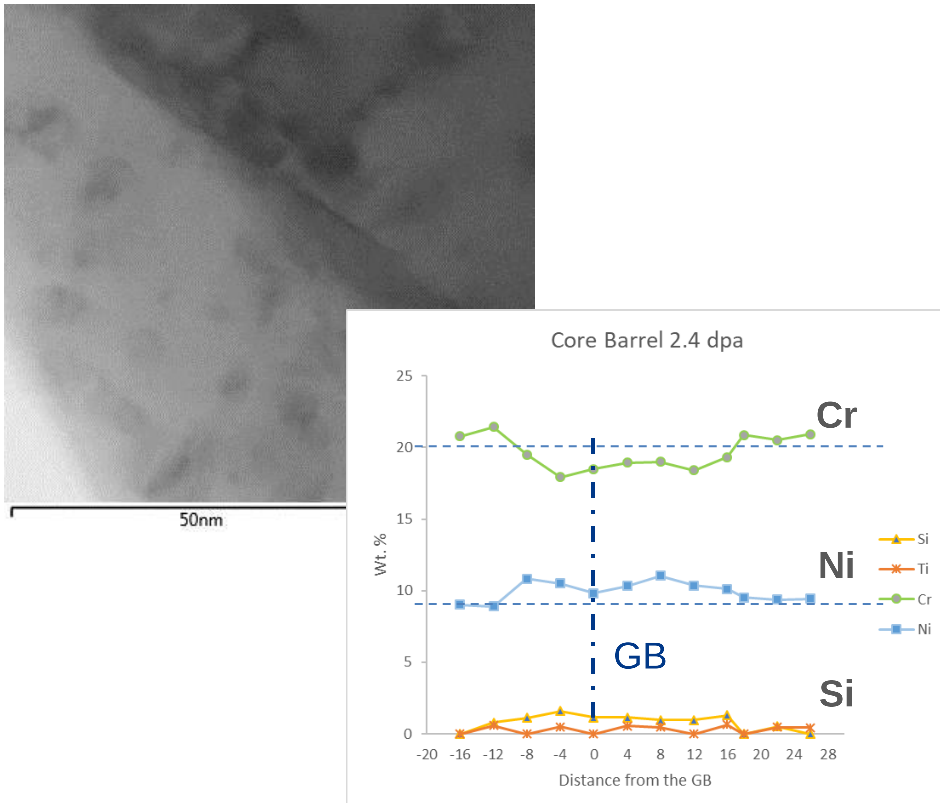
Core basket 5.2 dpa, 325ºC, ~10^{-8} dpa/s



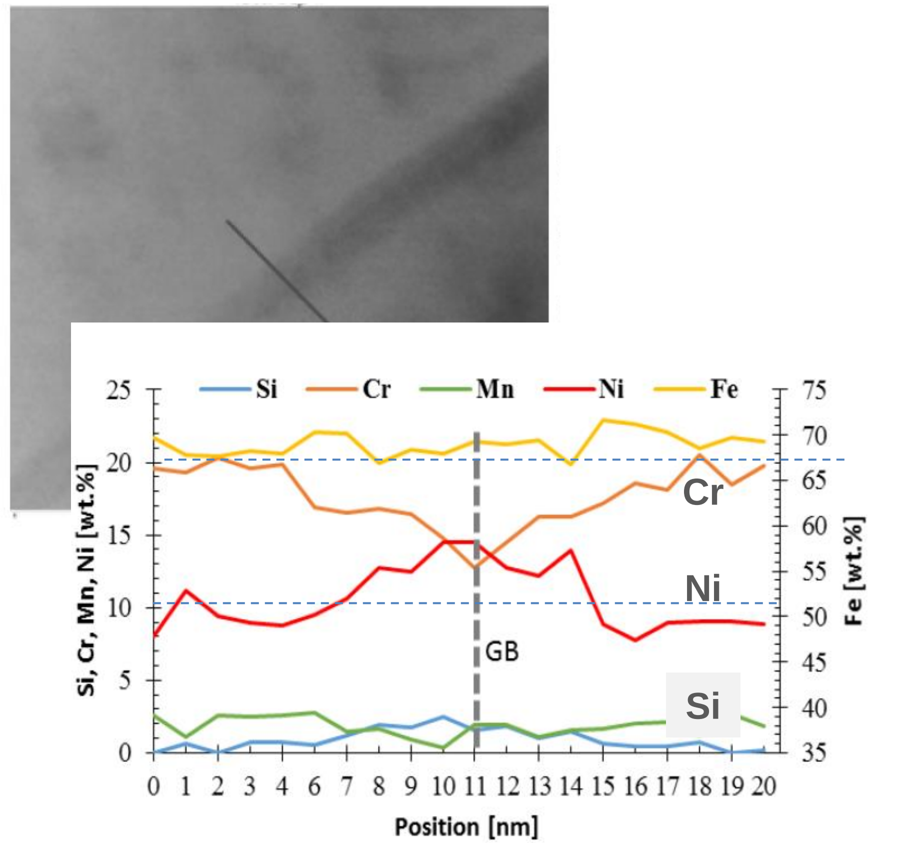
- The presence of cavities was evaluated using over and underfocus technique
- Core barrel: cavities were hardly observed, very small size
- Core basket:
  - clearly observed, 2-4 nm
  - Using EELS, it was confirmed the presence of a gas (He and/or H) in the cavities

