Anticipated dimensional and microstructural stability of ferritic-martensitic alloys irradiated in heavy liquid metal cooled reactors and ADS systems

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Background

- **Interest focused on ferritic-martensitic and ODS-ferritic alloys for service in ADS devices and advanced fast reactors cooled by Pb or Pb-Bi eutectic.**

- **The alloys chosen for use in Pb-Bi, however, have only a small amount of data on dimensional and microstructural stability, and all data were generated in sodium-cooled reactors.**

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- **Can we forecast the behavior at high neutron exposures of ferritic and ferritic-martensitic alloys in general?**

  EP-450, HT9, T91, EK-181, MA957, 14YWT

- **What forecast can be made for alloys specifically developed for service in Pb-Bi?**

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  EP-823, EP-852

Known to swell under ion and/or neutron irradiation?

Composition (wt%) of F/M steels used in Russian program

EP-450 **Fe-12.95Cr-0.20Ni-0.31Mn-1.54Mo-0.20Si - 0.47Nb-0.22V--0.14C-0.017P**

EP-823 **Fe-11.40Cr-0.70Ni-0.60Mn-0.67Mo- 1.05Si - 0.20Nb-0.40V--0.18C-0.012P-0.03Ti-0.04N_2-0.65W**

EP-852 **Fe-13.15Cr-0.27Ni-0.31Mn-1.69Mo-1.91Si - 0.13C-0.017P**

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Heavy silicon addition generally increases phase instabilities during irradiation in both austenitic and ferritic alloys. (Strains)

Silicon addition tends to delay onset of void swelling.

Assumptions based on previous studies of austenitic alloys

- **If dose exposures in fast reactors are specified in displacements per atom (dpa) then radiation-induced changes in mechanical and dimensional properties are independent of coolant identity.**
- **All alloys in a given class will exhibit similar swelling and creep behavior.**

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All Austenitics

Steady-state swelling rate of 1%/dpa

All material and environmental influence in transient duration

Creep compliance 1 X 10^{-6} Mpa^{-1} dpa^{-1}

Swelling-creep coupling coefficient 0.6 x 10^{-3} Mpa^{-1}

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All Austenitics All ferritics and ferritic-martensitics? ODS?

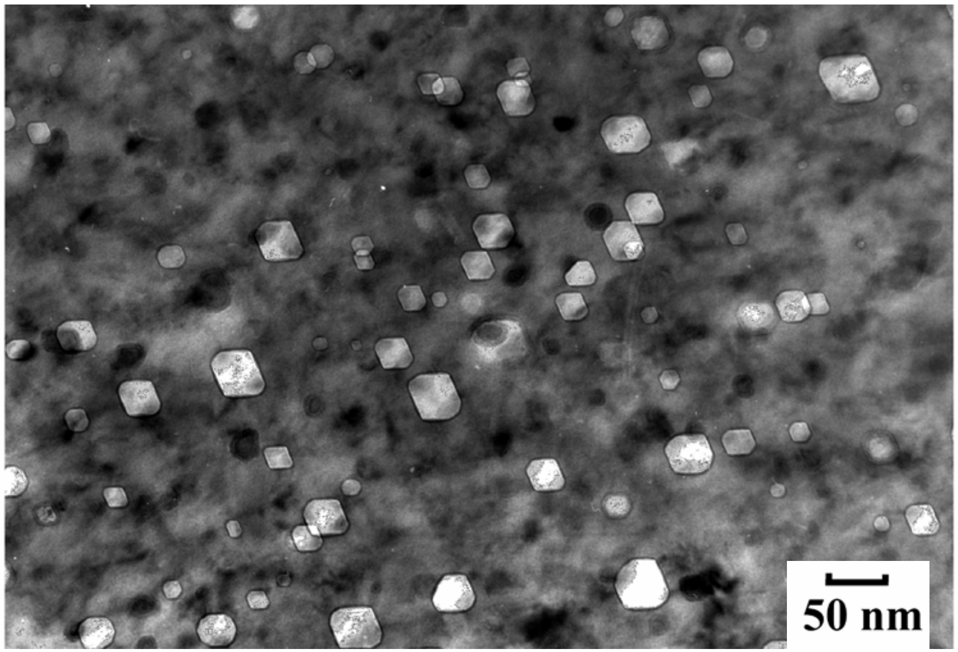
Steady-state swelling rate of 1%/dpa 0.2%/dpa?

All material and environmental influence in transient duration?

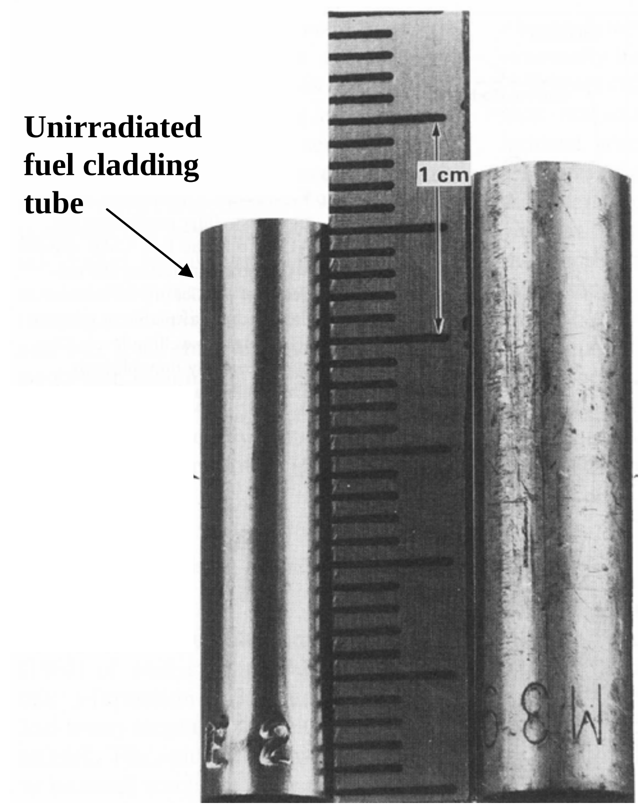
Creep compliance 1 X 10^{-6} Mpa^{-1} dpa^{-1} ~ 0.5 X 10^{-6} Mpa^{-1} dpa^{-1}

Swelling-creep coupling coefficient 0.6 x 10^{-3} Mpa^{-1} unchanged

Major concern is void swelling to increase dimensions and volume of structural steels



Voids introduce ~1% swelling in 304 stainless steel at 380°C and 22 dpa



~33% increase in volume of cold-worked 316 stainless steel at 510°C and 80 dpa

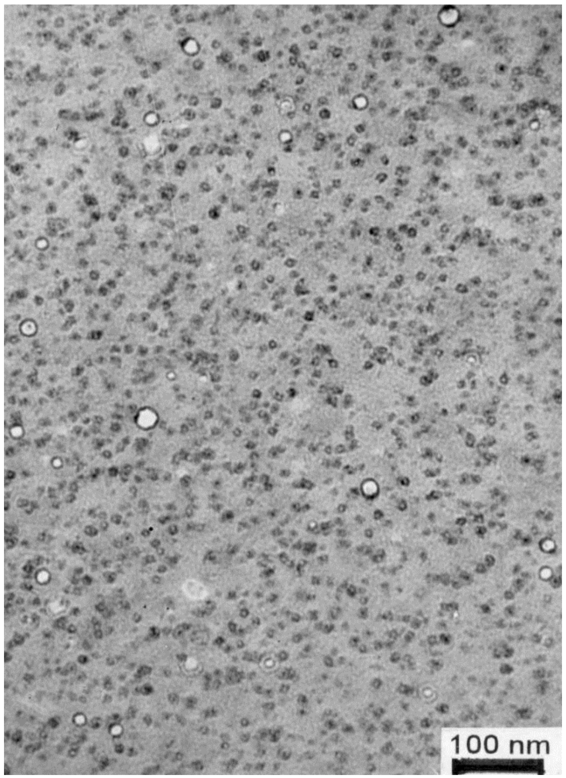
Questions to address

- **Nature of void swelling law? Mono-linear or bi-linear?**
- **Duration of transient regime of swelling?**
- **Steady-state swelling rate?**
- **Swelling-creep relationship?**
- **Other strain contributions, especially arising from phase instabilities.**

Precipitate microstructure of EP-450 after irradiation in BN-350 or BN-600

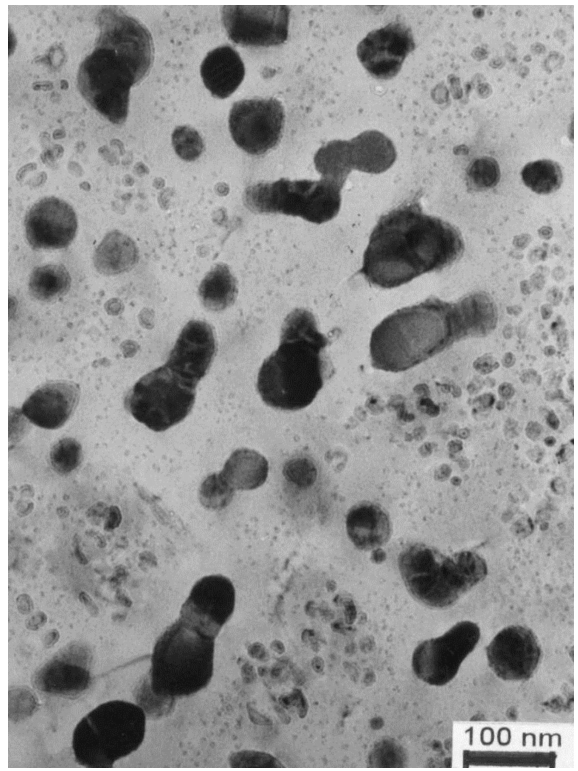
Dvoriashin, Porollo, Konobeev and Garner, 2004

α'-phase



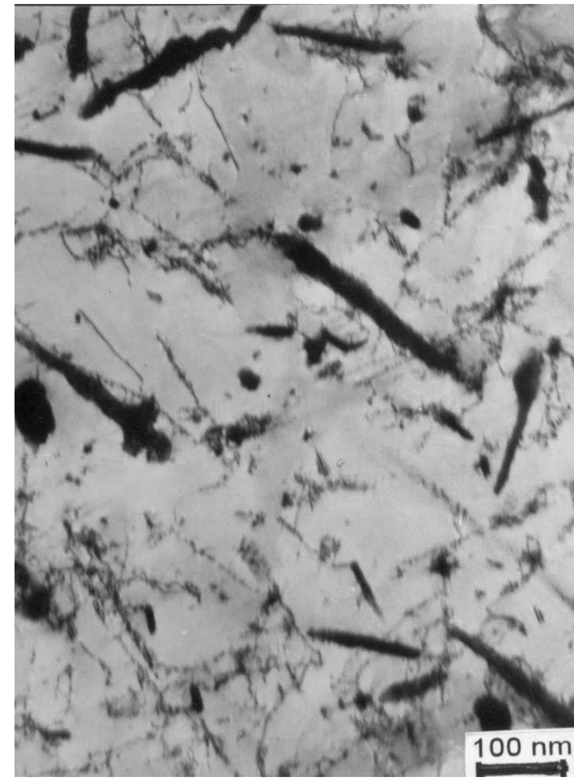
BN-350, 380°C, 56 dpa

χ-phase



BN-600, 465°C, 81 dpa

M_2X



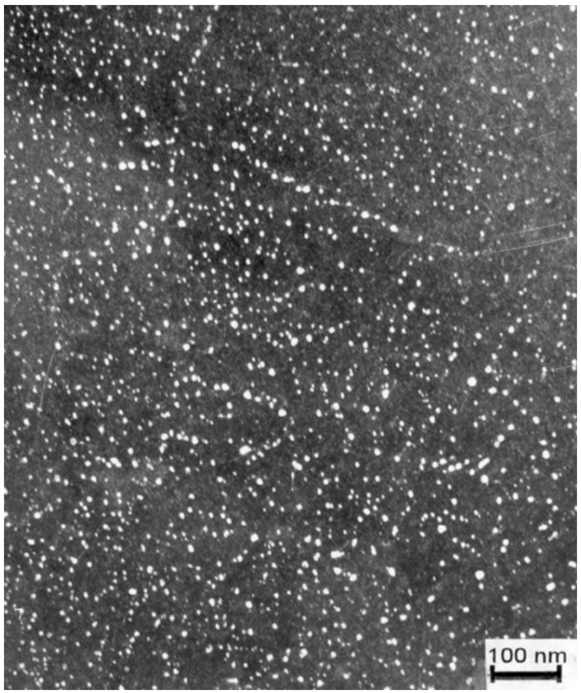
BN-350, 690°C, 17 dpa

Phase changes almost always generate volume changes and strains.

