Ion irradiation effects including high levels of helium and hydrogen on void swelling of 8-12%Cr ferritic-martensitic steels

Y. Kupriiyanova, V. Bryk, O. Borodin, A. Kalchenko, V. Voyevodin, G. Tolstolutskaya*

**voyev@kipt.kharkov.ua*

National Science Centre Kharkov Institute of Physics and Technology, 1, Akademicheskaya St., Kharkov, 61108, Ukraine

F.A. Garner

Radiation effects Consulting Richland, WA 99354, USA

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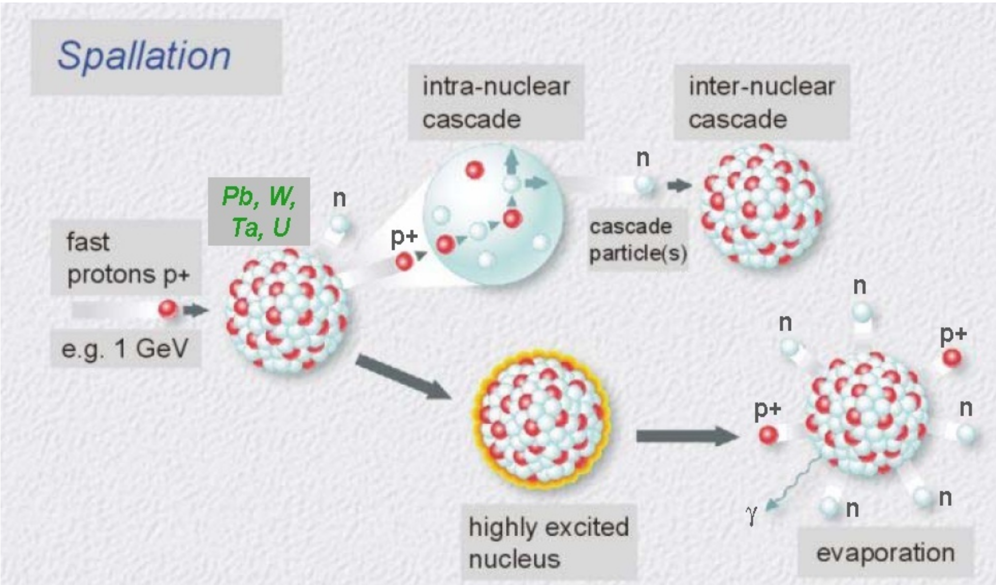
Motivation

- **Under the SNS environment, damage may be very high arising from the combination of very high energy protons (1GeV) and neutrons with energies ranging from the initial proton energy down to thermal energies.**
- **Structural materials exposed to mixed spectra of protons and neutrons found in accelerator-driven spallation neutron sources must withstand intensive generation and retention of large levels of helium and hydrogen.**
- **Is swelling of FM steels sensitive to these gases?**
- **Is there a common influence of helium and hydrogen?**
- **What is the effect of initial structure on swelling?**

Processes involved in “spallation”- ADS



Several classes of structural alloys find applications in more than one system. These include *austenitic stainless steels* for near-term fusion application, spallation neutron sources and several Generation IV fission reactor concepts, and *ferritic-martensitic steels* for long-term fusion systems, for advanced fission reactors and for long-term spallation neutron sources used for transmutation.

L.K. Mansur et.al.
Journal of Nuclear Materials
296 (2001) 1–16

	dpa/yr	appm He/yr	appm H/yr	He/dpa	H/dpa
Neutron	23	130	1800	5.7	78
Proton	13	1260	17 000	97	1310
Total	36	1390	18 800	39	522
p/n	0.57	9.7	9.4	17	16.8

Results calculated for displacement damage, He and H production rates at the innermost shell of the four-walled target container and shroud assembly. This is the wall of the assembly in contact with the bulk mercury on its inner surface and with the coolant mercury on the outer surface. The center of the nose is the location where radiation damage is highest, corresponding to peak proton beam flux of approximately 0.22 A/m^2

OUTLINE

1. Materials & Technical Methodology.

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3. Comparison of swelling characteristics under triple-ion beam irradiation in ferritic-martensitic (EP-450) and martensitic (EK181, F82H) steels.

Technical Methodology

For determination of the swelling characteristics of irradiated materials the following methods were employed:

- ✓ **Single, dual & triple beam ion irradiation;**
- ✓ **Transmission electron microscopy (TEM);**
- ✓ **X-ray micro-analyzer for determination of chemical composition;**
- ✓ **Scanning electron microscopy (SEM).**

Facility for triple-ion irradiation

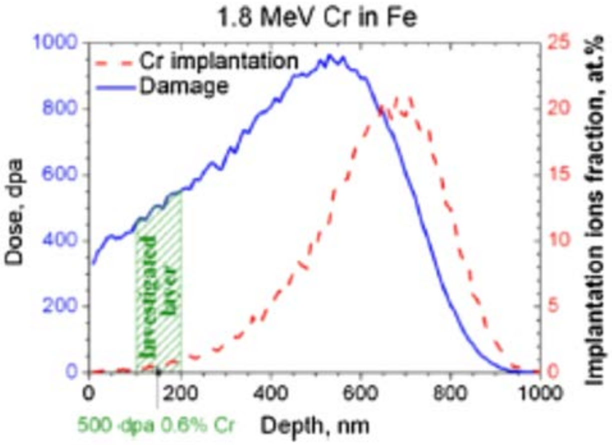
Electrostatic Accelerator with External Injector (ESUVI)

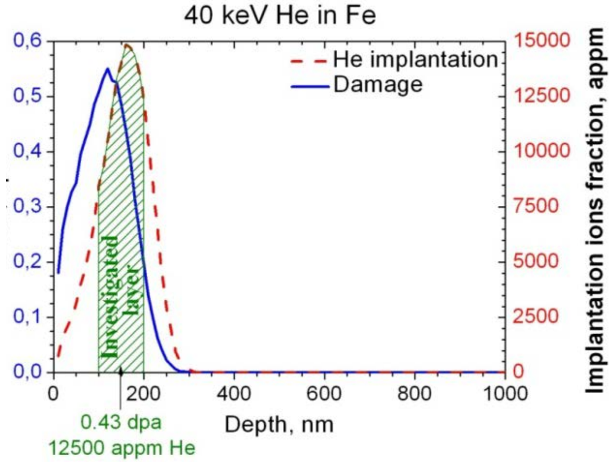
This facility for triple beam irradiation was first presented at the IWSMT-7,Thun, 2005

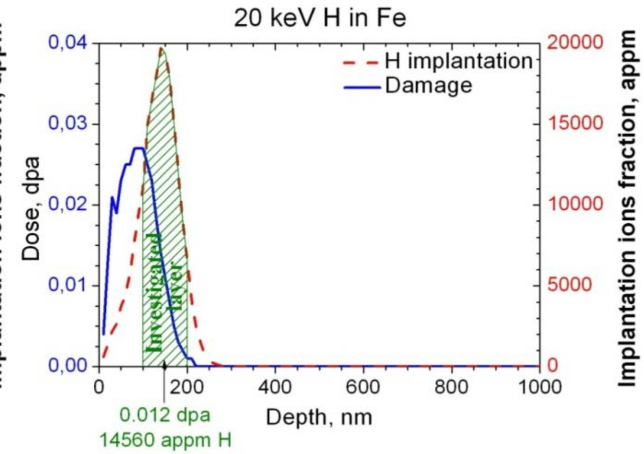
Irradiation	Parameters	
Cr Cr+He Cr+H Cr+He+H	E_{Cr}= 0.3 – 1,8 MeV j_{Cr} =1 - 35 μA/cm^2 E_{He}= 10 - 60 keV j_{He} = 1 – 500 nA /cm^2 E_H= 10 - 60 keV j_H = 1 – 500 nA /cm^2	T_{irr}= 350^0 – 800^{0}C D = 0 - 1200 dpa k = 7·10^{-5} - 10^{-2} dpa/s 0.1 – 1.3 appmHe/s 0.1 – 1.0 appmH/s

Profiles of displacement damage of Cr ions and ions of He & H

Large amount of helium & hydrogen can be implanted without adding much damage







For calculation of dpa the NRT-standard and "Quick damage" **option was used (**E_d=40eV**).**

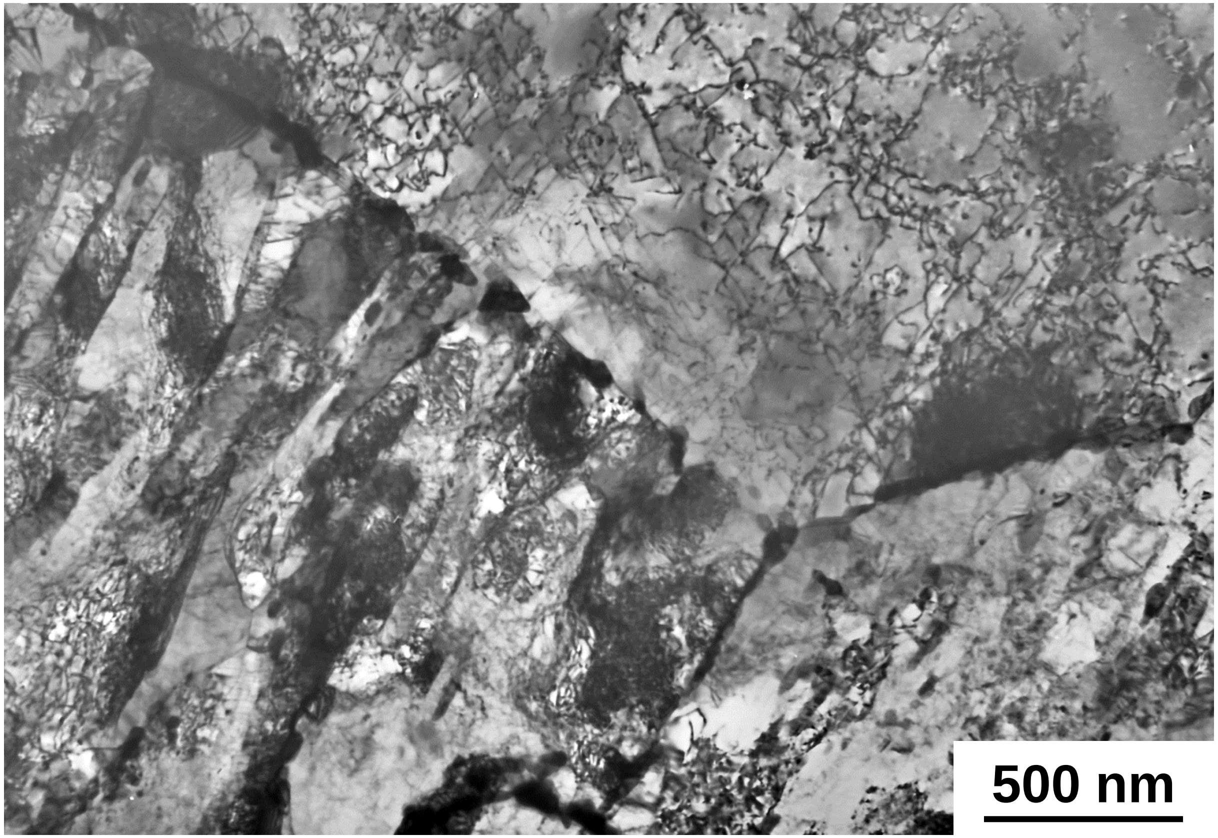
Investigated materials

Steel structure	Initial precipitates	Chemical composition, %wt									
		C	Cr	W	Si	Mn	Mo	V	Nb	Ni	Other
EP-450 ferrite+ tempered martensite 1:1	$M_2(C,N)$; $M_{23}C_6$	0.10-0.15	11.0-13.5	-	0.6	0.6	1.2-1.8	0.1-0.3	0.25-0.55	0.30	B-0.004
EK-181 tempered martensite	$M_{23}C_6$; M_6C; M_3C; TaC; VC	0.14	11.2	1.17	0.37	0.94	0.04	0.29	0.01	0.03	P-0.010, B-0.006, Ta- 0.17, Co-0.01, N-0.04
F82H martensite	$M_{23}C_6$; M_6C	0.10	7.44	2.0	0.14	0.5	-	0.20	-	-	Ta-0.04, N-0.002, P-0.001, S-0.001, Al-0.019

Steel	Heat treatment conditions
EP-450	$1050^0C/30min+720^0C/1h$
EK-181	$1100^0C/1h+720^0C/30$ min
F82H	$1040^0C/30min+740^0C/2h$