Core barrel 2.4 dpa, 265ºC, ~10^{-9} dpa/s
Microchemistry: RIS at GB

Core barrel 2.4 dpa, 265ºC, ~10^{-9} dpa/s



Core basket 5.2 dpa, 325ºC, ~10^{-8} dpa/s



- Grain boundary areas were investigated with focus on local chemical changes.
- Radiation-induced segregation of Cr, Ni, Si and Fe was observed across high-angle grain boundary using STEM EDS.
- The segregation of elements (depletion of Cr and enrichment of Ni, Si) is taking place in a region of about 20 nm.

Objective and scope

- TEM examination of available neutron irradiated austenitic stainless steel from a WWER reactor
- Same material has been ion irradiated at two different **ion** fluxes up to the same dose as the neutron irradiated material and examined by TEM
- In situ TEM PIA experiments have been performed on ion irradiated material

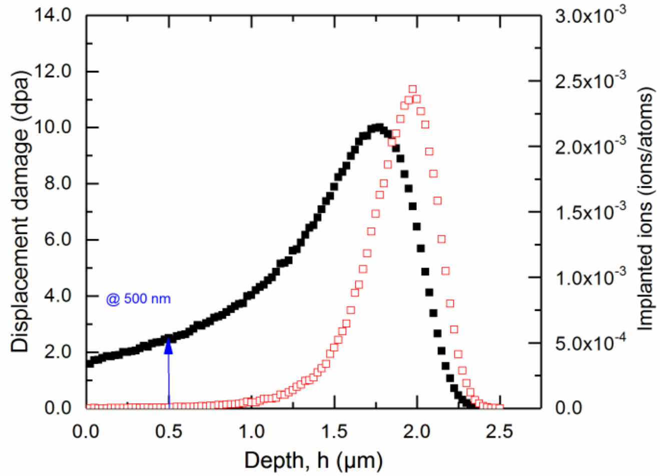
Irradiation conditions: Ions

- Austenitic stainless steel 08Ch18N10T (AISI **321SS**) ion irradiated at the **IBC, HZDR (Germany)**
- **Fe ions of 8 MeV**
- **2.5 dpa** (1.04×10^{16} ions/cm^2) at 500 nm from surface
- Two irradiation experiments.
- Average damage rates were :
  - **HF: 4.6×10^{-4} dpa/s**
  - **LF: 1.5×10^{-5} dpa/s**

x30

- Irradiation temperature: **350°C**

8 MeV Fe ions



Radiation damage and ion implantation profiles as functions of sample depth calculated from SRIM

Ion irradiation

- Ion irradiation aims at reproducing microstructural damage produced by neutrons
- The set of conditions suitable for emulating all neutron irradiation features in a single ion irradiation experiment is not known
- Approach:
  - To fix the dose to be the same as for neutron irradiation
  - As the ion irradiation is at higher flux, the irradiation temperature can be adjusted to achieve:
    - the same total flow of defects to sinks (which drives processes like RIS)
    - or the net flux of one defect over another (which drives processes like cavity or loop growth).
  - The magnitude of T shift is usually estimated using Mansur's invariant relations and is different for these two processes (L. Mansur, JNM, 1993)