Precipitate microstructure of EP-823 fuel pin cladding after irradiation in BOR-60

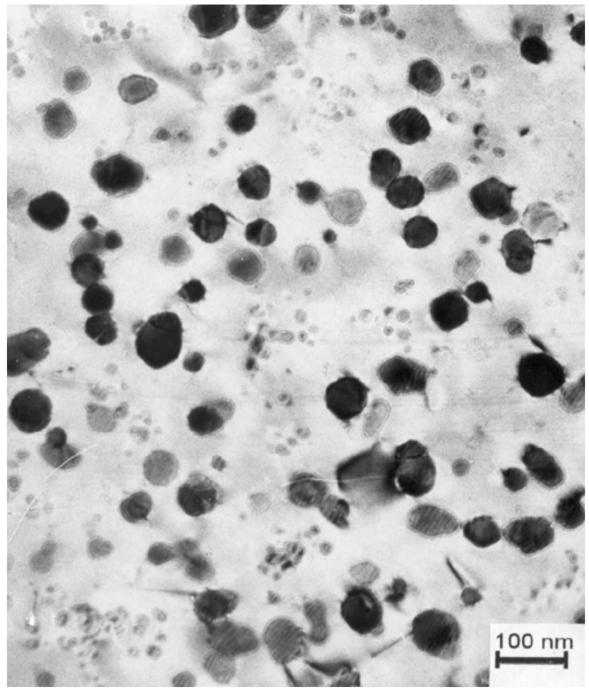
Porollo, Dvoriashin, Konobeev and Garner, 2004

M_6C



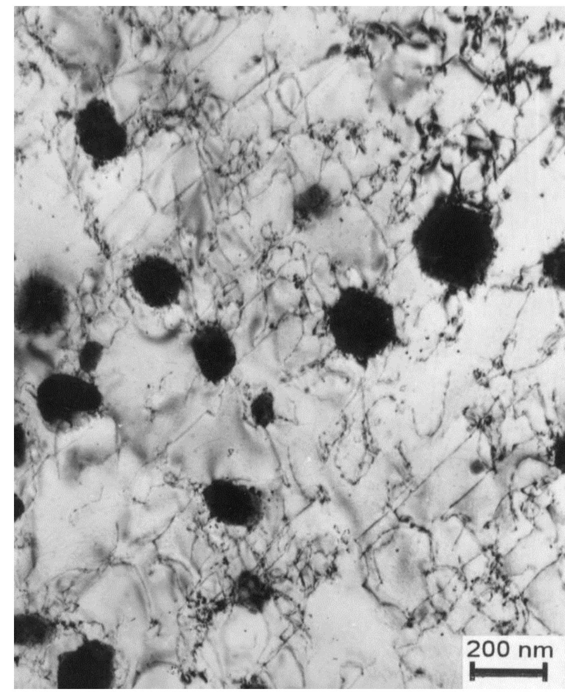
365-385°C, 50.5 dpa

M_6C and χ-phase



465-535°C, 63 dpa

χ-phase



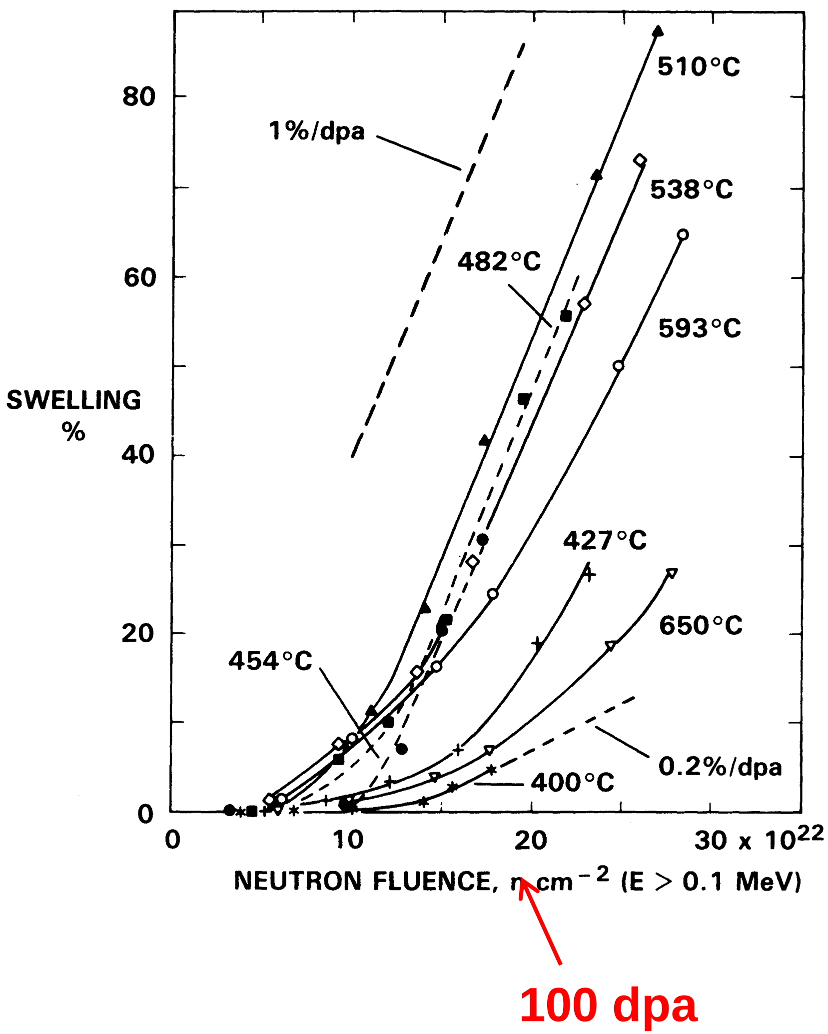
580-680°C, 33 dpa

Isolated voids observed only at 365-385°C.

Relative swelling suppression compared to EP-450 is attributed to higher silicon level of EP-823.

Swelling of 20% CW 316 stainless steel in EBR-II

Garner and Gelles, 1990

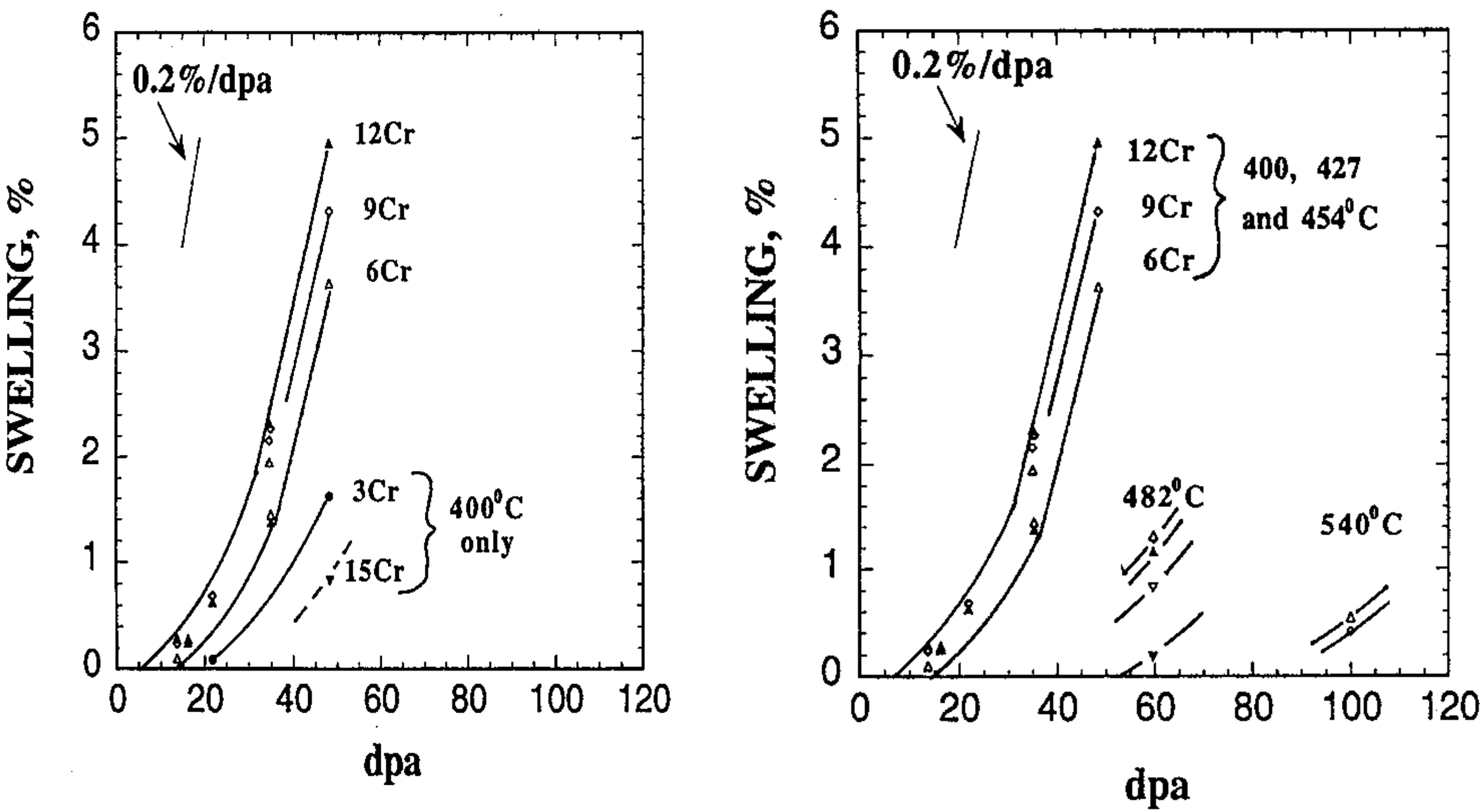


- Transient followed by steady-state swelling. "Bilinear"
- The eventual swelling rate of 316 at all reactor-relevant temperatures is ~1%/dpa.
- Dependence on compositional, fabricational and environmental variables lies primarily in the duration of the transient regime.