Initial structure of materials

EP- 450 steel, 12%Cr

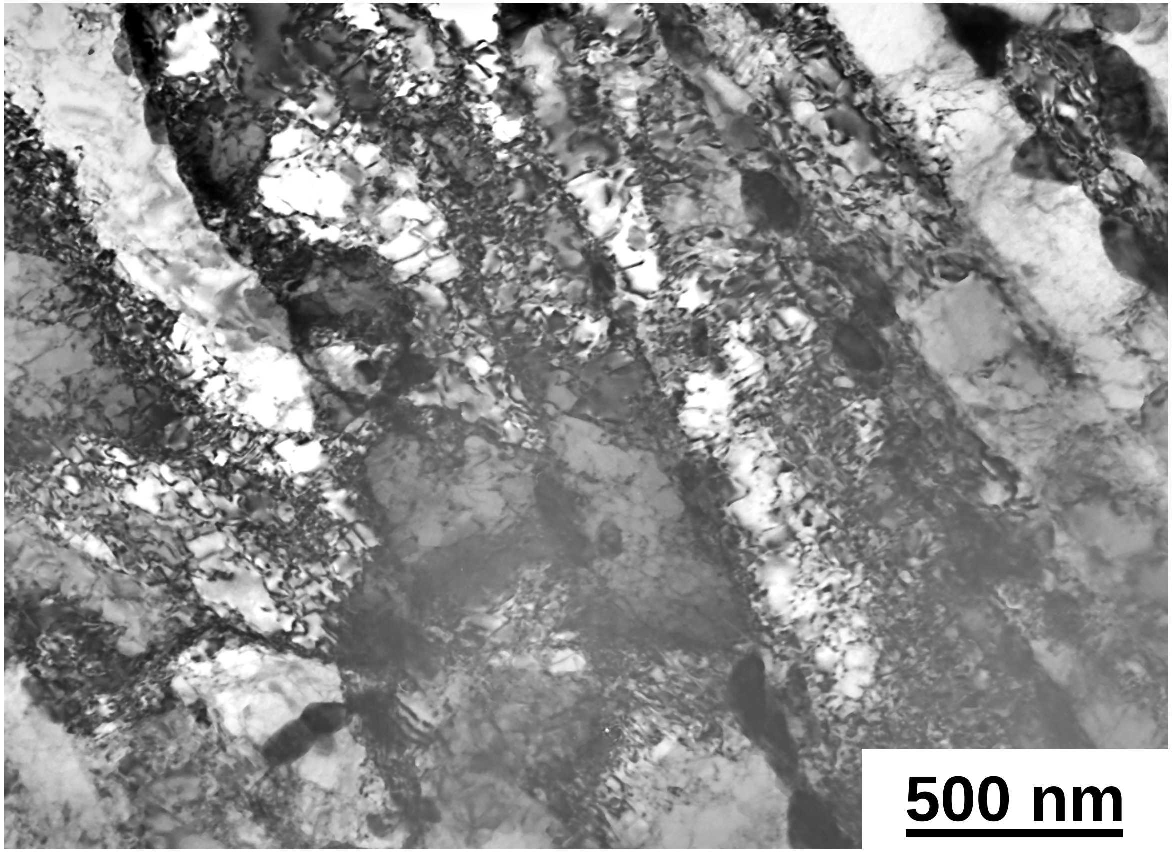


Duplex structure of tempered martensite and ferrite (1:1)

Large globular $M_{23}C_6$ carbides on grain boundaries and some $M_2(C,N)$ carbides in martensite matrix .

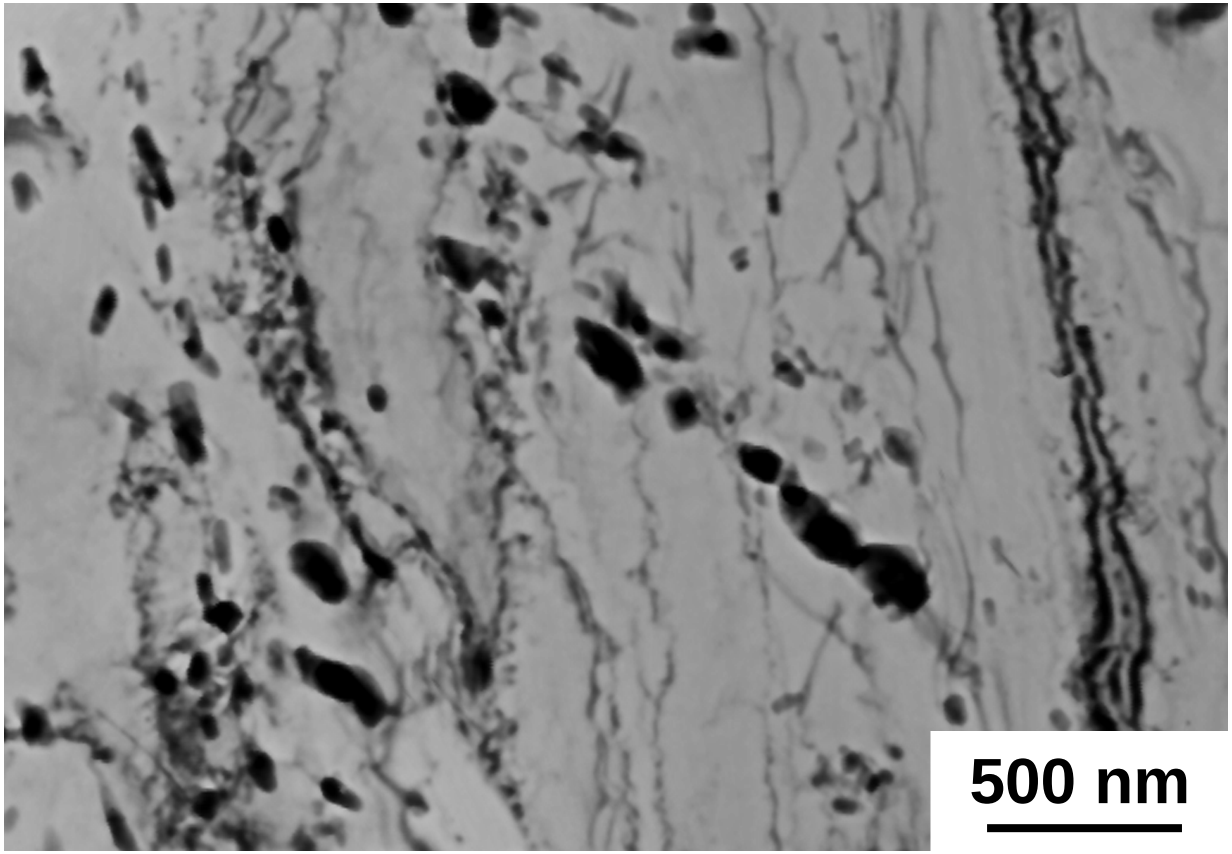
Initial structure of materials

EK-181, 12%Cr



Structure is lath tempered martensite + <3% residual austenite.
The average size of martensite grains is 250 nm.
Carbides are different in composition and size: $M_{23}C_6$; M_6C, M_3C, TaC, VC.
VC (TaC) and $M_{23}C_6$ size of 3-5 nm are an essential part of carbide phases.

Initial structure of materials
F82H, 8%Cr



Structure is martensite. Dislocation line density of ~ $1x10^{14}$ m^{-2}.
$M_{23}C_6$ carbides in matrix and on grain boundaries, at $6{\cdot}10^{19}$ m^{-3}, avg. size 73 nm.
Only a few MC carbides were observed in the matrix at $1{\cdot}10^{20}$ m^{-3} and 14 nm.
The mean width of the lath structure was 440 nm.

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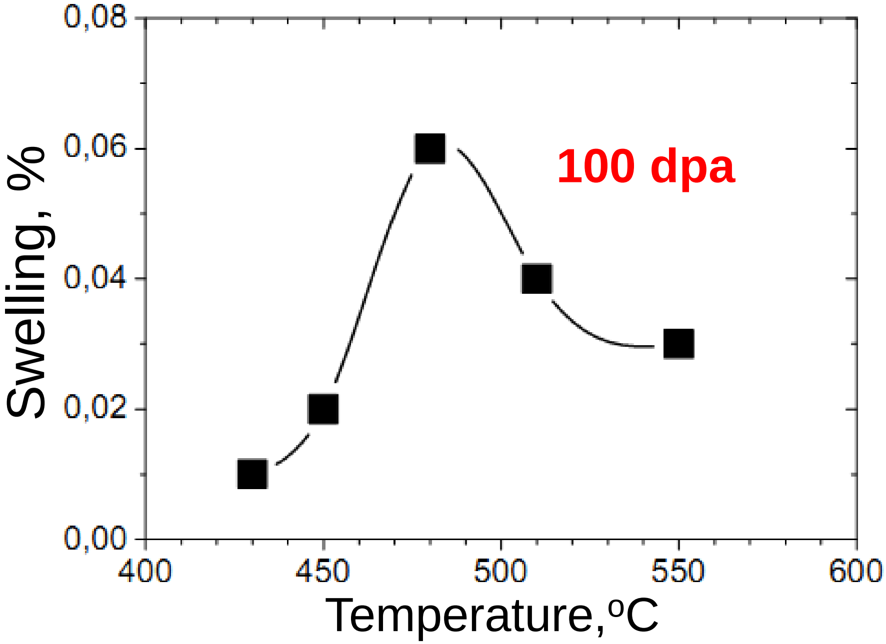
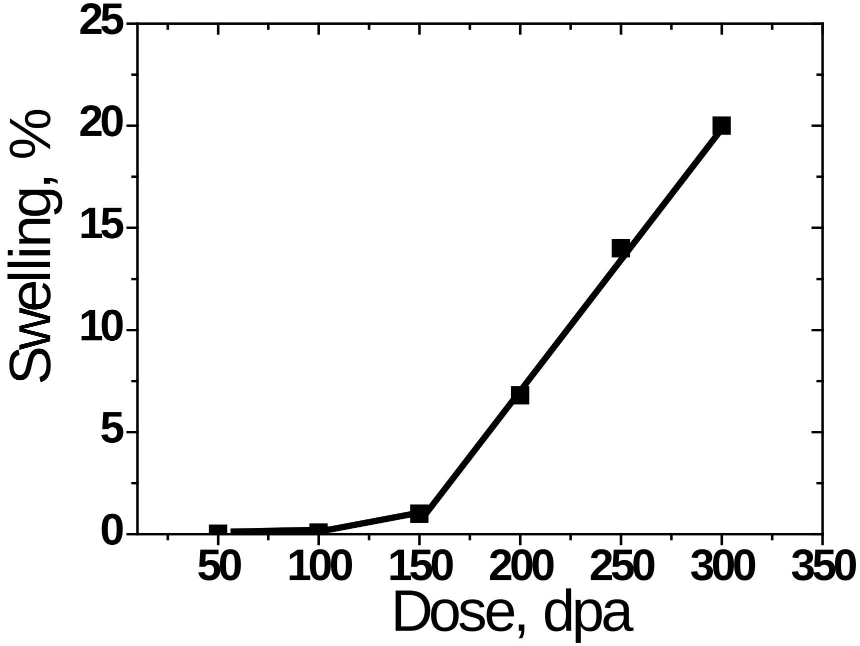
Temperature and dose dependence of swelling
EP-450 steel





The swelling maximum is at T=480°C in EP-450 steel .

O. Borodin et. al. PAST (Problem of Atomic Science and Technologie), 2011№2(97).p.10-15

Dose dependence of swelling in EP-450 at 480°C
Swelling increasing strongly was observed above 150 dpa.