T shift

	Neutrons	Ions LF	Ions HF
dose	2.4 dpa	2.5 dpa at 500 nm	2.5 dpa at 500 nm
Irrad Temperature	**265ºC**	**350ºC**	**350ºC**
Flux	**$3x10^{-9}$ dpa/s**	$1.5x10^{-5}$ dpa/s	$4.6x10^{-4}$ dpa/s
T_{shift} for RIS		T_{shift}=264 ºC →T_2= 529ºC	T_{shift} = 460ºC →T_2=725ºC
T_{shift} for loops and cavities		T_{shift}= 81ºC → **T_2= 346ºC**	T_{shift} = 121ºC →**T_2=386ºC**

- T shift according to L. Mansur, JNM1993
  - Vacancy formation energy Evf=1.5 eV and Vacancy migration energy Evm= 1.2 eV

RIS

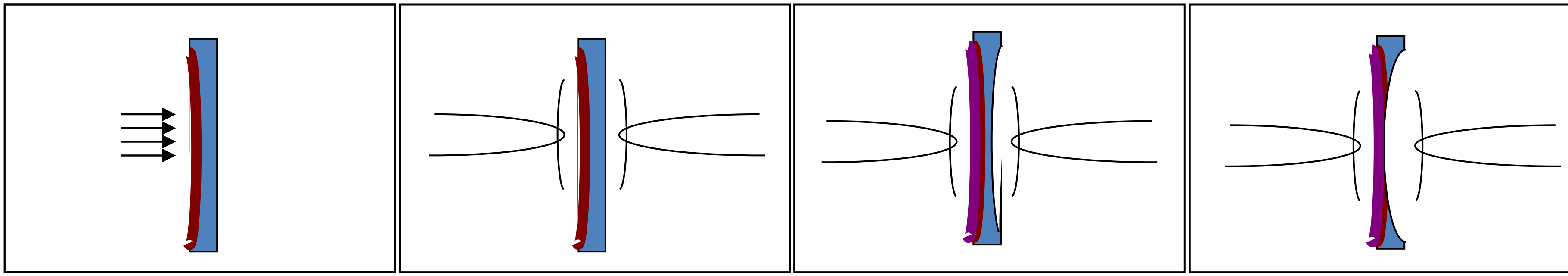
$$T_2 - T_1 = \frac{\frac{kT_1^2}{E_m^v}\ln\left(\frac{K_{0_2}}{K_{0_1}}\right)}{1 - \frac{kT_1}{E_m^v}\ln\left(\frac{K_{0_2}}{K_{0_1}}\right)}$$

Loops and cavities

$$T_2 - T_1 = \frac{\frac{kT_1^2}{E_m^v+2E_f^v}\ln\left(\frac{K_{0_2}}{K_{0_1}}\right)}{1 - \frac{kT_1}{E_m^v+2E_f^v}\ln\left(\frac{K_{0_2}}{K_{0_1}}\right)}$$

Results ion irradiation

- Sample preparation for TEM when extracting samples from ion irradiated material
  - TEM samples are 3 mm diameter discs with initial thickness of 100 µm
  - Ion irradiation on one side



(1) Ion beam | **(2) 3’ electropolishing** | **(3) Protection with lacquer and back thinning by electropolishing** | **(4) Electropolishing until perforation**

Ion damaged area

Lacquer layer to protect the ion damaged area

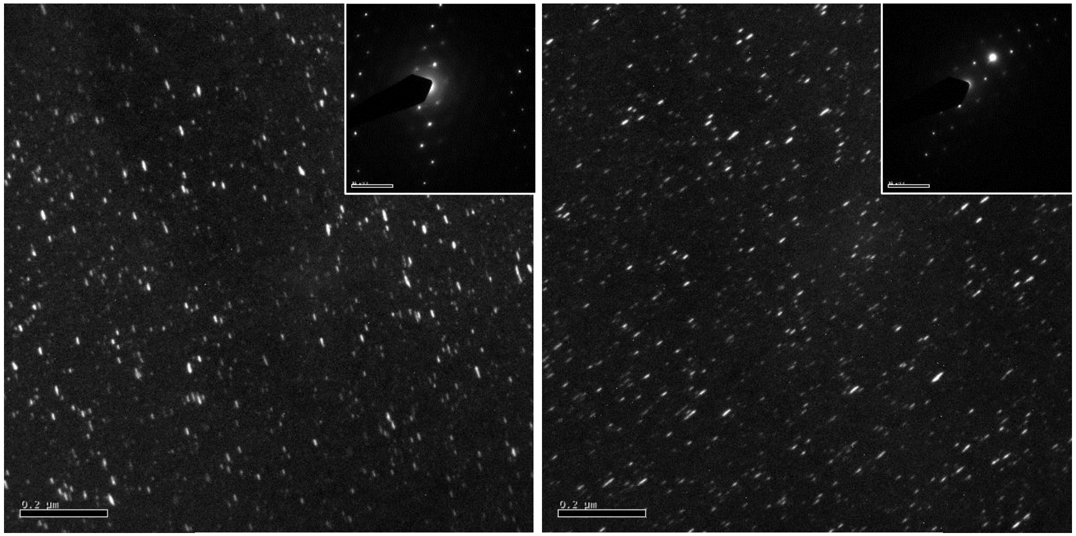
TEM characterization of ion irradiated material