- **Is there a similar behavior for ferritic and ferritic-martensitic alloys?**

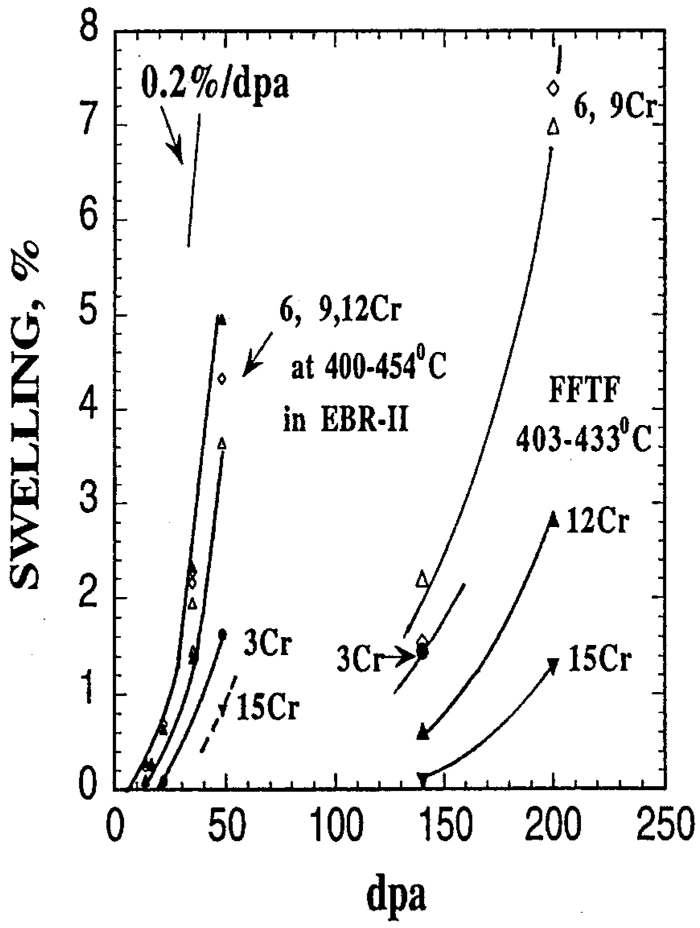
Swelling of Fe-Cr binary alloys in EBR-II at 400-450°C

Garner, Toloczko and Sencer, 1998



Very similar to behavior of that of Fe-Cr-Ni austenitic alloys

Swelling of Fe-Cr binary alloys in EBR-II and FFTF at 420-450°C



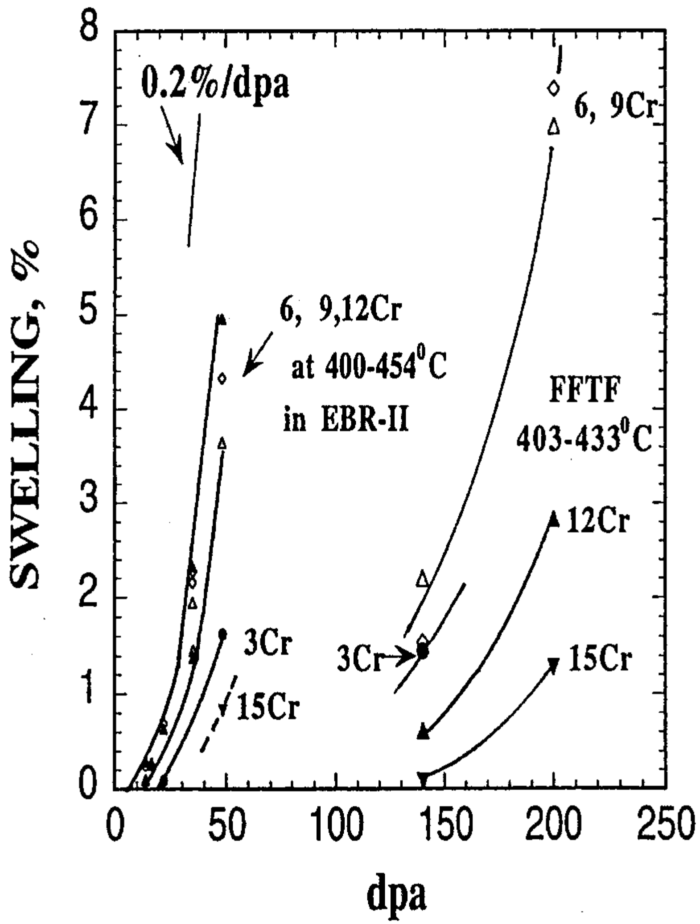
Garner, Toloczko and Sencer, 1998

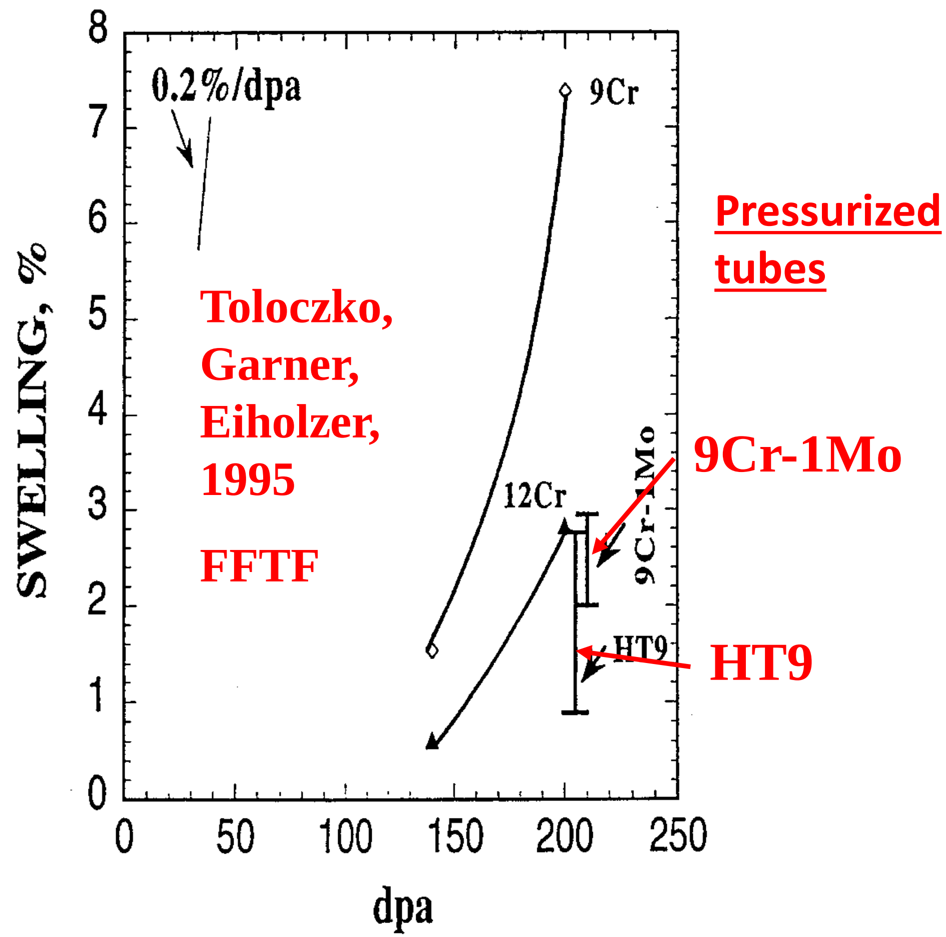
Steady-state swelling rate is ~0.2%/dpa.

Compared to EBR-II, FFTF has a higher dpa rate and a much lower helium generation rate, leading to a much longer transient regime of swelling.

Swelling of Fe-Cr binary alloys and F/M steels in EBR-II and FFTF at 420-450°C

Garner, Toloczko and Sencer, 1998

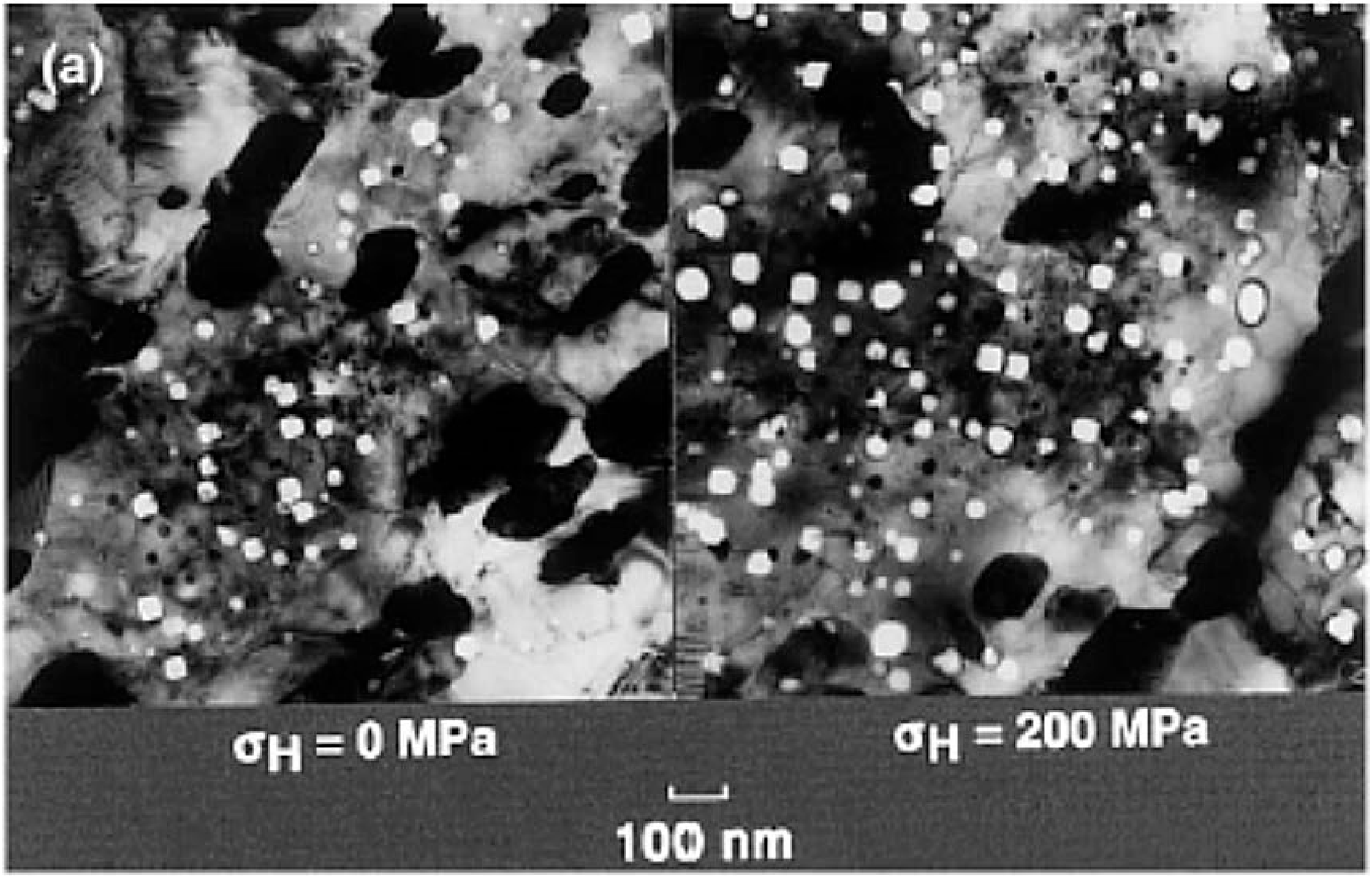




Microstructure of HT9 pressurized tubes after irradiation in FFTF to 208 dpa at ~400ºC