Parameters of gas effect experiments

1.The single, dual and triple beam irradiations of EP-450, EK-181 and F82H were carried out at temperature of swelling maximum:

EP-450 $T_{max} = 480^0C$

F82H $T_{max} = 450^0C$

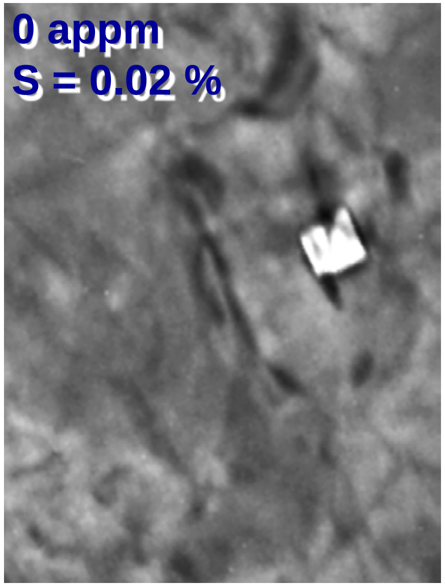
EK-181 $T_{max} = 450^0C$

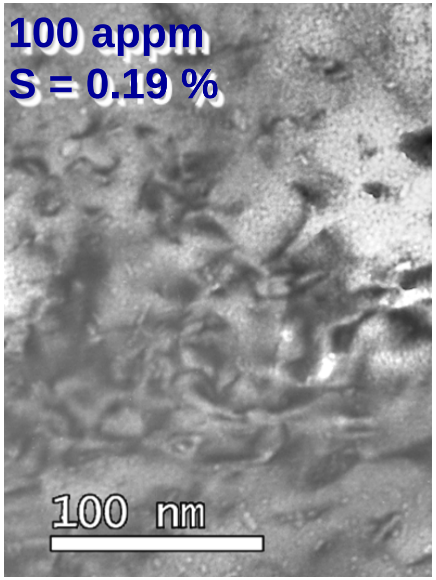
2. The dose rate was 2×10^{-2} dpa/s.

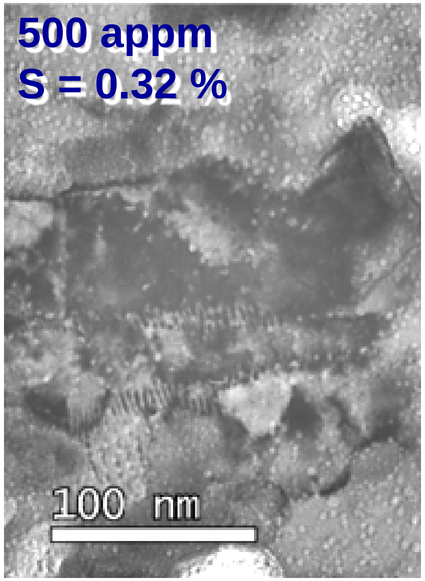
3. Doses 50 dpa (incubation period) and 200 dpa (steady–state period) was chosen to investigate gas influence on swelling.

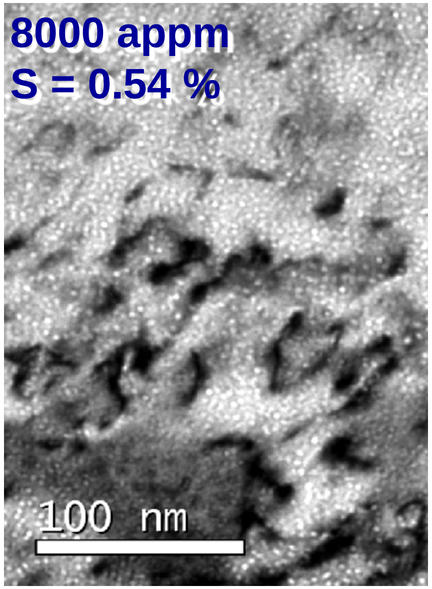
4. Gases levels were: helium 0 to 8,000 appm,
hydrogen 0 to 10,000 appm.

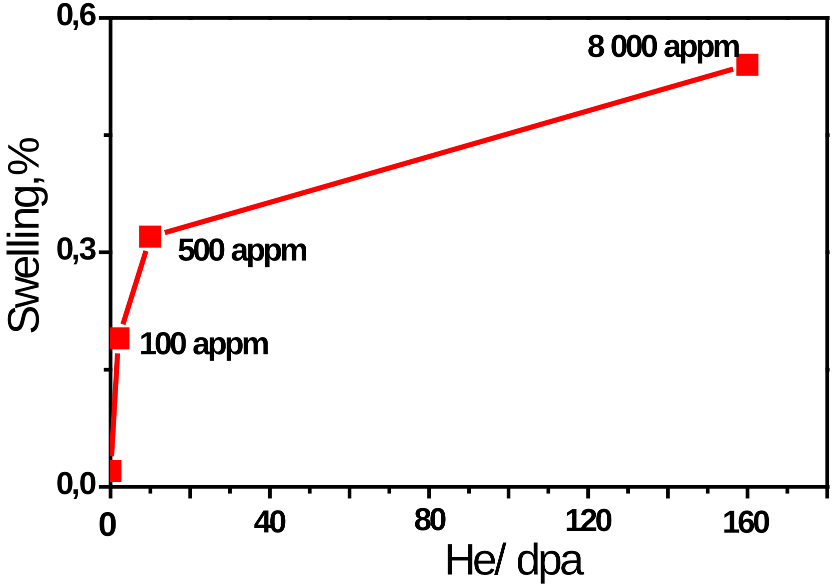
Helium effect on swelling at 50 dpa
EP-450, 480°C, He = 0 to 8,000 appm











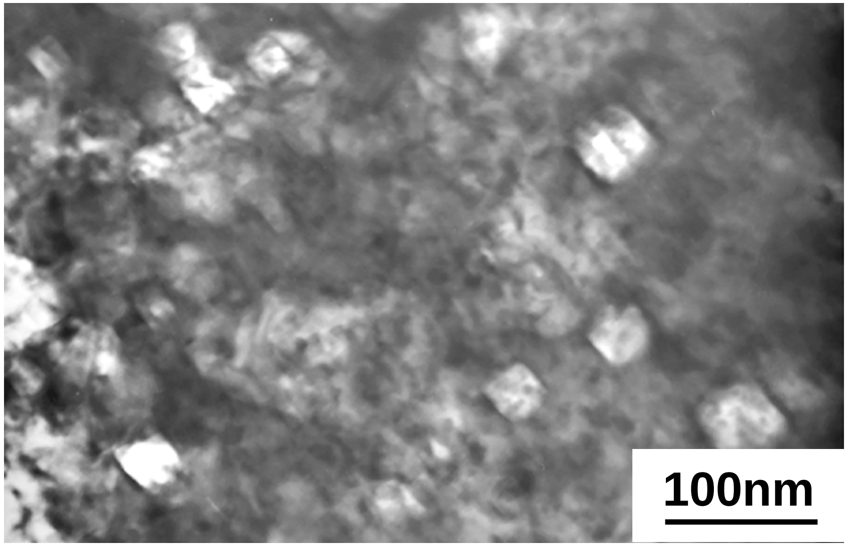
Co-injection of helium leads to strong increase of swelling in EP-450 at 50 dpa.

Swelling increasing strongly to ~500 appm followed by a slower state.

Helium effect on swelling at 200 dpa

EP-450, 480°C, He = 0-100 appm

Irradiation	Void size nm	Number density cm^{-3}	Swelling %
Cr (200 dpa)	32	$5 \cdot 10^{15}$	6.8
Cr+He (200 dpa+100 appm)	7	$2.8 \cdot 10^{16}$	0.37



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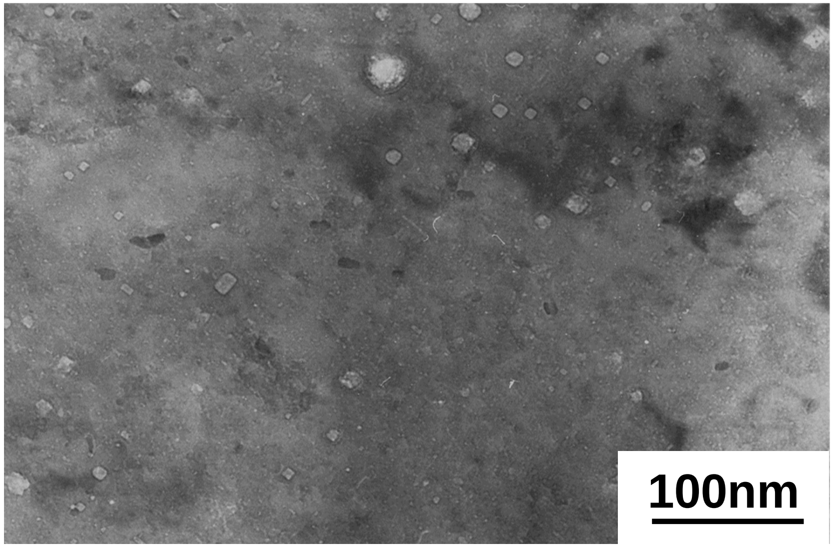
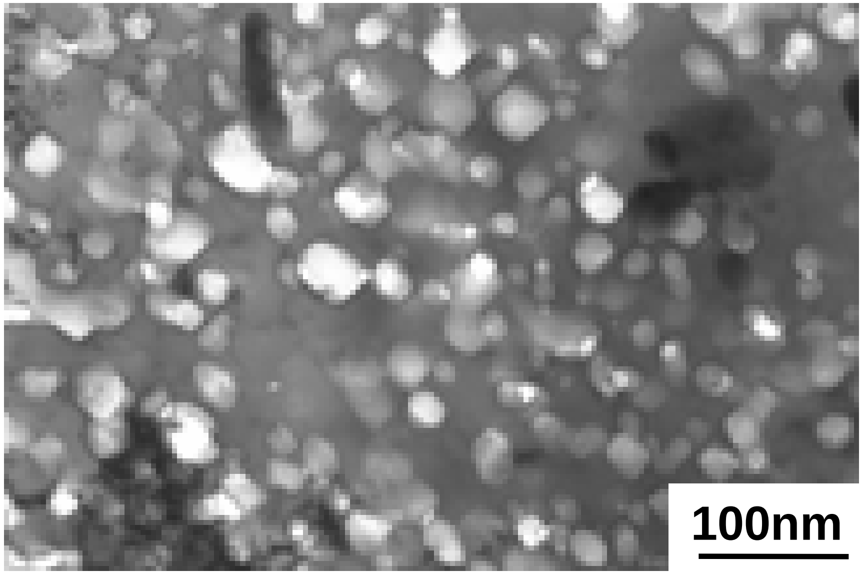


dual

Under dual irradiation with co-injection of helium, swelling decreases in EP-450.
Helium reduced the void size.

Helium effect on swelling at 200 dpa
EK181, 450°C, He = 0 or 100 appm

Irradiation	Void size , nm	Number density, cm^{-3}	Swelling, %
Cr (200 dpa)	30	$3 \cdot 10^{15}$	8.9
Cr+He (200dpa+100appm)	7	$7.5 \cdot 10^{16}$	1.34



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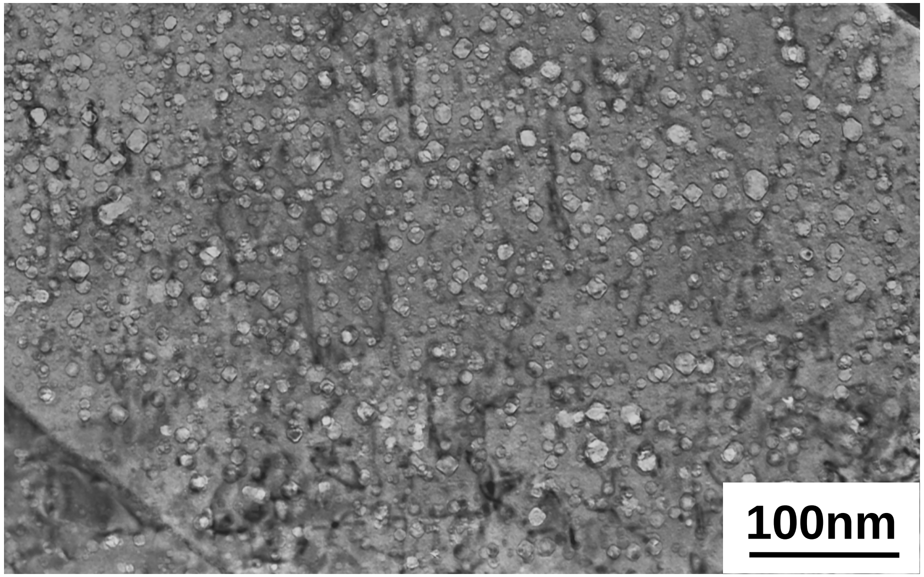
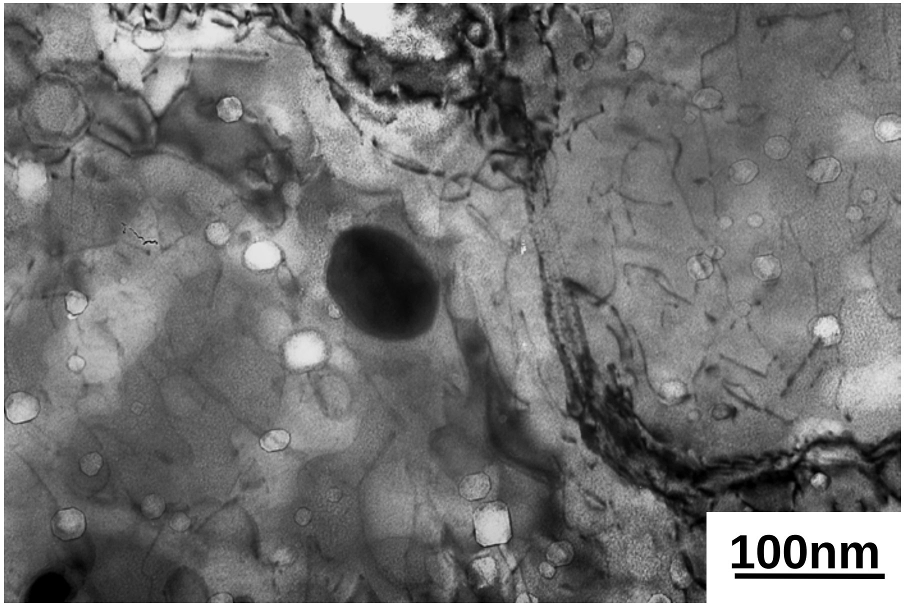


dual

Under dual irradiation with co-injection of helium the swelling decreases.
In EK-181 changes in voids size and density are similar to EP-450.