- Microscope at CVR facilities:
  - TEM JEOL JEM 2200FS
  - Electron source: Field Emission Gun (FEG)
  - Accelerating voltage: 200 kV
  - In-column Omega filter
  - CTEM, STEM
  - HAADF STEM detector for Z-contrast imaging
  - EDX detector 80 mm2
  - EELS
  - Gatan double-tilt heating holder

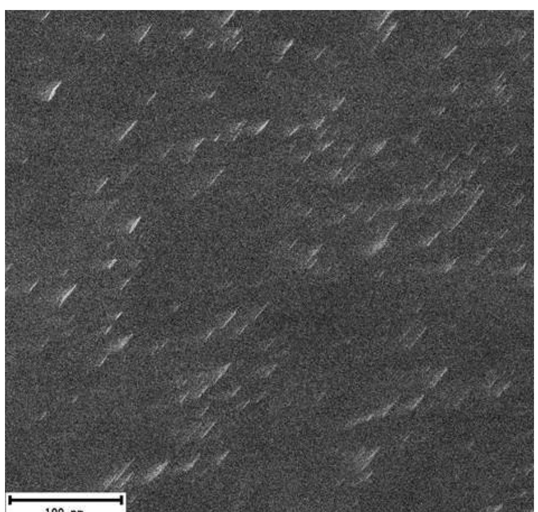
- Microscope at CIEMAT facilities
  - TEM JEOL JEM 2010
  - LaB6 filament
  - Accelerating voltaje: 200 kV
  - CTEM
  - EDX detector 80 mm2
  - Gatan double tilt heating holder
  - Video recording