Toloczko, Garner and Eiholzer, 1994

~1% density change ~2.6%



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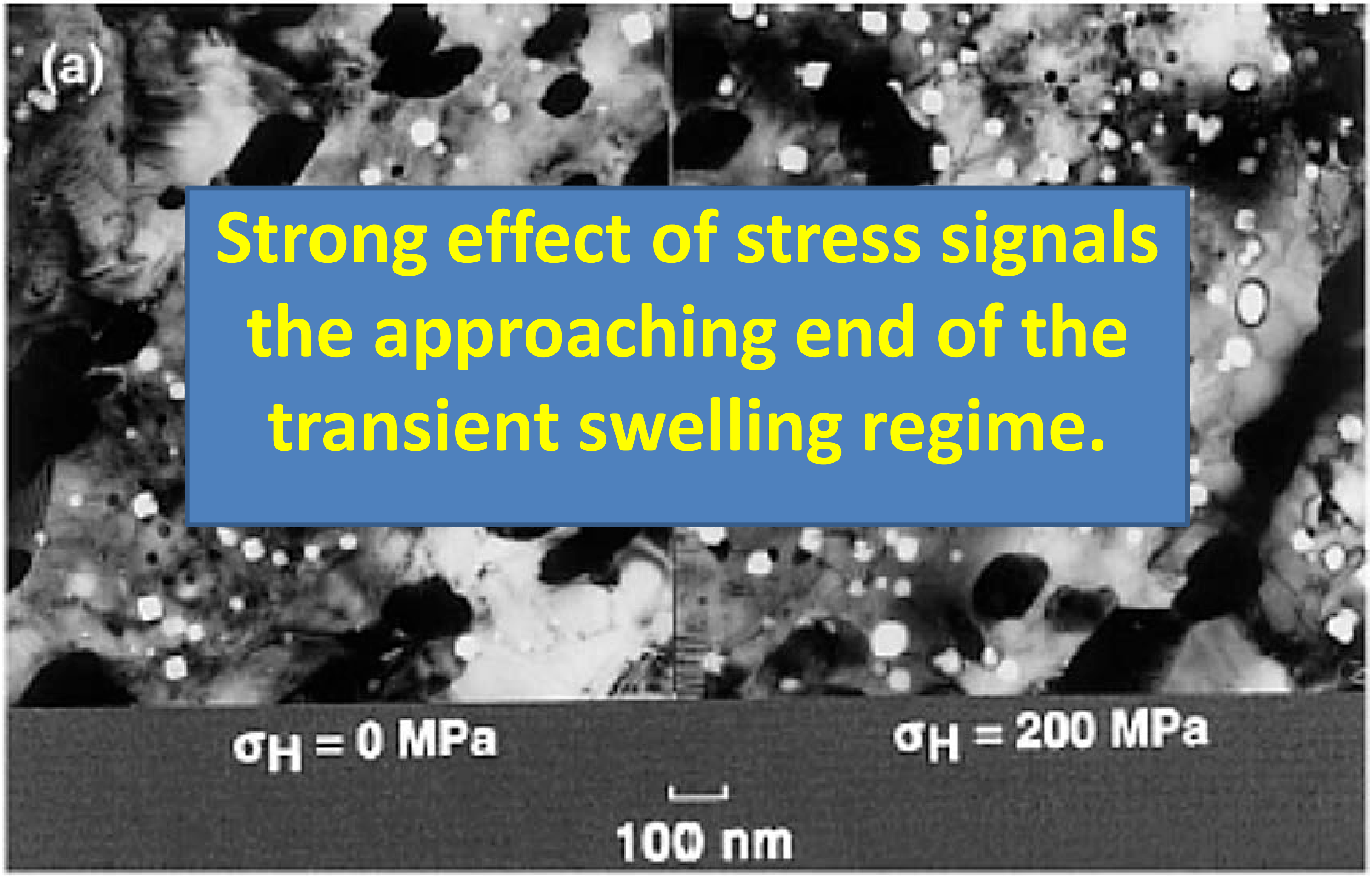
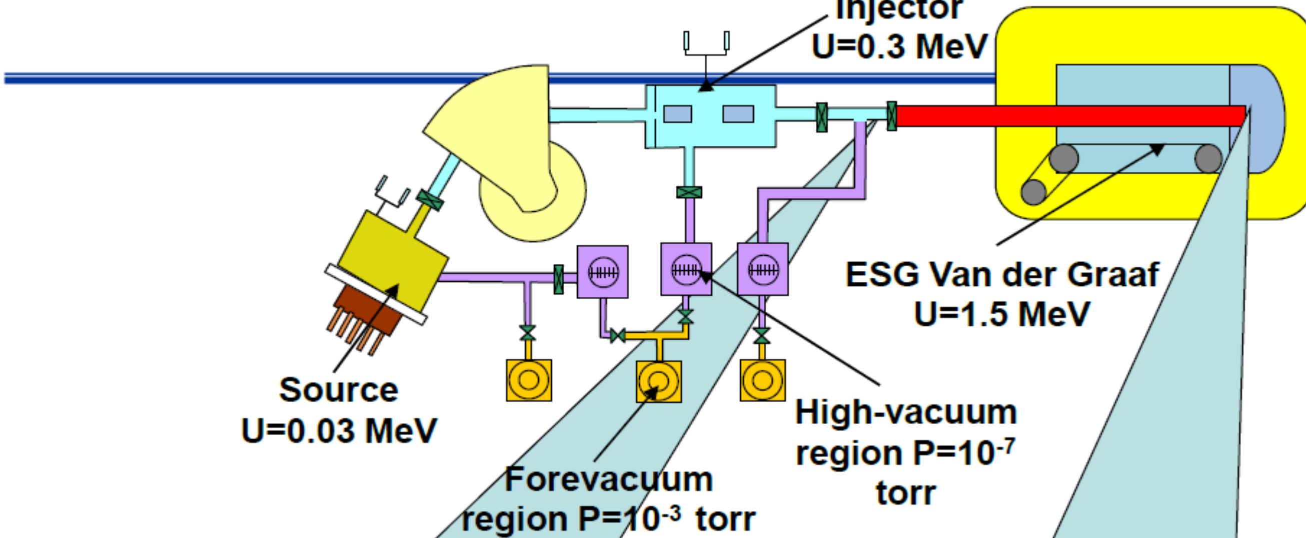


Strong effect of stress signals the approaching end of the transient swelling regime.

Electrostatic Accelerator with External Injector (ESUVI) at KIPT



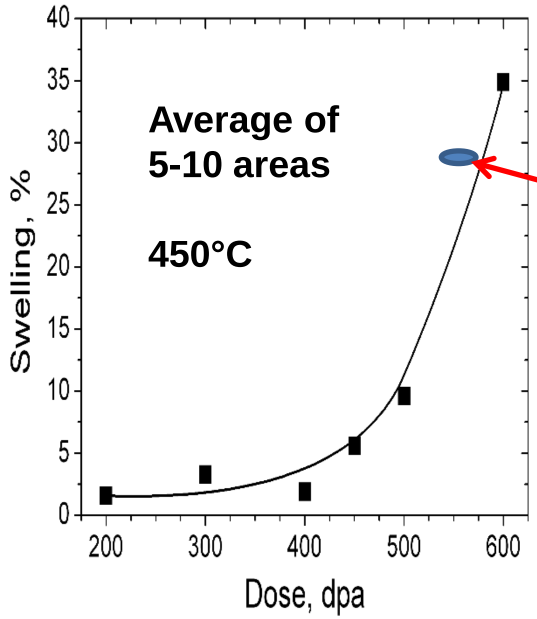
Group of
Borodin, Bryk,
Kalchenko,
Melnichenko,
Voyevodin

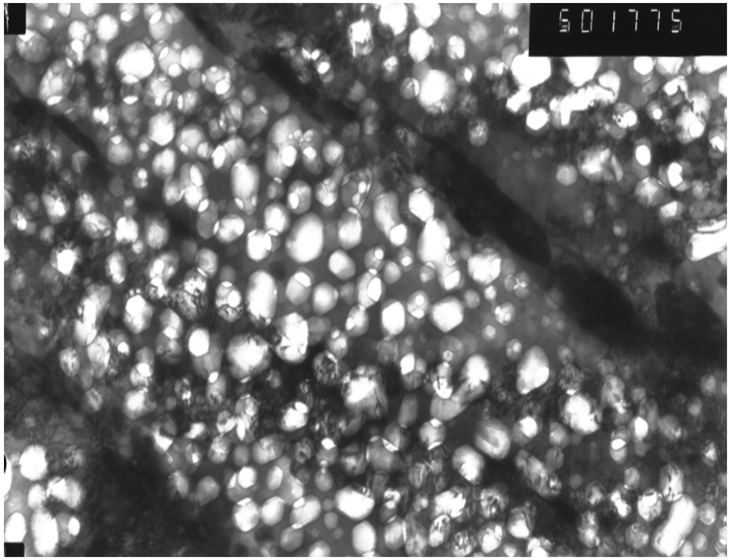
1.8 MeV Cr^{+}

Ion-induced swelling of 33% CW HT9 at 480°C at 200 to 600 dpa

Bryk, Borodin, Melnichenko, Kalchenko, Voyevodin, Garner, 2013

760°C/0.5hr + 33% CW





28% swelling at 450°C and 550 dpa

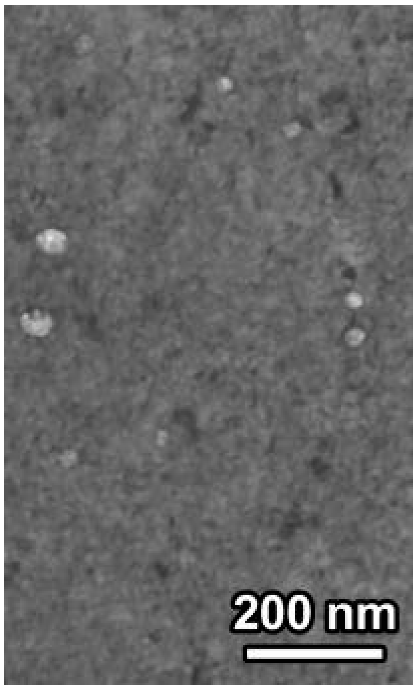
A high rate of void swelling develops after ~400 dpa.