Helium effect on swelling at 200 dpa
F82H, 450°C, He = 0 or 100 appm

Irradiation	Void size , nm	Number density, cm^{-3}	Swelling, %
Cr (200 dpa)	14	$2.4 \cdot 10^{15}$	0.50
Cr+He (200dpa+100appm)	4	$5.2 \cdot 10^{16}$	0.10



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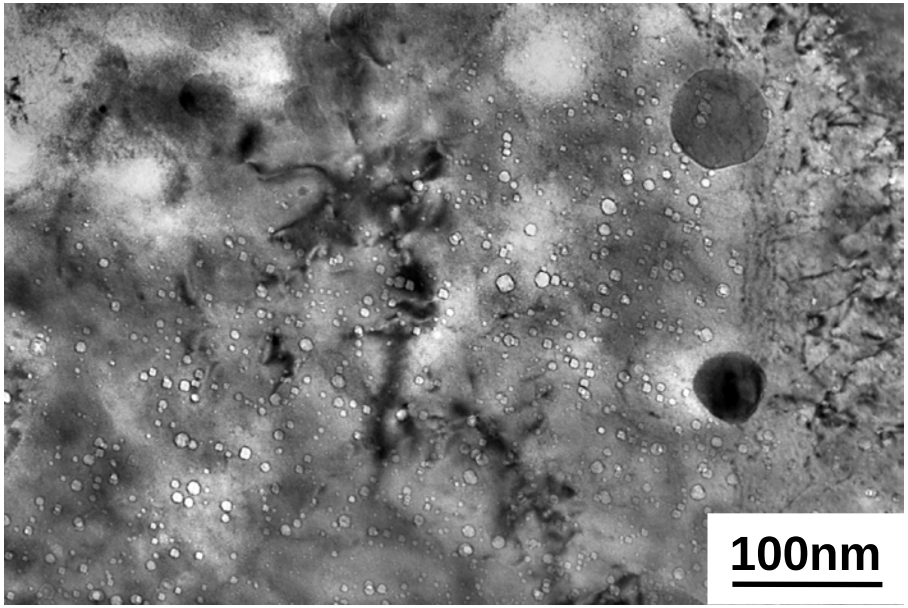
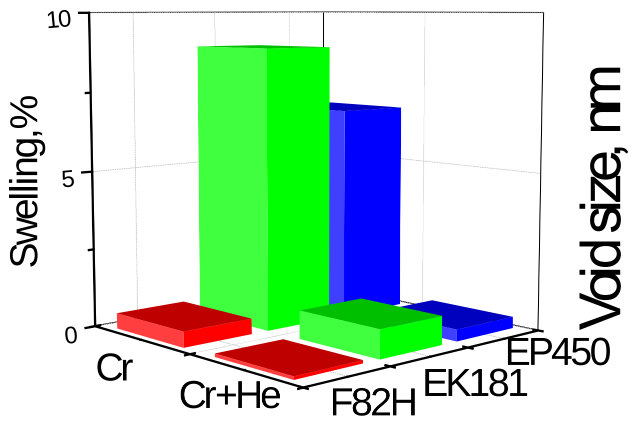


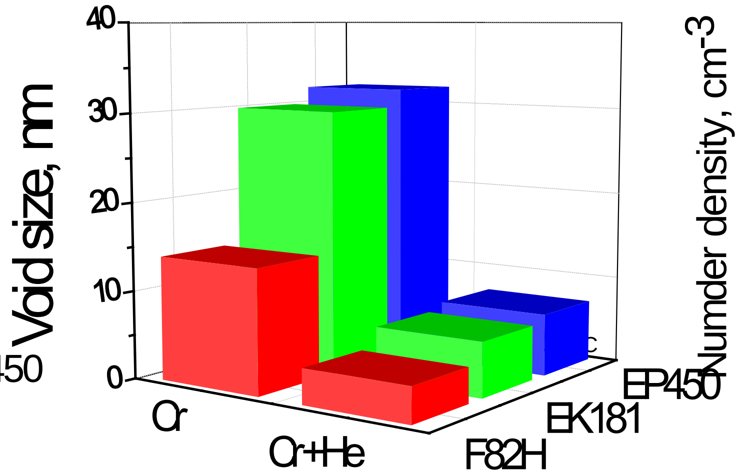
dual

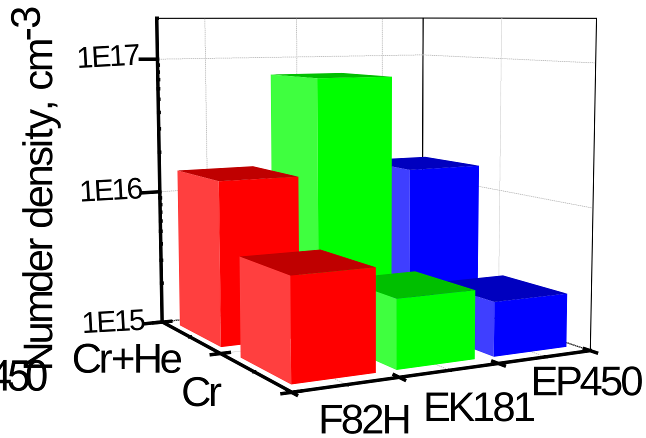
Under dual irradiation with co-injection of helium, the swelling decreases, but not so significant as in EP-450 steel.

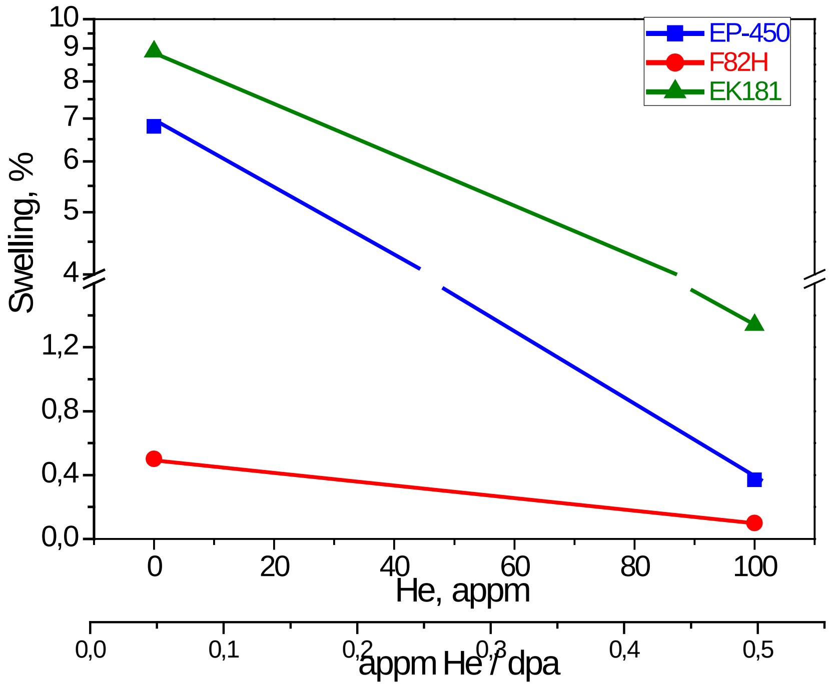
Damage and He effect on swelling in F-M steels T^{sw}_{max}, 200 dpa, He =0 or 100 appm

He-100 appm



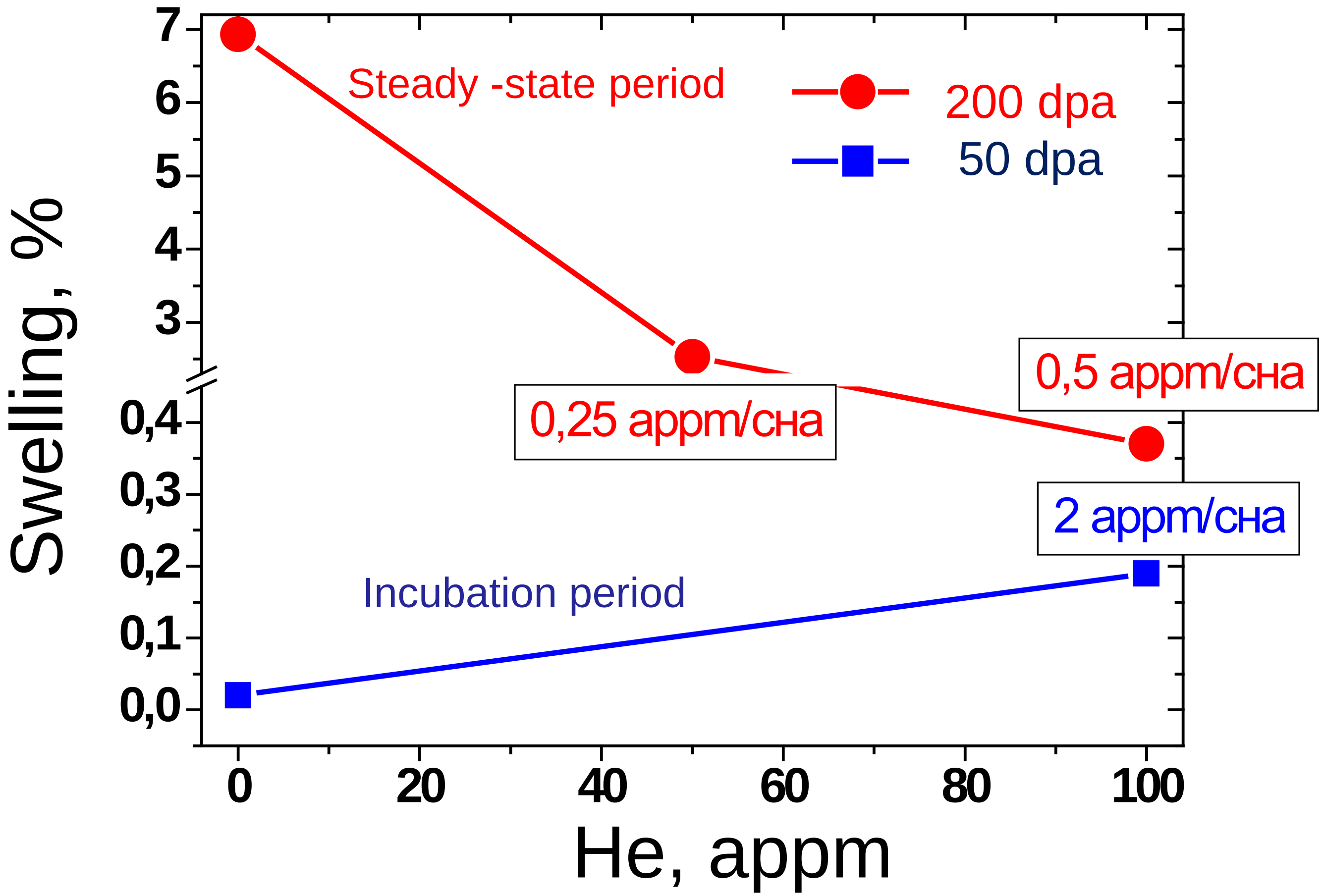






Dual irradiation at 200 dpa and co-injection of 100 appm He leads to swelling decrease (swelling have the same tendency).