Ion irradiation : 2.5 dpa, 350°C
Dislocation loop component: FL

LF: 2.5 dpa, 350°C, 1.5×10^{-5} dpa/s



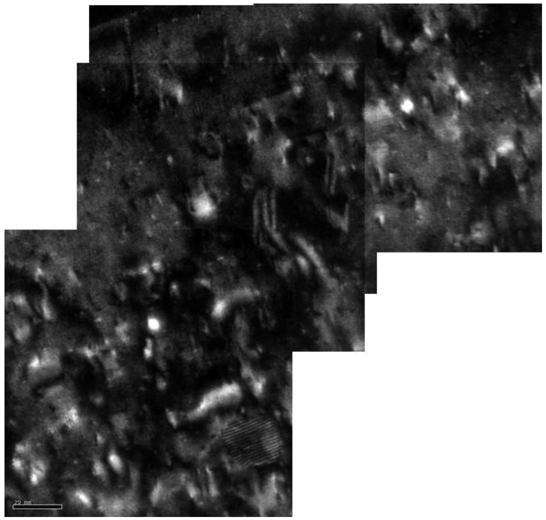
HF: 2.5 dpa, 350°C, 4.6×10^{-4} dpa/s



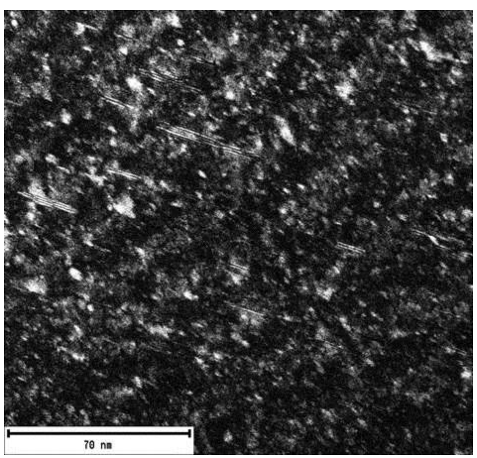
Increase in size
and density

Ion irradiation : 2.5 dpa, 350°C
Dislocation loop component: fine microstructure

LF: 2.5 dpa, 350°C, 1.5×10^{-5} dpa/s



HF: 2.5 dpa, 350°C, 4.6×10^{-4} dpa/s



Decrease in size
Increase in density

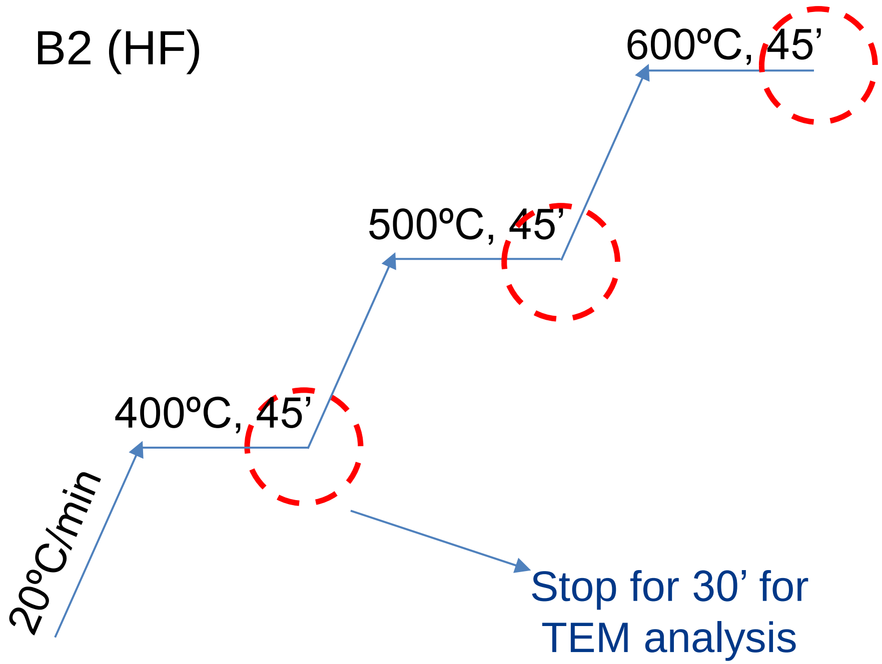
- Two beam imaging condition with g type (111) excited:

- TEM examination of available neutron irradiated austenitic stainless steel from a WWER reactor
- Same material has been ion irradiated at two different ion fluxes up to the same dose as the neutron irradiated material and examined by TEM
- In situ TEM PIA experiments have been performed on **ion** irradiated material

TEM: in-situ annealing experiments

- FL and fine microstructure are present on ion and neutron irradiated material
- The stability of these features detected after ion irradiation has been studied by PIA experiments
- We may also apply other types of studies (corrosion, SCC tests, etc.): isolate the effect of some features on IASCC.

TEM: in-situ annealing experiments



- The experiments were performed both at CIEMAT and at CVR with a double tilt-annealing sample-holder.
- Possible to heat up to 1000ºC under the high vacuum of the TEM
- The evolution of the microstructure during the annealing was directly observed by employing diffraction contrast: bright field (BF) and weak beam dark field (WBDF) images.
- The experiments were video recorded: interpretation in course, will be reported in D2.5, mid-July, 2019.

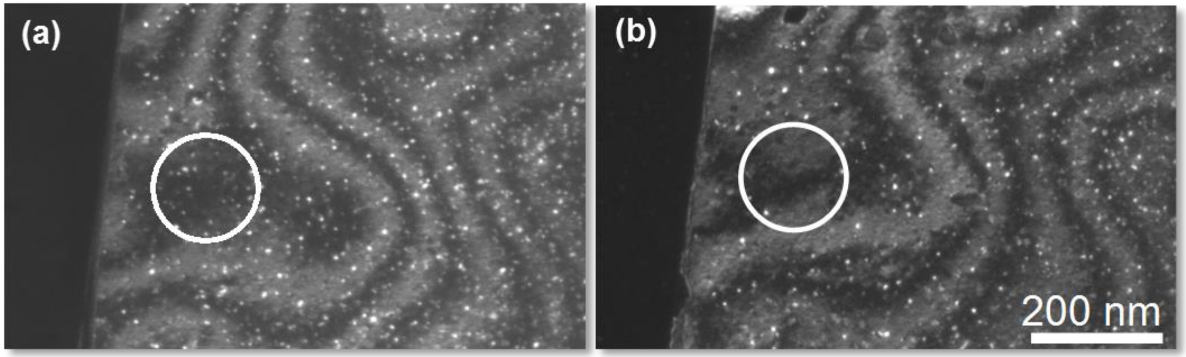
Annealing effect on population of fine microstructure