Bi-linear swelling behavior

EP-450 after ion irradiation at 480ºC at ≥150 dpa

Voyevodin, Bryk, Borodin, Melnichenko, Kalchenko, Garner, 2012

150 dpa

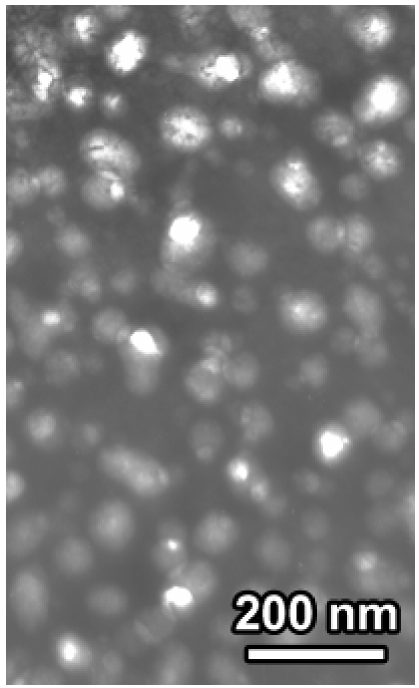


S = 0.6 %

$\langle d \rangle$ = 28 nm

$\rho = 5.0 \cdot 10^{14}\ cm^{-3}$

200 dpa

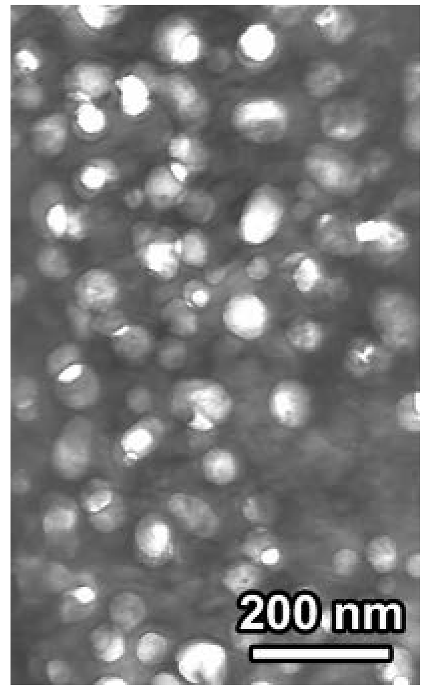


S = 6.9 %

$\langle d \rangle$ = 43 nm

$\rho = 1.5 \cdot 10^{15}\ cm^{-3}$

250 dpa

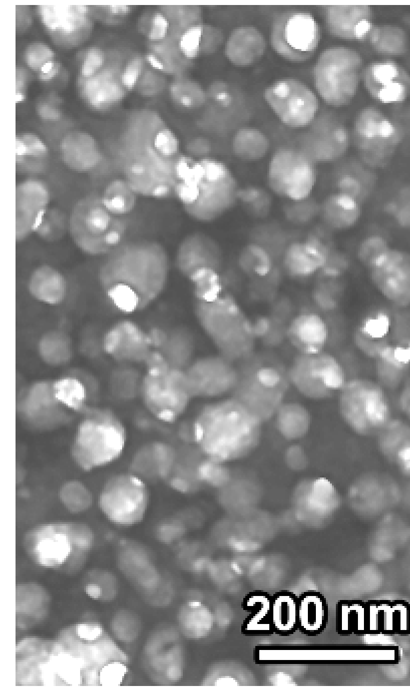


S = 13.3 %

$\langle d \rangle$ = 52 nm

$\rho = 1.8 \cdot 10^{15}\ cm^{-3}$

300 dpa



S = 20.4 %

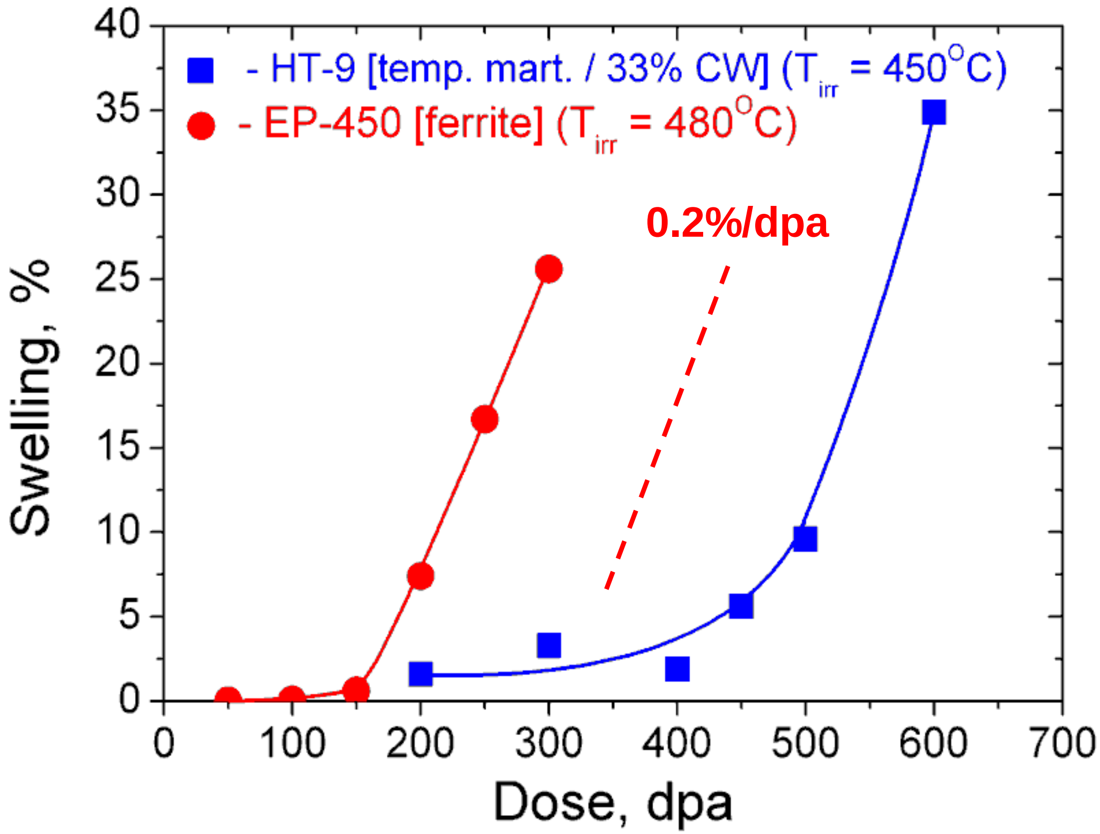
$\langle d \rangle$ = 58 nm

$\rho = 2.0 \cdot 10^{15}\ cm^{-3}$

Ferrite grains only are shown above.

Comparison of ion-induced swelling in two ferritic-martensitic alloys

Voyevodin, Bryk, Borodin, Melnichenko, Kalchenko, Garner, 2012

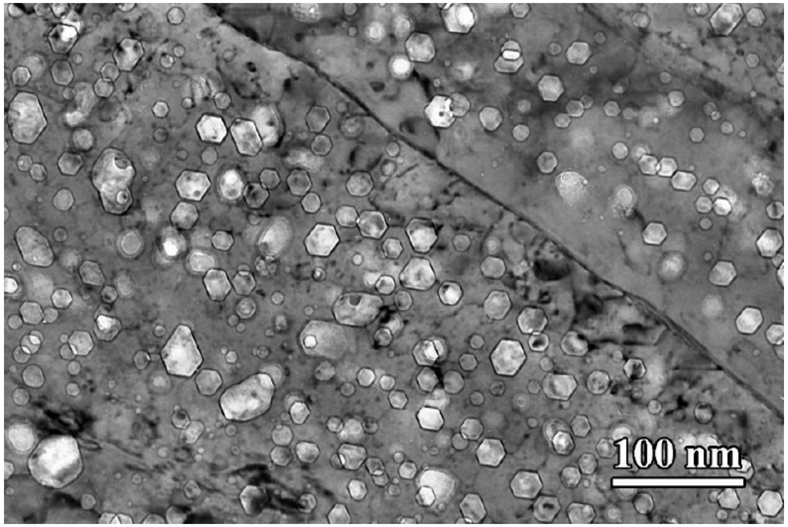


Iron-based bcc alloys appear to eventually swell at ~0.2%/dpa during ion irradiation.

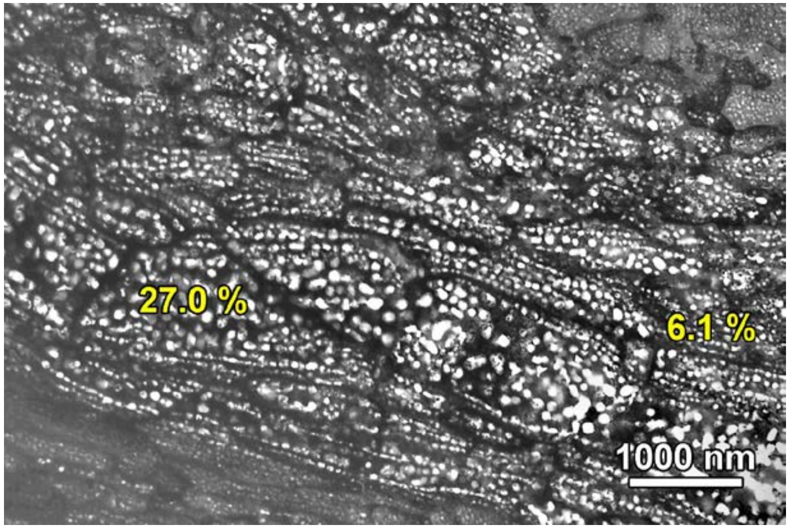
Similar results in other ion-irradiated alloys (unpublished)

Similar behavior seen in both EBR-II and FFTF fast reactors.

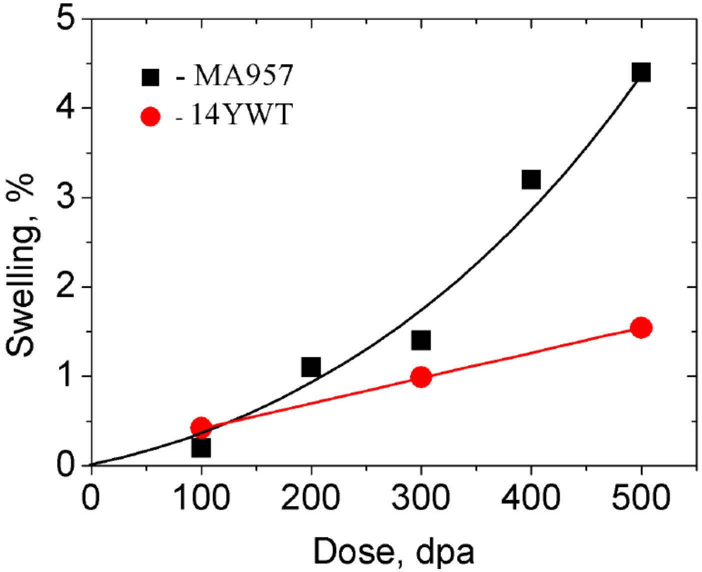
Ion-induced swelling at 450 °C and 500 dpa



MA 957



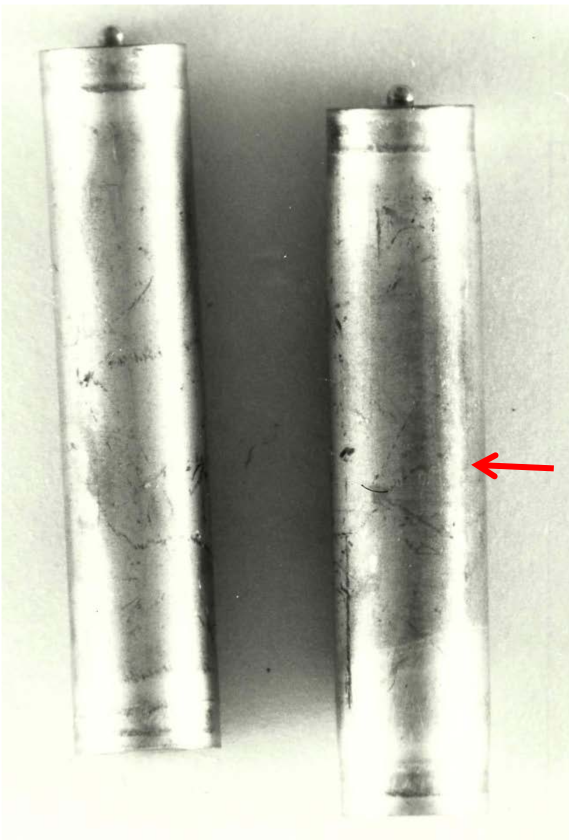
14YWT



ODS alloys have much longer transient regimes of swelling but eventually all iron-base alloys will begin to swell.

Temperature sensitivity of irradiation and/or thermal creep in FFTF-MOTA

D9 steel at 40 dpa, 520°C



0 MPa 146 MPa

~6% uniform strain

Gas-pressurized thin-wall tubes

Swelling is determined from diameter change of tube without stress.

Swelling plus creep contributes to diameter change of tube under stress.

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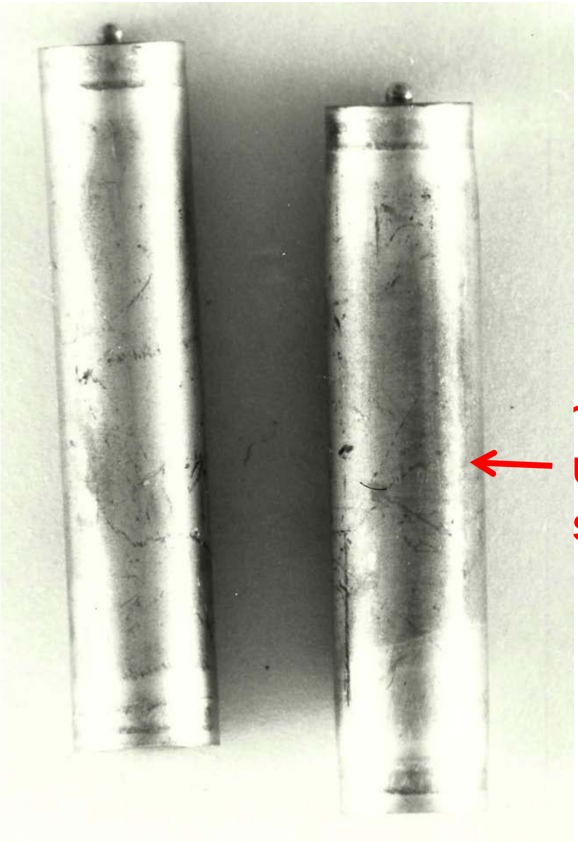