Helium effect at different dose ranges on EP-450 swelling



At the incubation period helium increase the swelling of EP-450 steel.
At the steady state period helium decrease the swelling.
Under increase of helium concentration the effect of damage is level.

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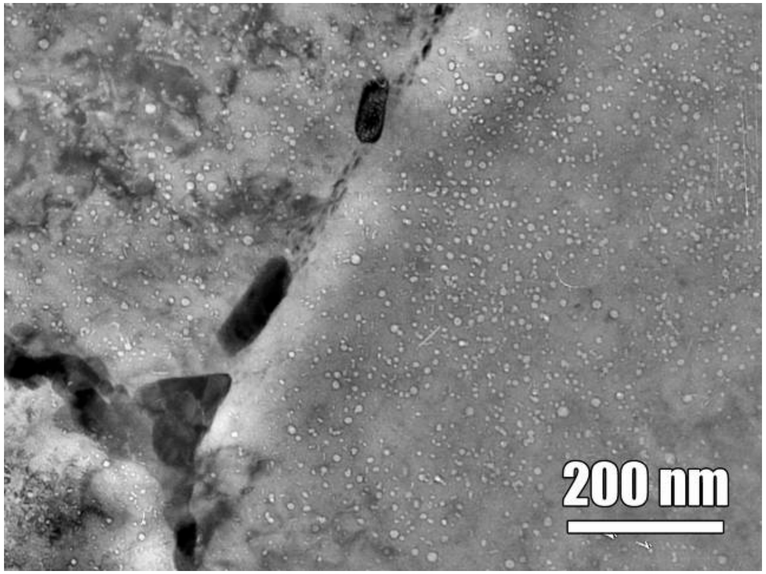
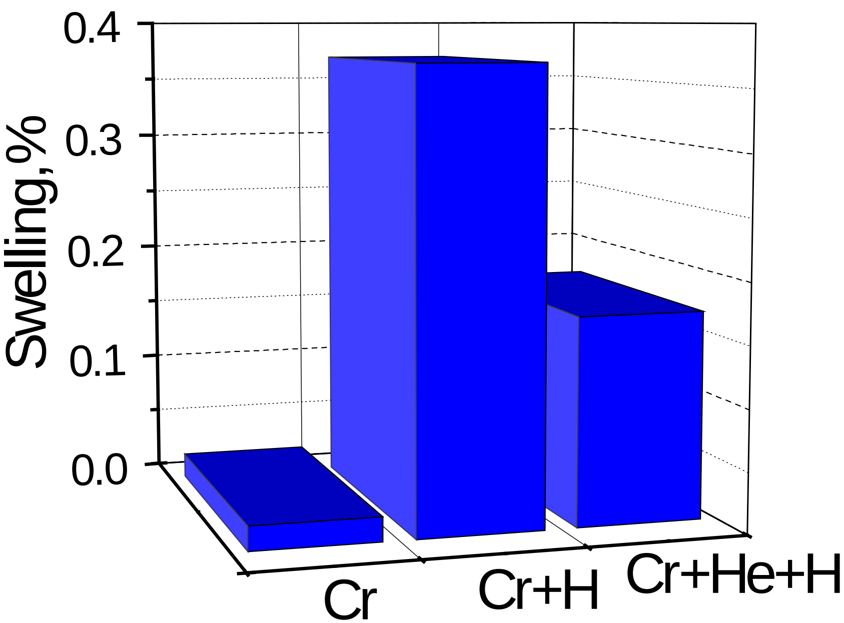
Effect of He & H in EP-450 steel (T_{irr}= 480^{0}C)

incubation period (50 dpa)

Irradiation	Void size , nm	Number density, cm^{-3}	Swelling, %
Cr (50 dpa)	15	$7.3 \cdot 10^{13}$	0.02
Cr+H (50 dpa + 10000 appm)	7	$7 \cdot 10^{16}$	0.37
Cr+He+H (50 dpa+1000 appm + 10000 appm)	20	$1.7 \cdot 10^{15}$	0.17

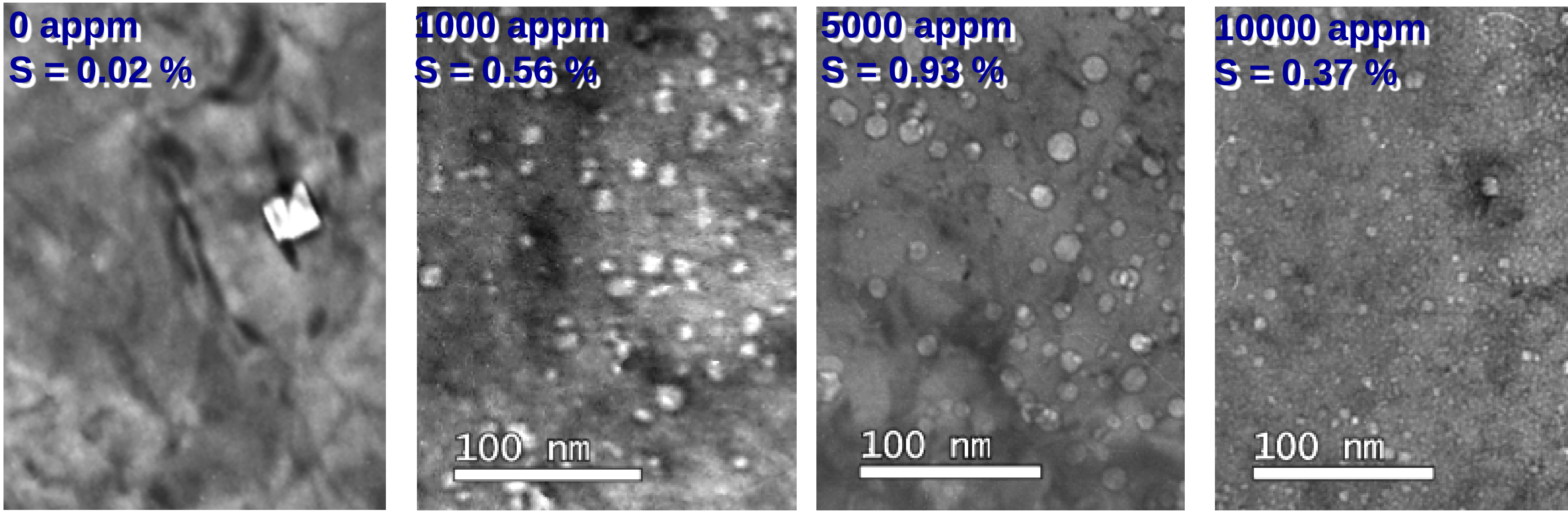


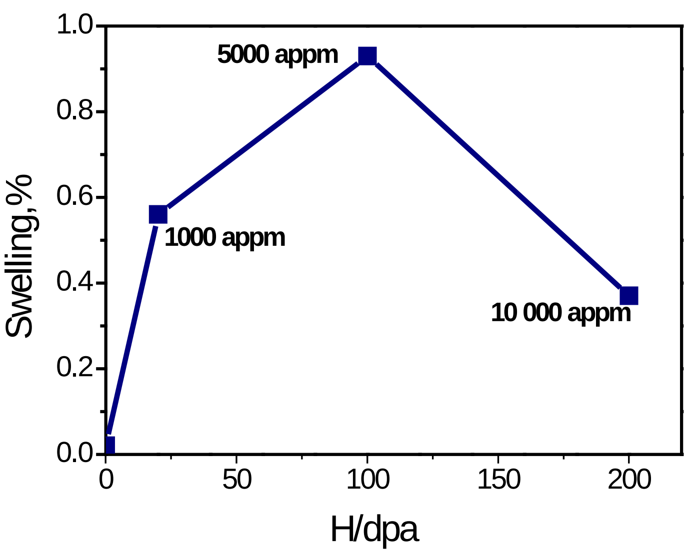
Formation of He-H-V complex?



Under dual irradiation at 50 dpa with hydrogen swelling increases ~15 times, and under triple beam irradiation ~ 8 times.

Damage & Hydrogen effect on EP-450 swelling
50 dpa, 480°C, H = 0 to 10,000 appm



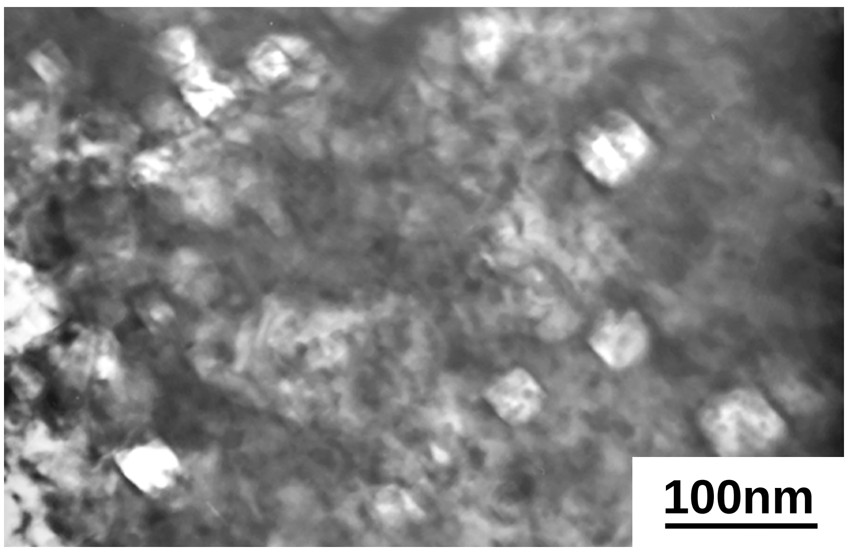


Co-injection of hydrogen at 1000-5000 appm in EP-450 steel leads to an increase of swelling.

Increase of hydrogen up to 1 at.% decreases the swelling.

Effect of He & H on swelling in EP- 450
480°C 200 appm He, 2000 appm H

Irradiation	Void size , nm	Concentration, cm^{-3}	Swelling, %
Cr (200 dpa)	32	$5 \cdot 10^{15}$	6.8
Cr+He+H (200dpa+200appm+ 2000appm)	7	$1.5 \cdot 10^{16}$	0.4



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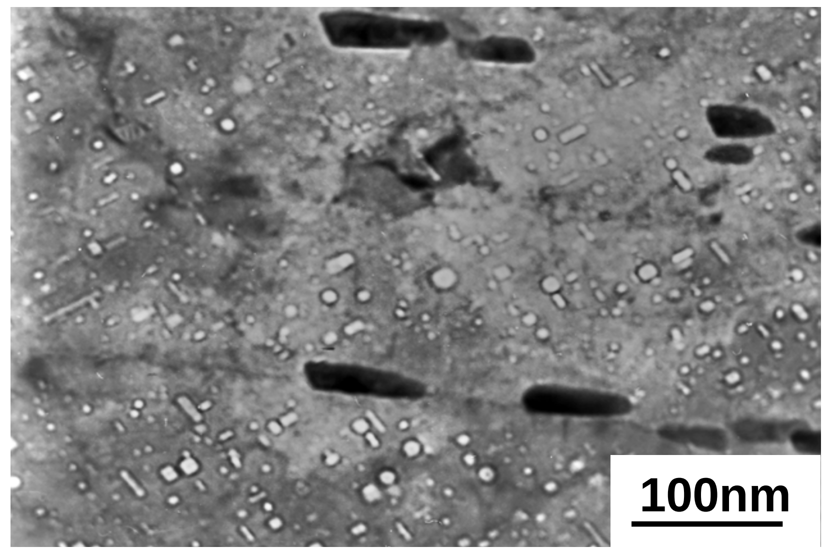
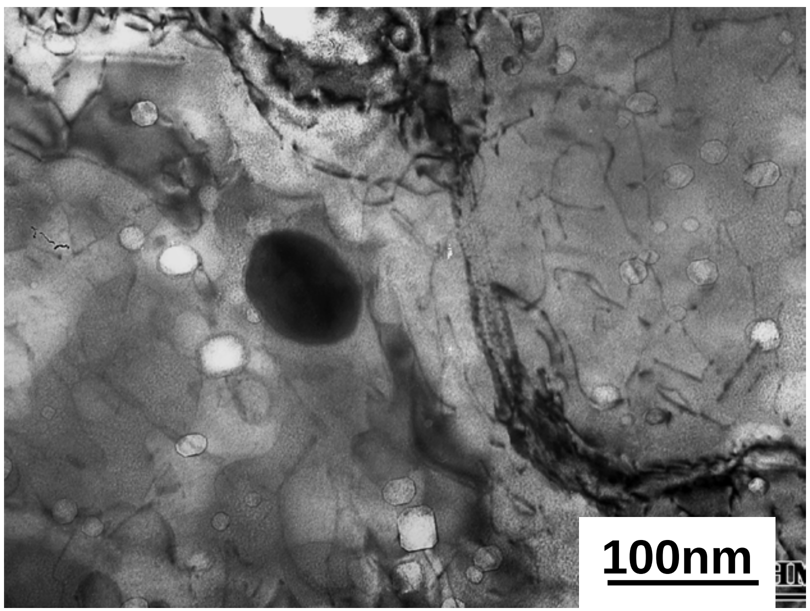


triple

Effect of He & H on swelling in F82H

450°C, 200 appm He, 2000 appm H

Irradiation	Void size , nm	Number density cm^{-3}	Swelling, %
Cr (200 dpa)	20	$2.4 \cdot 10^{15}$	0.50
Cr+He+H (200dpa+200appm+2000appm)	3	$1.7 \cdot 10^{17}$	0.05