- Present results are still under interpretation will be reported in D2.5
- Results shown in next slide come from previous experiment: 316L ion irradiated

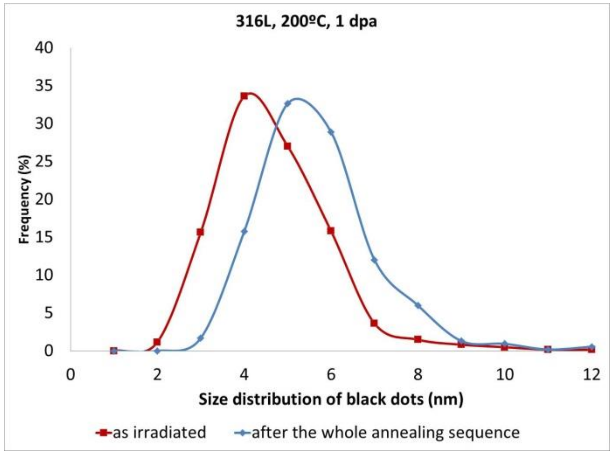
Previous experiments : **Evolution of the 316L steel irradiated microstructure at 200ºC, 1 dpa, during in-situ annealing**

initial as-irradiated state

the microstructure at the end of the whole annealing sequence, final temperature 450ºC



Loops appearing at the interior of the circle disappear after the annealing.



Size distribution of the black dots in the 316 steel as-irradiated at 200ºC/1dpa and after the whole annealing sequence.

- Disappearance of defects was observed to occur as a consequence of the annealing
- Changes in density or in size are not easy to follow during the experiments, but it is possible to provide information at the different steps of the annealing from the analysis of the video frames.
- A slight increase of the average size is observed and the size distribution is shifted towards larger sizes →the increase in size is due to the preferential disappearance of the smaller defects or black dots and not to actual growth.
- *"Since the black dots showed slightly faster recovery during PIA than the dislocation loops, the black dots may contain some fraction of thermal unstable clusters, which migth be vacancy type"* (Fukuya et al.)

Implication for IASCC

- By PIA, the stability of specific microstructures can be assessed
- Together with mechanical property tests and corrosion tests it may be possible to correlate IASCC with a specific microstructure.
- From literature:
  - *(…) Cracking susceptibility was removed during PIA before any significant change in RIS, dislocation loop microstructure or hardening occurred.*
  - *None of the defects were the sole controling factor in IASCC and that some other irradiation induced change may be responsible (…)*"
    → **fine scale radiation damage** as a contributing factor.
  - *(…) The rapid removal of a dense population of small obstacles impeding dislocation motion could result in a change in deformation mode (…).*

- J. Busby, G. S. Was, E. A. Kenik. "*Isolating the effect of radiation-induced segregation in irradiation-assisted stress corrosion cracking of austenitic stainless steels*" Journal of Nuclear Materials. 302 (2002)
- K. FUKUYA, M. NAKANO, K. FUJII, T. TORIMARU, Y. KITSUNAI, "*Separation of Microstructural and Microchemical Effects in Irradiation Assisted Stress Corrosion Cracking using Post-irradiation Annealing*" Journal of NUCLEAR SCIENCE and TECHNOLOGY, Vol. 41, No. 12, p. 1218–1227 (December 2004)

Summary

- Type AISI 321SS has been examined by TEM after neutron and ion irradiation
- Neutron irradiation at 2.4 dpa, 265°C, 3x10-9 dpa/s: FL, black dots perfect loops, cavities and RIS to GB has been detected
- Ion irradiation up to the same dose, 2.5 dpa, 350°C and two ion fluxes, 1.5x10-5 dpa/s and 4.6×10^{-4} dpa/s
  - Formation of FL, black dots and perfect loops
  - No cavities, no RIS
- Part of neutron radiation induced features has been reproduced with ion irradiation
- PIA: in situ annealing experiments,
  - employed to assess radiation induced features stability under annealing
  - May provide information about the nature of fine radiation induced microstructure

Acknowledgments

- The ion irradiations have been performed at the IBC, at HZDR, with support from C. Heintze and S. Akhmadaliev
- The preparation of ion irradiated samples by M. González and neutron irradiated samples by CVR staff.

The SOTERIA Consortium

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