0 MPa 146 MPa

~6% uniform strain

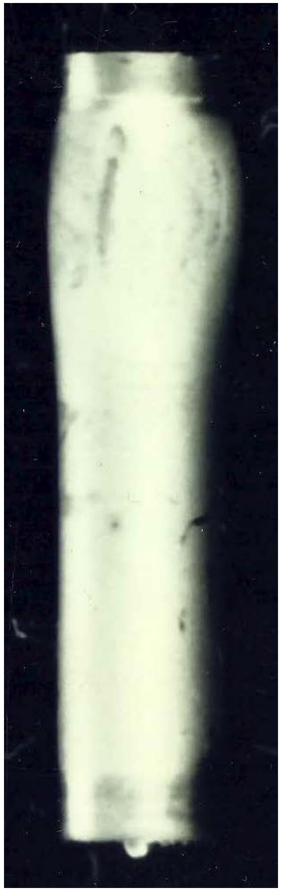
HT9

Irradiated at 45 Mpa and nominal temperature of 605°C

Top of tube ~30°C higher in temperature

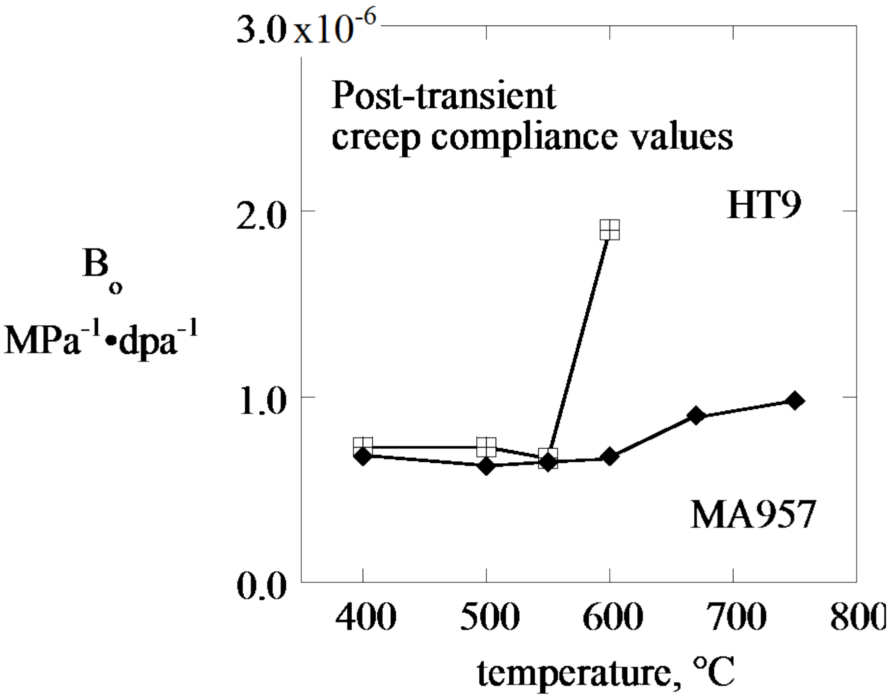
Average diametral strain of ~8%

Maximum strain ~25%



Temperature sensitivity of irradiation and/or thermal creep in FFTF-MOTA

Toloczko, Garner, Maloy, 2012



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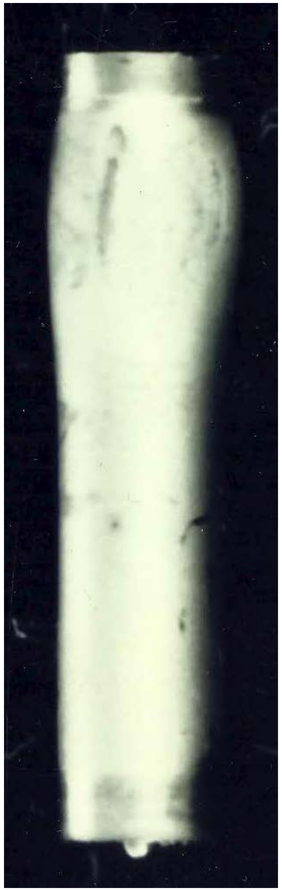

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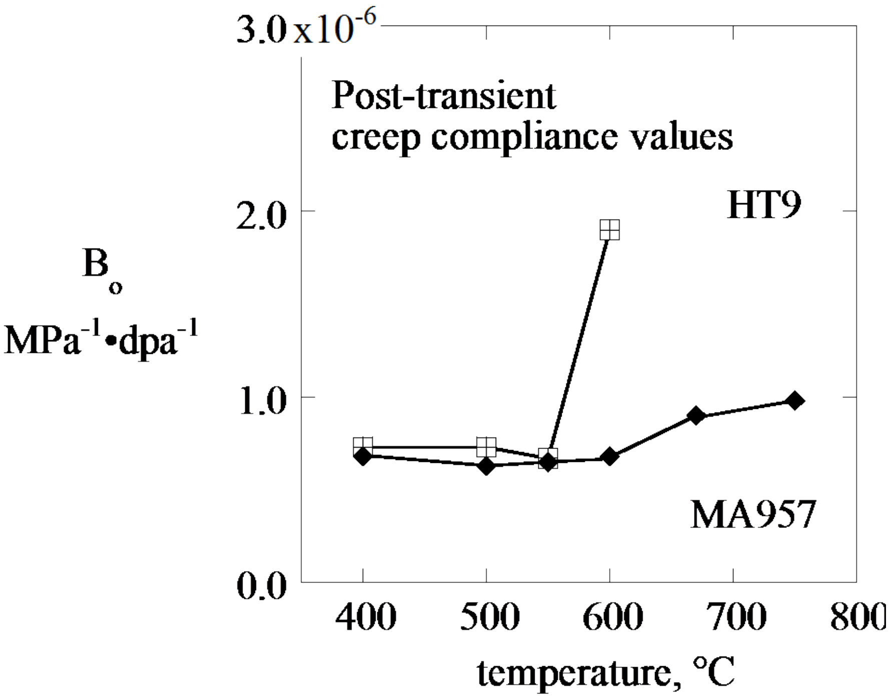

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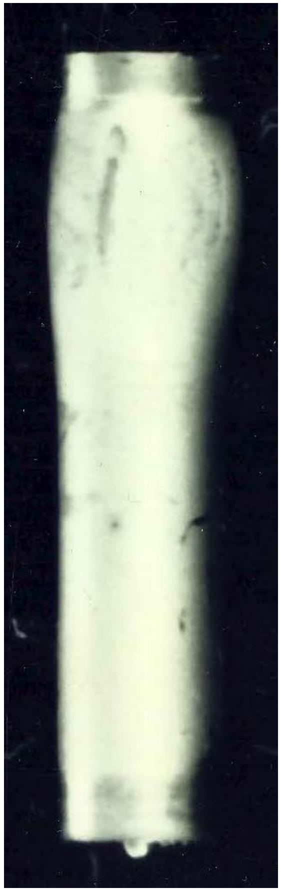
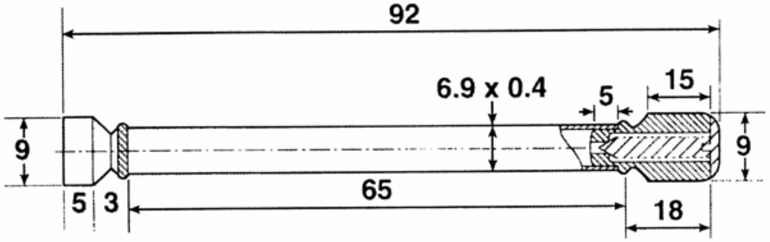


Note that the high dispersoid density does not reduce the creep rate!

Approach used in IPPE-Bochvar experiment on irradiation creep of F-M steels

- **Collect extensive data on EP-450 from four Soviet fast reactors to use as reference or "template" for F/M steels.**

All dimensions in mm

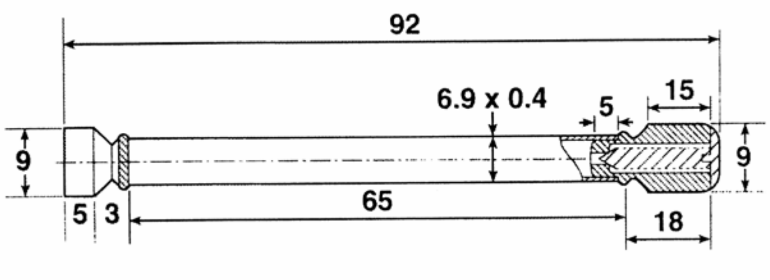


- **Argon-pressurized tubes and rings in flowing sodium.**
- **Diameter and density change measurements**
- **Ring-pull tests**
- **Microscopy**

Approach used in IPPE-Bochvar experiment on irradiation creep of F-M steels

- **Collect extensive data on EP-450 from four Soviet fast reactors to use as reference or "template" for F/M steels.**
- **Use "side-by-side" irradiation of EP-450, EP-823 and EP-852 at 280-520ºC in BN-350 to establish the relative behavior of the two high-silicon types of steels.**

All dimensions in mm



- **Argon-pressurized tubes and rings in flowing sodium.**
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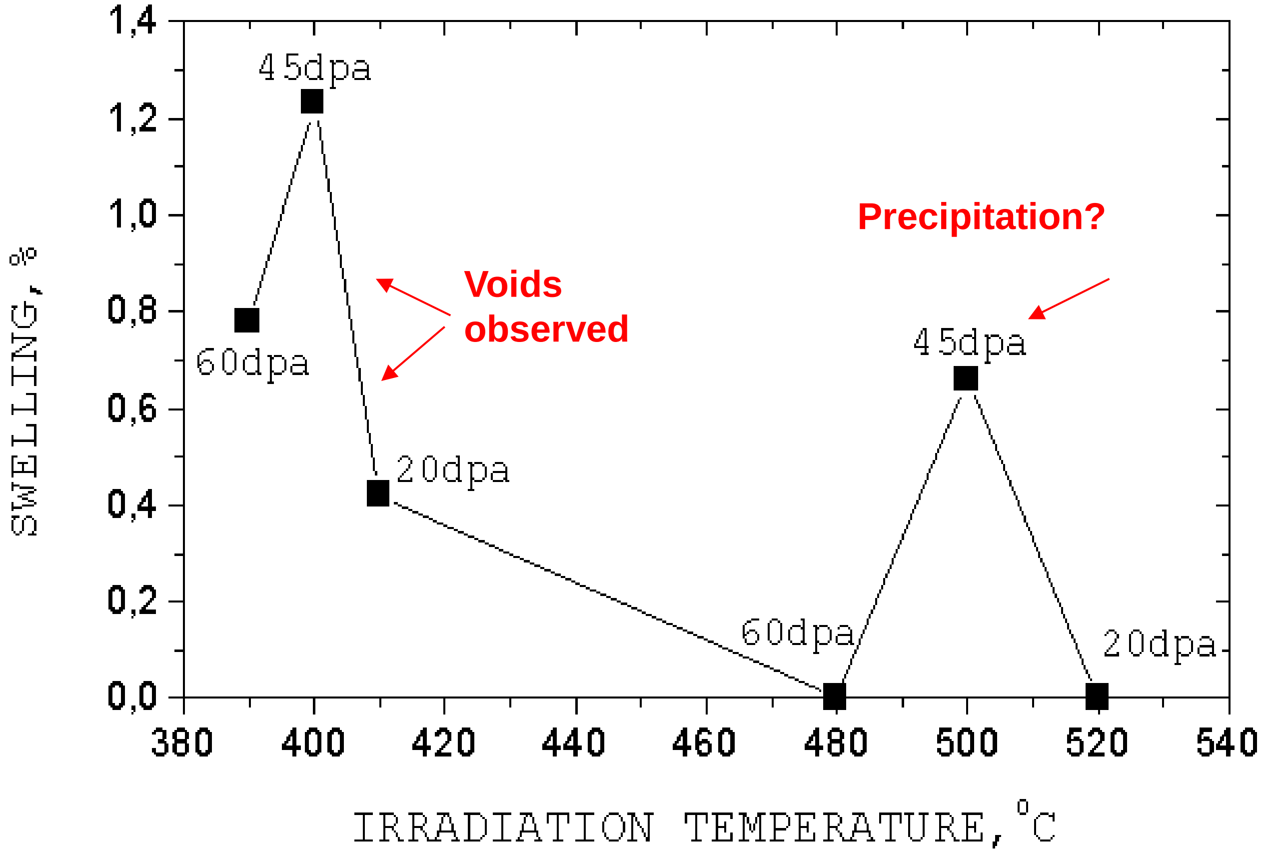
Extrapolation of expected behavior of the high-silicon steels from the <u>template</u> of the larger data base available on EP-450.