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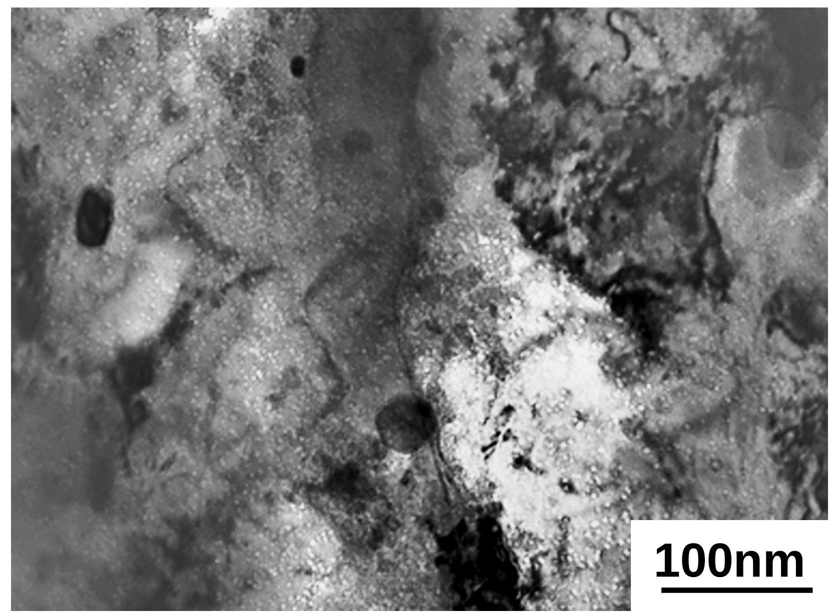
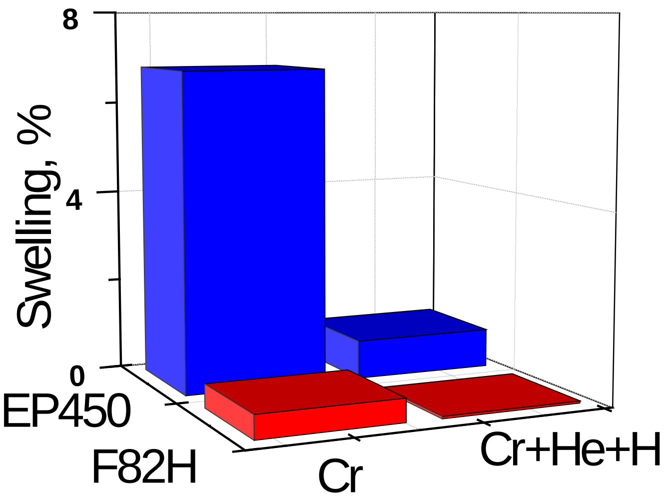
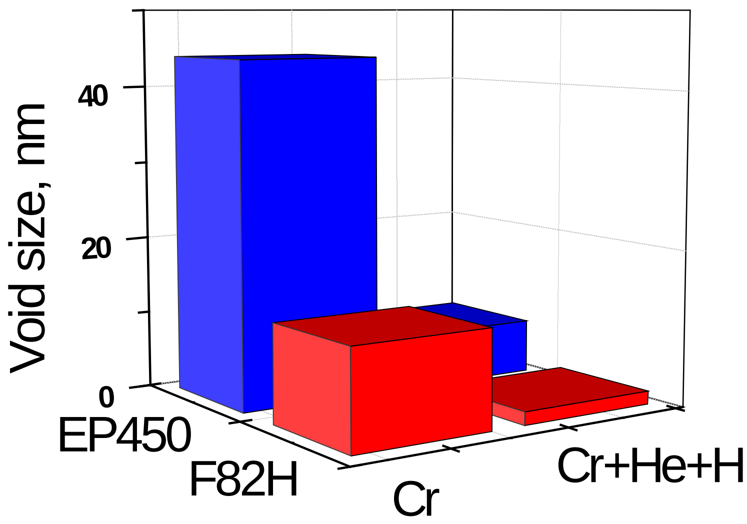


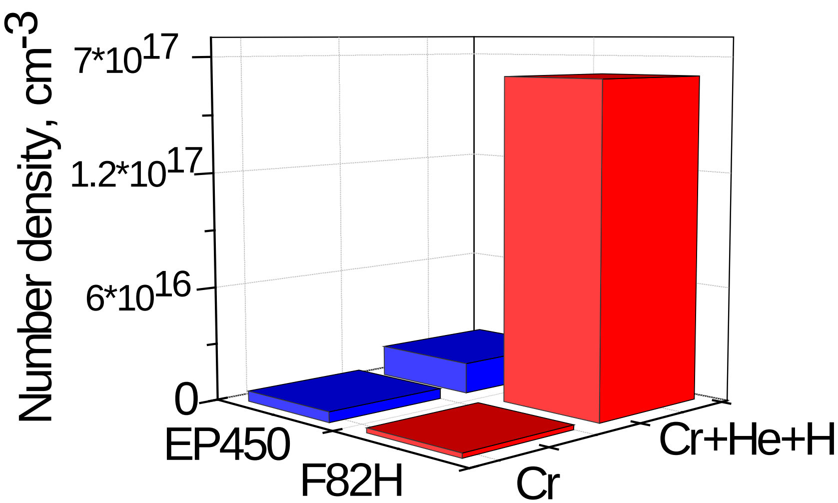
triple

Effect of He & H on swelling in EP- 450 & F82H
200 appm He, 2000 appm H

200 dpa, steady-state period



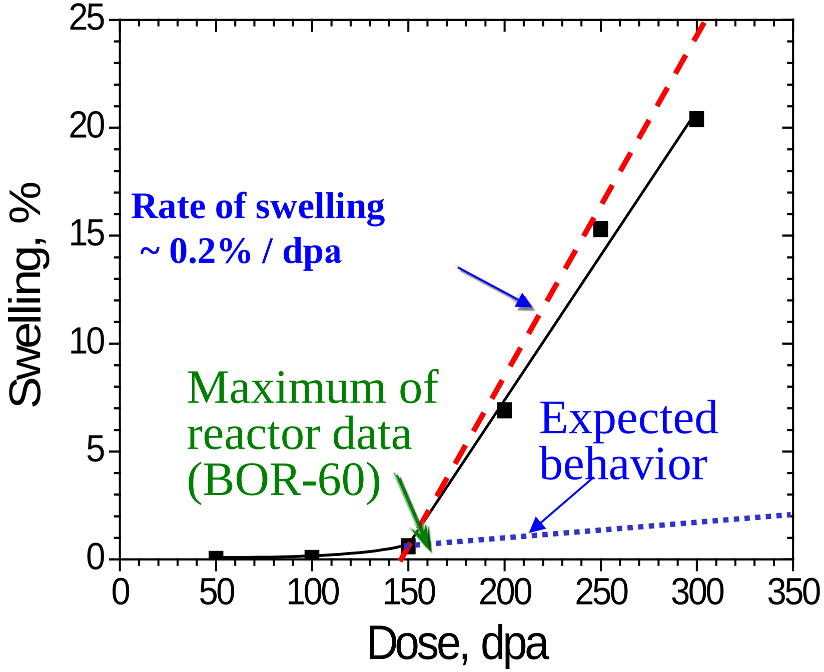


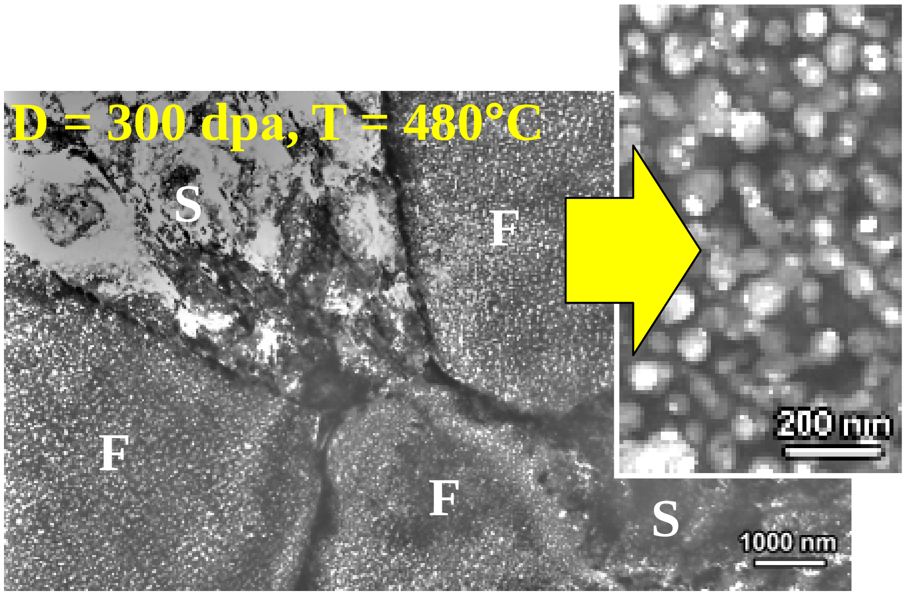


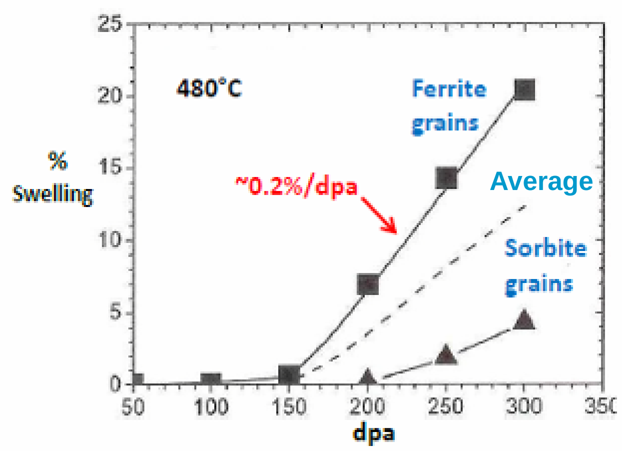
Under triple beam irradiation the swelling of both EP-450 and F82H decreases.

Under triple beam irradiation the swelling of EP-450 (ferritic/martensitic) is higher than of F82H (martensitic) steel.