Swelling and/or phase-induced changes in density implied by stress-free strains observed in EP-450 pressurized tubes irradiated in BN-350

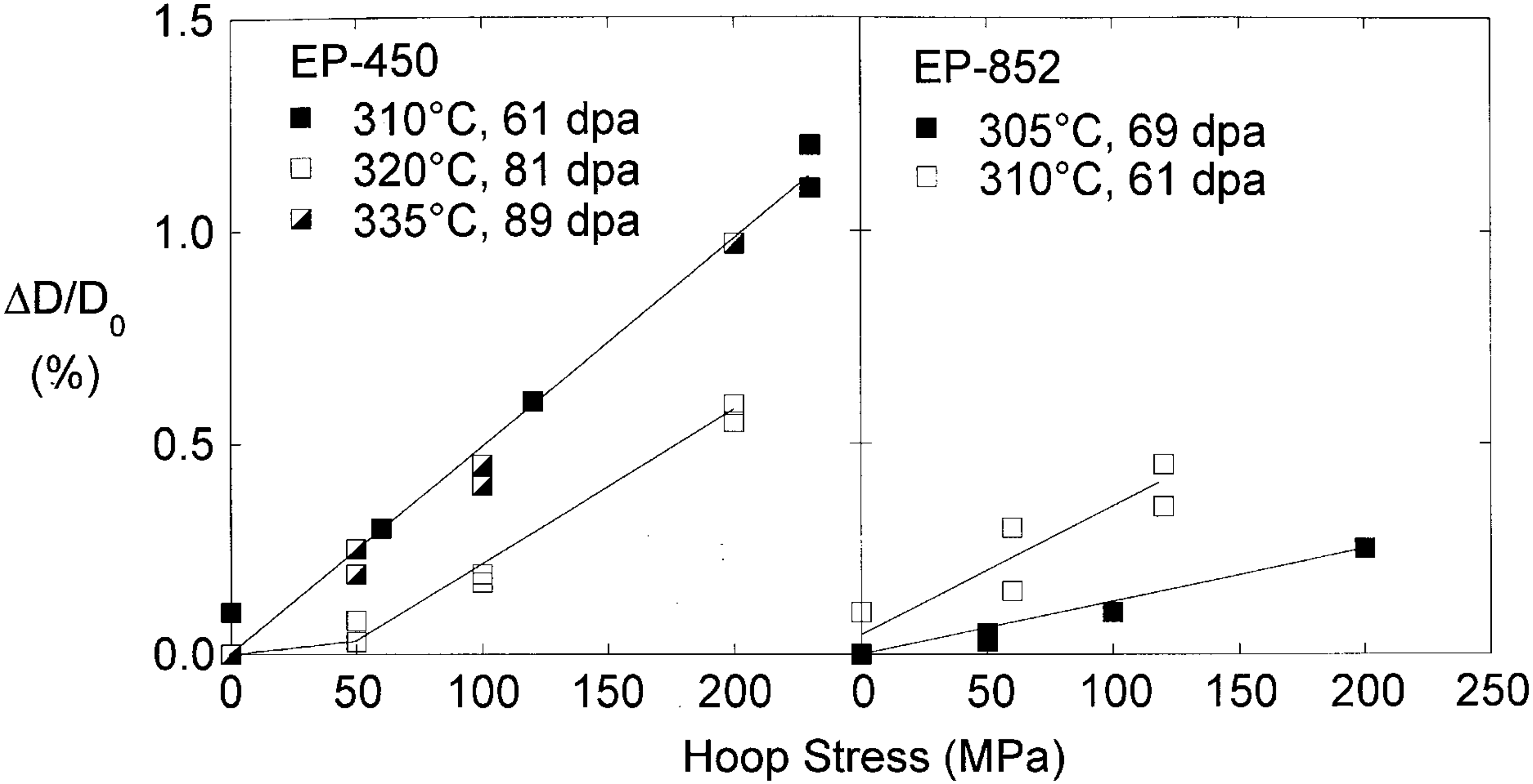
Shulepin, Dvoriashin, Ivanov, Konobeev, Porollo, Budylkin, Mironova, Garner, 2004



If flux effects are ignored, it appears that there may be two "swelling" peaks.

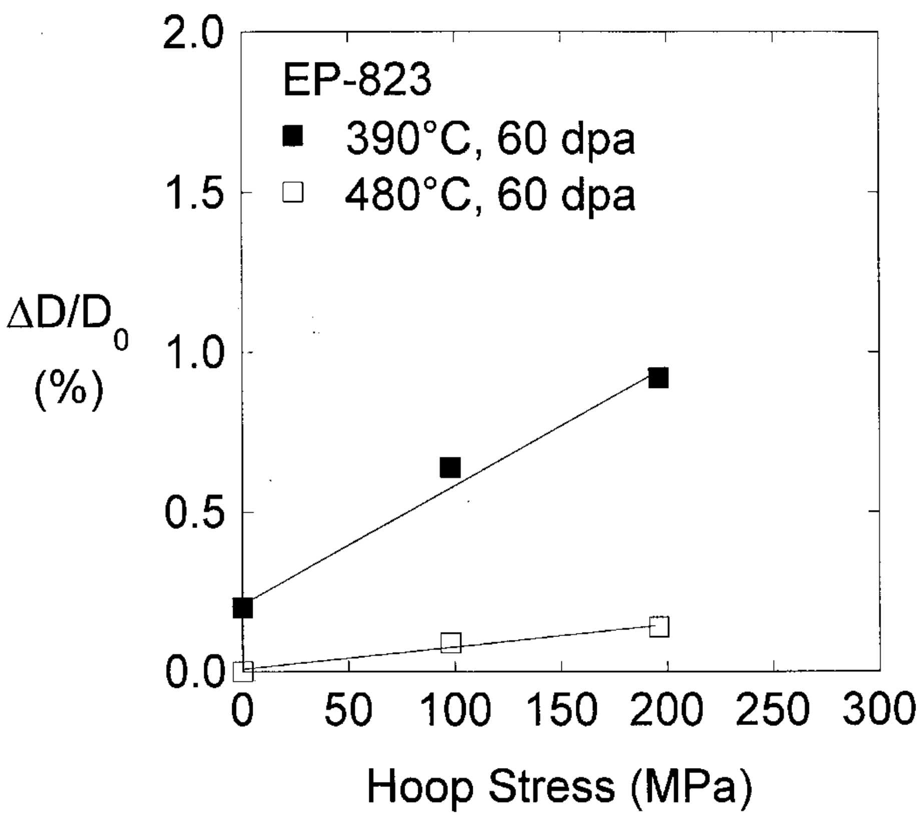
High temperature peak is probably due to phase instability.

Total strains in “low temperature” series of F/M steels after irradiation in BN-350



Creep coefficients on the order of 0.5-0.8 x 10^{-6} (MPa dpa)$^{-1}$

Total strains in "high temperature" series of F/M steels after irradiation in BN-350

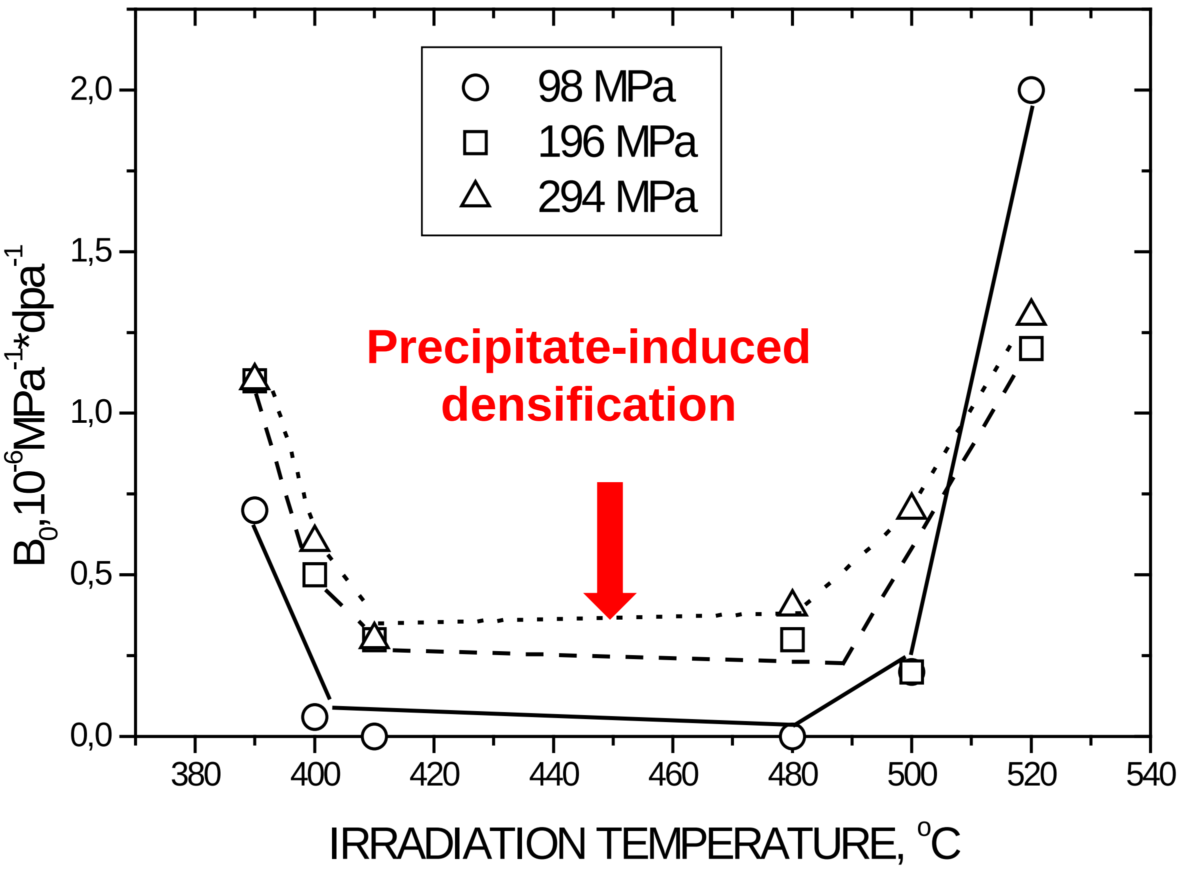


Total strains may include

- **densification or dilation from precipitates**
- **transient creep**
- **irradiation creep**
- **swelling-enhanced creep**
- **swelling**
- **thermal creep**

Creep coefficients observed in EP-450 in “high temperature” series at 380-520ºC

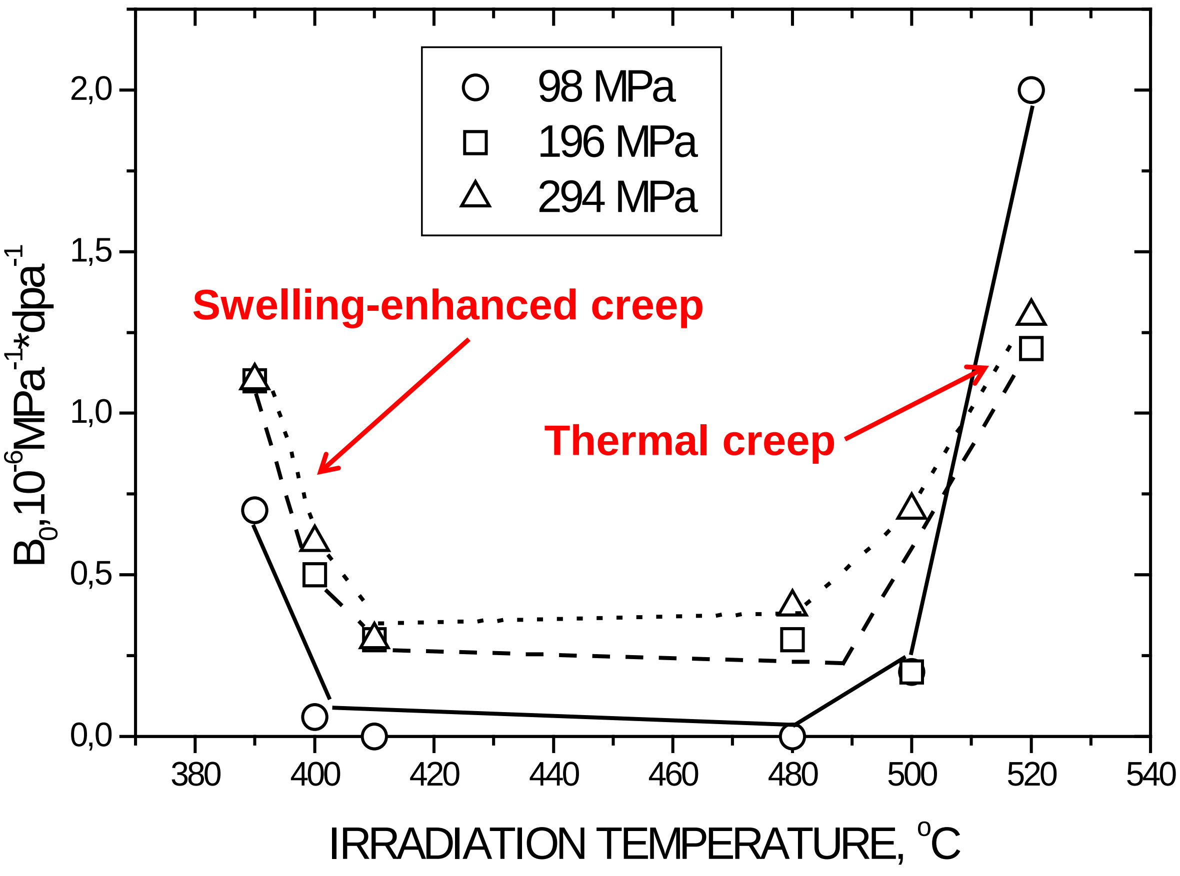
Shulepin, Dvoriashin, Ivanov, Konobeev, Porollo, Budylkin, Mironova, Garner, 2004