Swelling of ferritic-martensitic steels EP-450 irradiated to super-high doses







➢Swelling of BCC steels may reach more than 20% at super-high doses;

➢Ferritic grains in EP-450 begin swelling earlier than sorbite grains;

➢Average swelling rate depends on volume ratio of ferrite to sorbite grains;

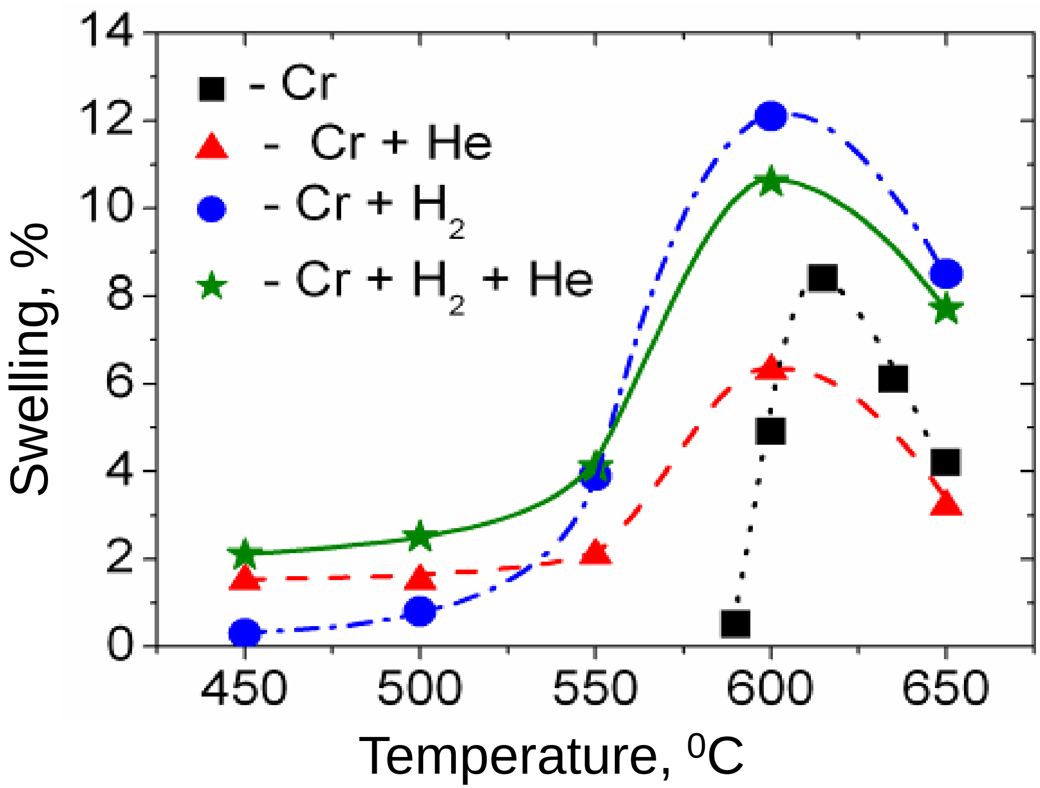
➢The rate of swelling in steady-state stage is 0.2%/dpa, agreeing with observed swelling of binary Fe-Cr alloys irradiated in EBR-II and FFTF.

Conclusions

- Investigations of swelling in EP-450, F82H and EK-181 under single, dual and triple beam irradiation were carried out.
- Ferritic–martensitic steels are sensitive to co-injection of helium and hydrogen:
  - in the incubation period helium increases swelling due primarily to number density increase .
  - helium exhibits a strong influence on void nucleation, increasing their concentration.
  - in the incubation period hydrogen is as effective as helium in accelerating the onset of swelling, but the cavity size distribution is different.
  - in the steady-state period helium co-injection decreases the swelling of EP-450, EK-181, F82H.
- Under triple ion irradiation the swelling decreases in all investigated materials.
- Under dual and triple irradiation the behavior of swelling has the same tendency for EP-450 (12%Cr,F/M), F82H(8%Cr, M) and EK-181 (12%Cr, M).

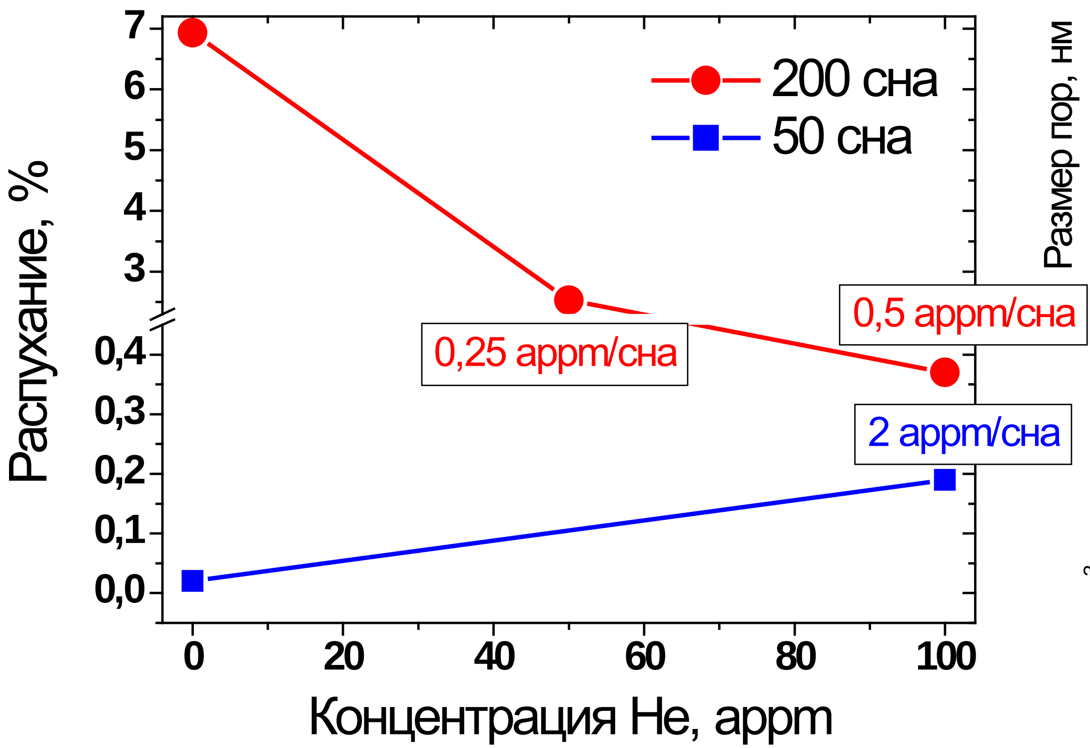
THANK YOU FOR YOUR ATTENTION!!!

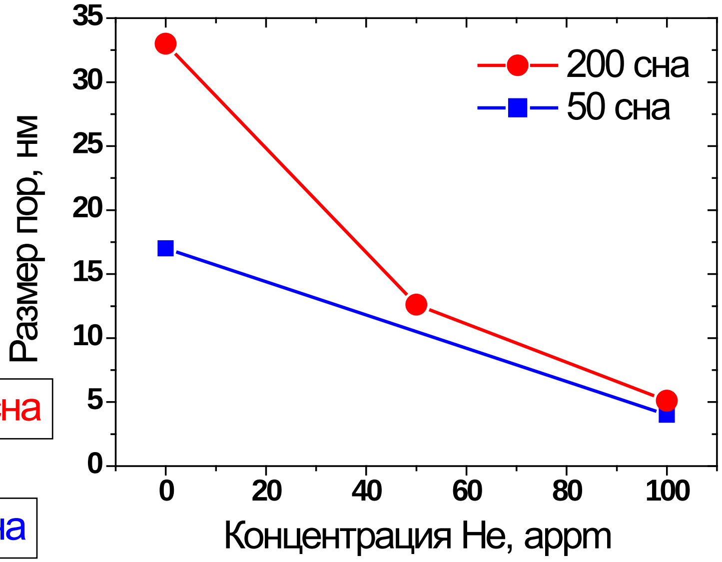
Effect of He & H in 18Cr10NiTi steel (50 dpa)

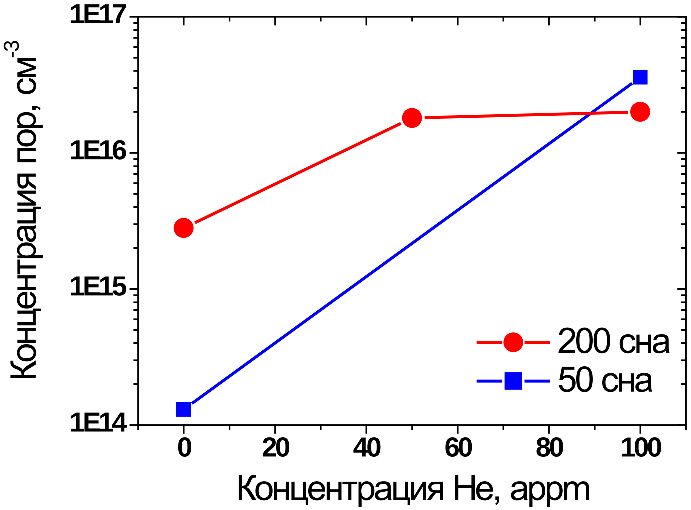


- **Helium injection leads to enhanced void nucleation at all temperatures examined, but depresses swelling somewhat at higher temperatures.**
- **Hydrogen injection is also very effective to nucleate voids, but exhibits different behavior with temperature.**
- **Triple beam irradiation does not show a strong synergism between helium and hydrogen in this alloy.**

Effect of He on swelling of EP-450 irradiated at T=480 ºC and doses 200 and 50 dpa

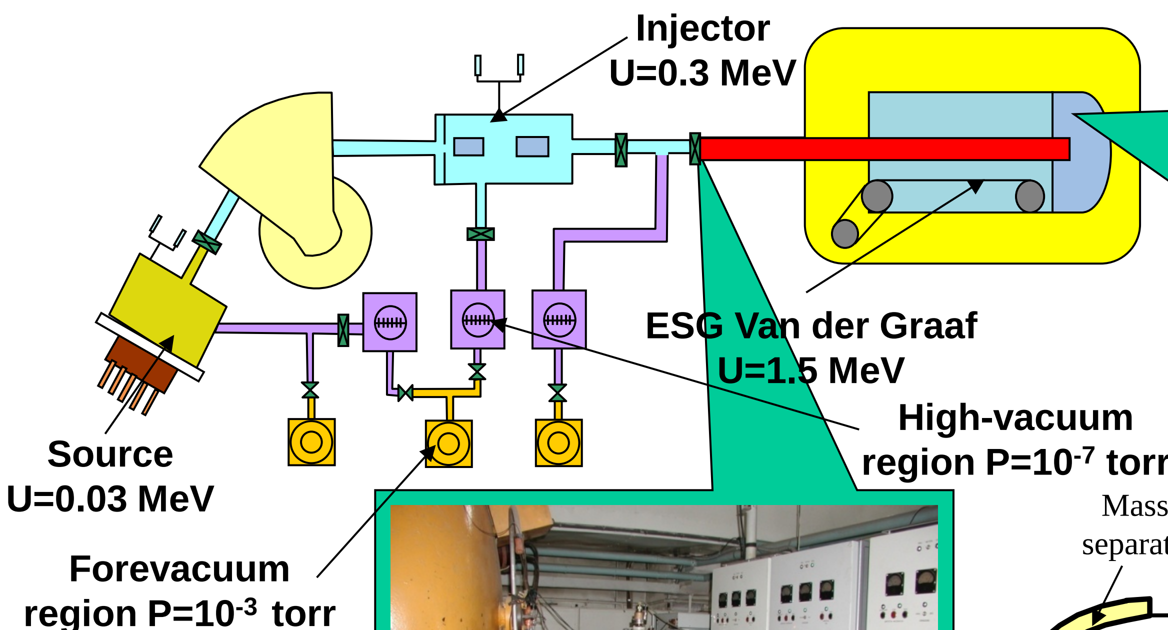


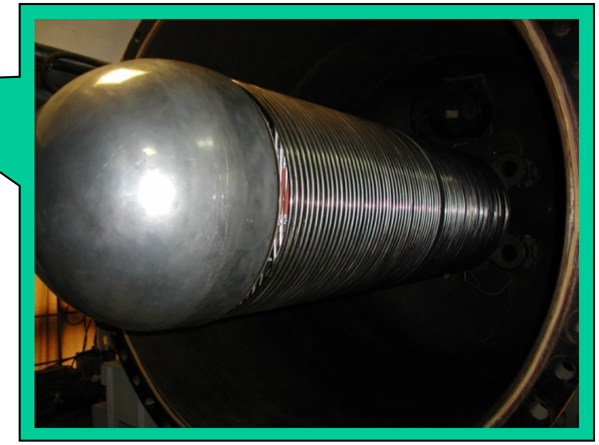


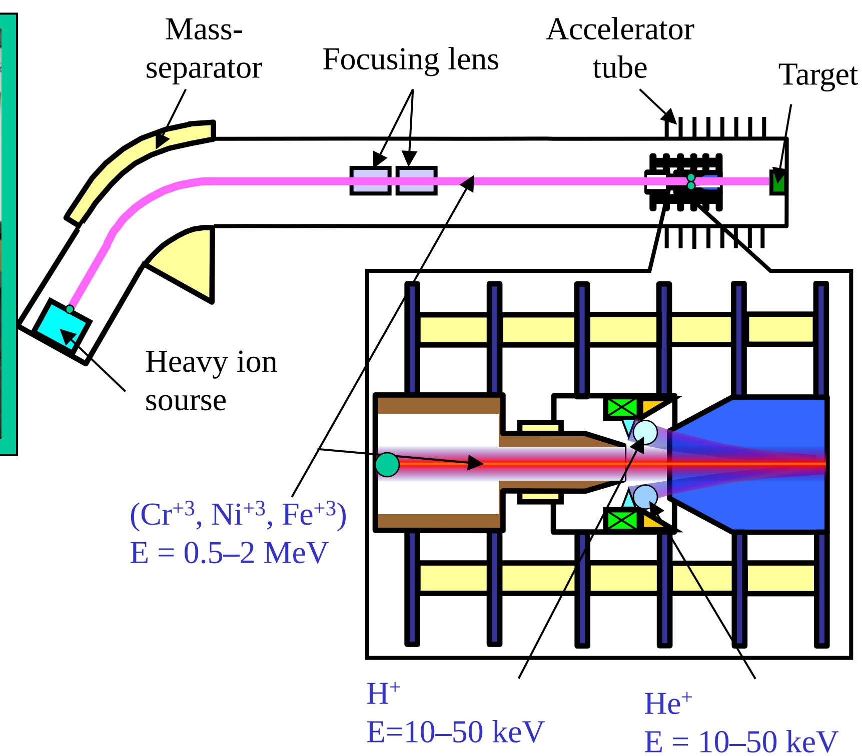


At incubation period helium increase of swelling due to number density increase . At steady-state period helium decrease swelling due to voids size

Electrostatic Accelerator with External Injector (ESUVI)

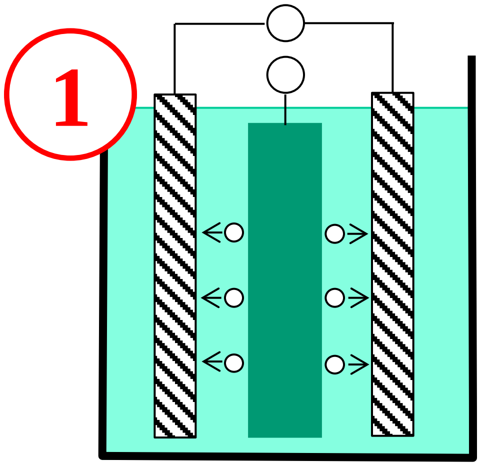




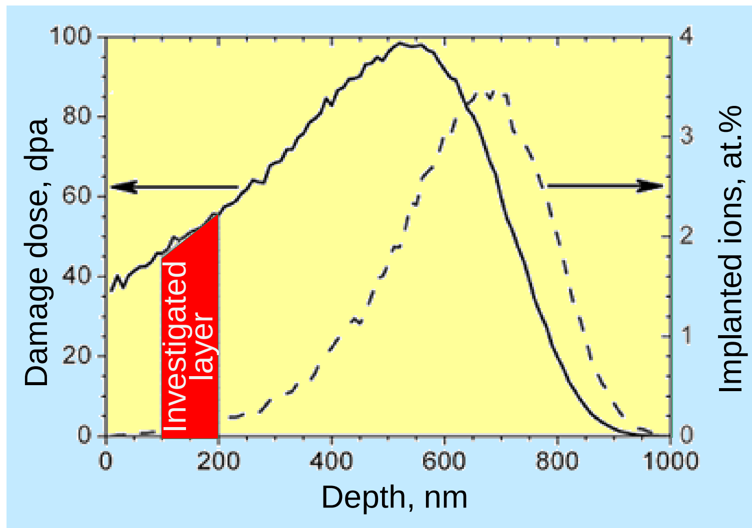


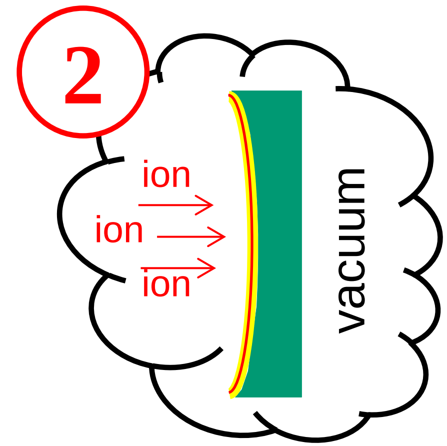
Irradiation	Parameters	
Cr Cr+He Cr+H Cr+He+H	E_{Cr}= 0.3 - 3 MeV j_{Cr} =1 - 45 μA/cm^2 E_{He}= 10 - 60 keV j_{He} = 1 – 500 nA /cm^2 E_H= 10 - 60 keV j_H = 1 – 500 nA /cm^2	T $_{irr}$= 60 – 800^{0}C D = 0 - 250 dpa k =7·10^{-5} - 7·10^{-3} dpa/s 0.13 – 1.3 appmHe/s 0.1 – 1 appmH/s

Specimen's preparation

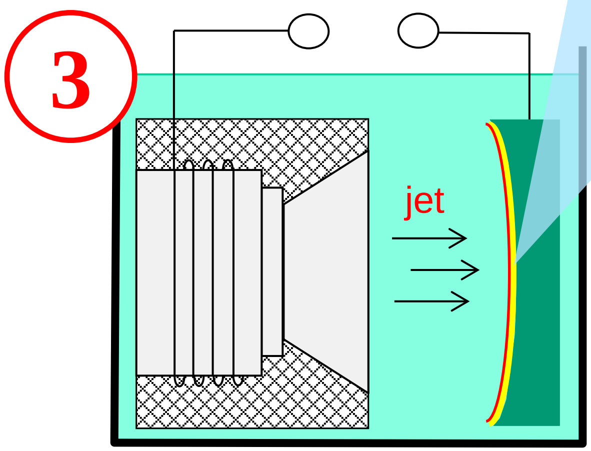


Electropolishing

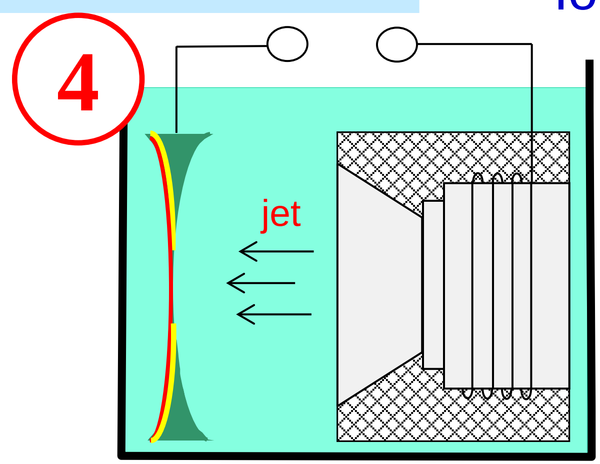




Ion irradiation

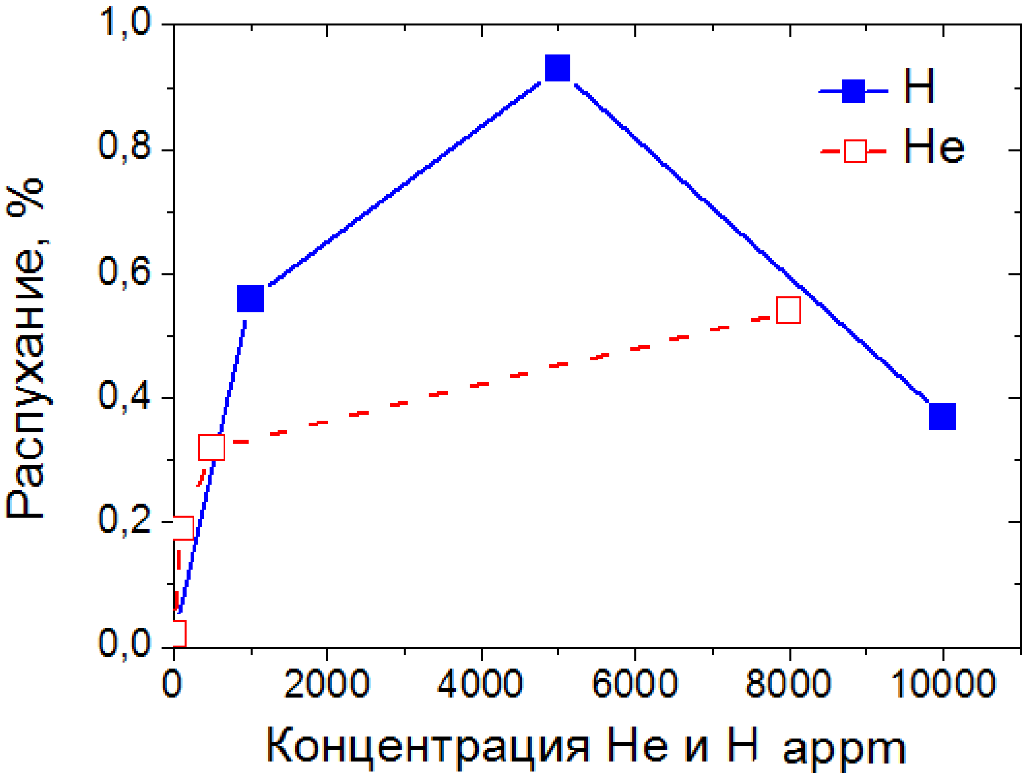


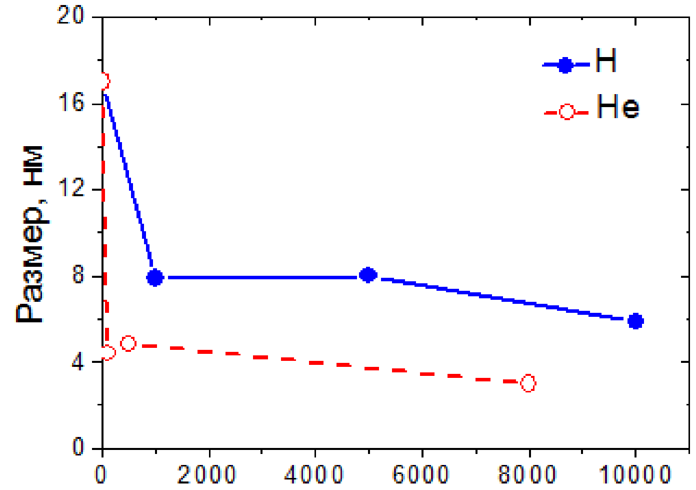
Control thinning irradiated side

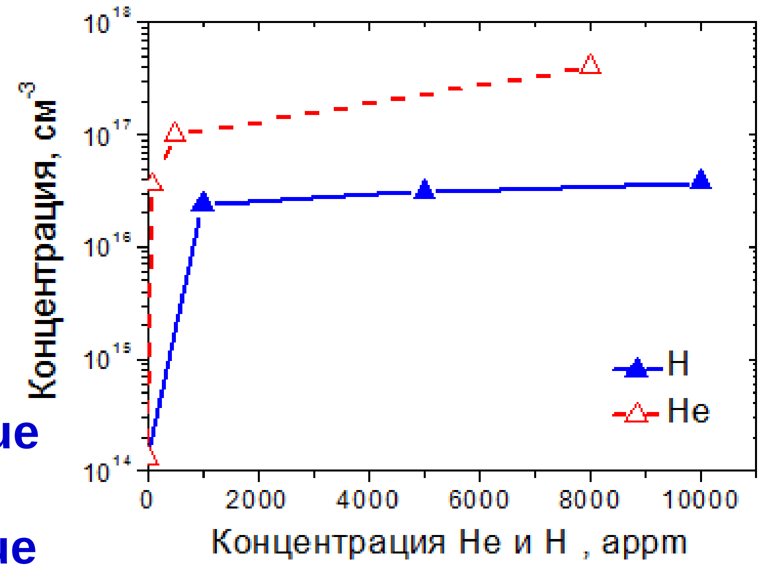


One-side thinning
by jet electropolishing

Effect of He и H on swelling on EP-450 irradiated at T=480 ºC to dose 50 dpa







At 0-1,000 appm H increases swelling due to number density increasing .
At 10,000 appm H decreases swelling due to voids size reduction