- **Contributions from densification at all temperatures,**

Creep coefficients observed in EP-450 in "high temperature" series at 380-520ºC

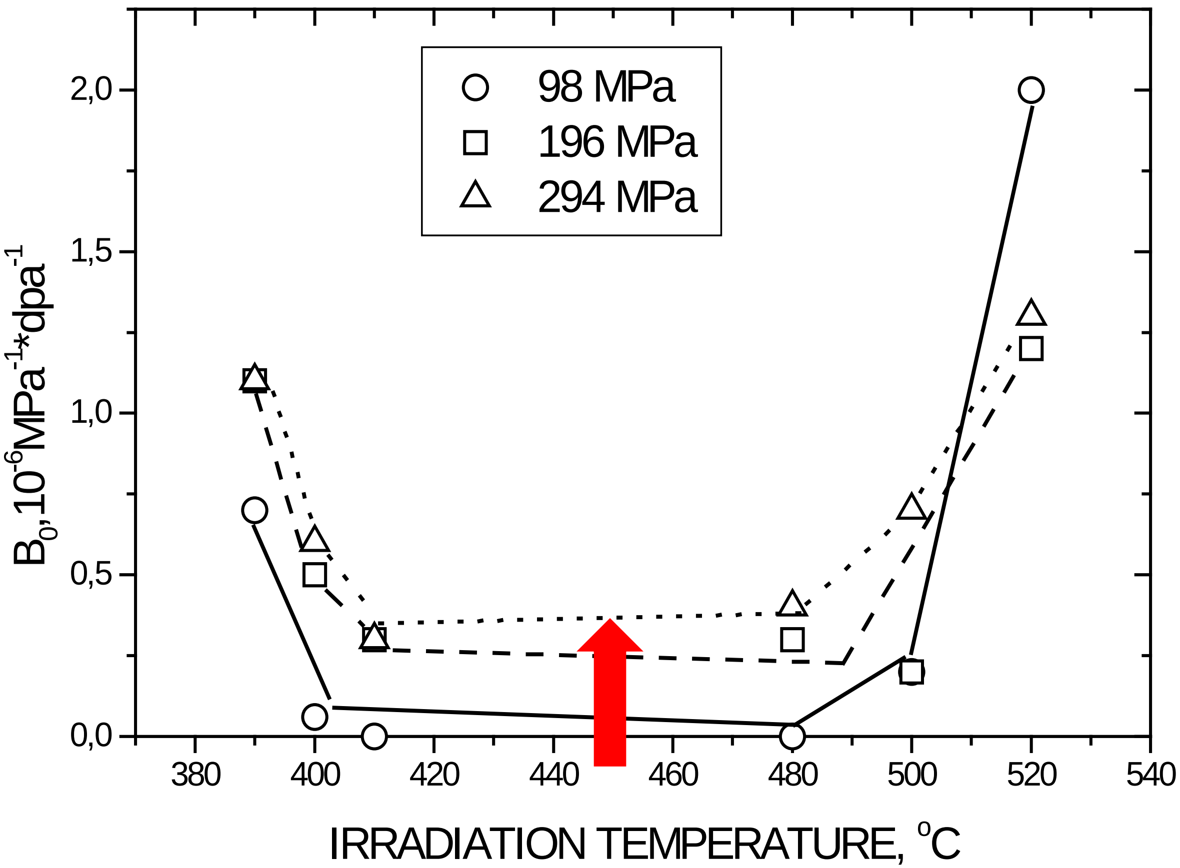
Shulepin, Dvoriashin, Ivanov, Konobeev, Porollo, Budylkin, Mironova, Garner, 2004



- **Contributions from densification at all temperatures,**
- **void swelling at low temperatures,**
- **and thermal creep at high temperatures.**

Creep coefficients observed in EP-450 in "high temperature" series at 380-520ºC

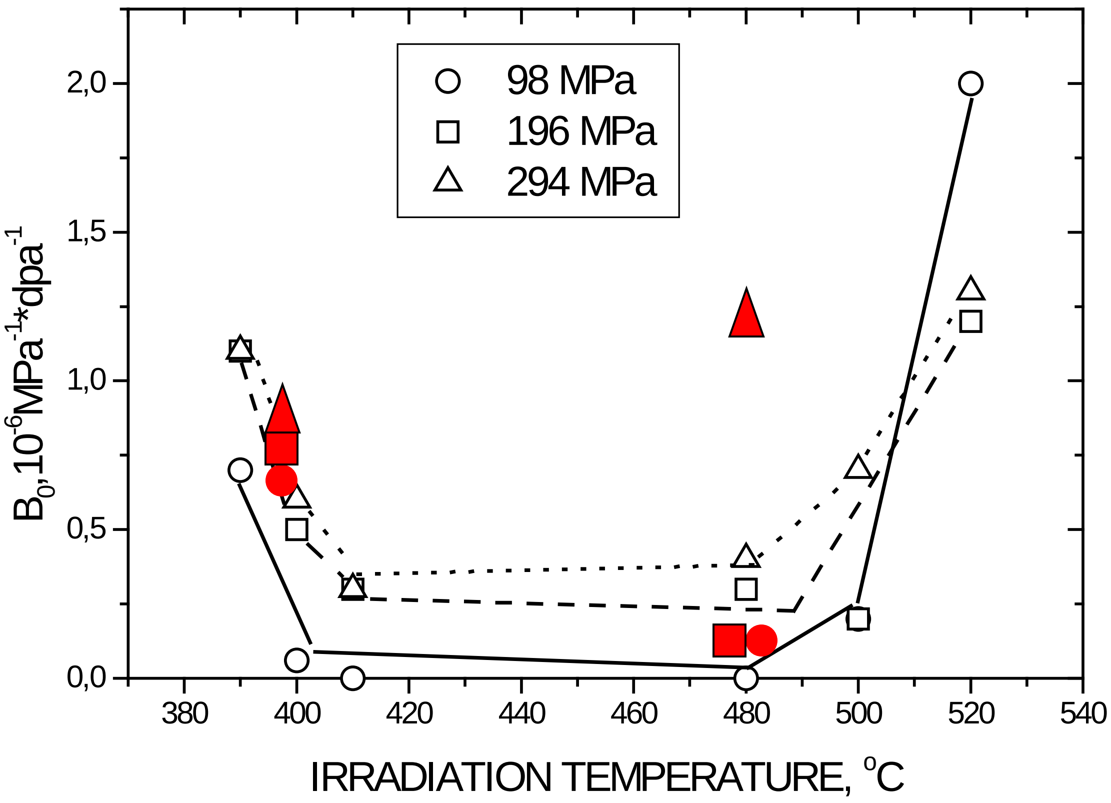
Shulepin, Dvoriashin, Ivanov, Konobeev, Porollo, Budylkin, Mironova, Garner, 2004



Convergence to creep coefficient B = ~0.5 x 10^{-6} at higher stress level reflects decreasing <u>fraction</u> of densification strain.

Creep coefficients observed in EP-450 and EP-823 in BN-350 at 380-520ºC

Shulepin, Dvoriashin, Ivanov, Konobeev, Porollo, Budylkin, Mironova, Garner, 2004



EP-823 results are shown in red.

Creep coefficients are in general similar except for highest stress at 480ºC, suggesting earlier onset of thermal creep in EP-823.

Conclusions

- **Ferritic-martensitic (F-M) steels are superior to austenitic steels for fast reactor service, with lower swelling and irradiation creep compared to austenitic steels. Upper temperature limit due to thermal creep.**

- **For reduction of corrosion in lead-bismuth for F-M steels, higher silicon addition has been used.**

Conclusions

- Ferritic-martensitic (F-M) steels are superior to austenitic steels for fast reactor service, with lower swelling and irradiation creep compared to austenitic steels. Upper temperature limit due to thermal creep.

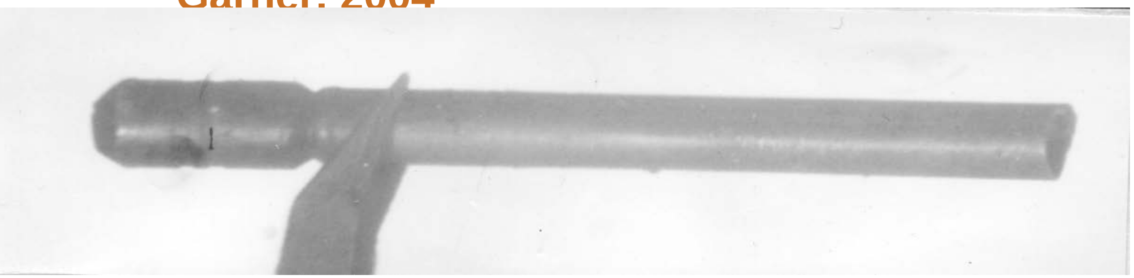
- For reduction of corrosion in lead-bismuth for F-M steels, higher silicon addition has been used.

- There is no indication that the creep and swelling behavior of the high-silicon steels is fundamentally different from that of EP-450 or HT9.

- For service less than ~200 dpa, ferritic-martensitic steels with high silicon will probably provide acceptable behavior concerning dimensional stability.

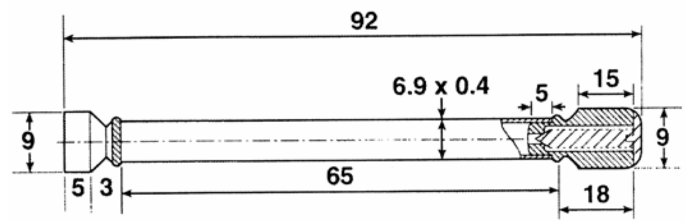
Embrittlement of F-M steels after irradiation in BN-350 at 310° C to 61 dpa

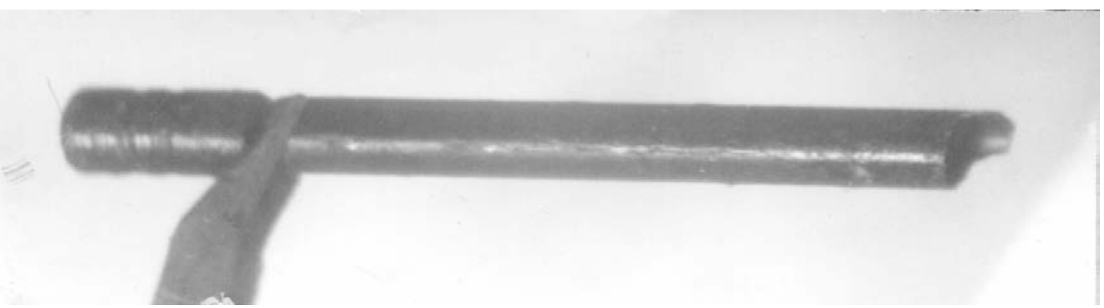
Shulepin, Dvoriashin, Ivanov, Konobeev, Porollo, Budylkin, Mironova, Garner, 2004



EP-450, 100 MPa

All dimensions in mm





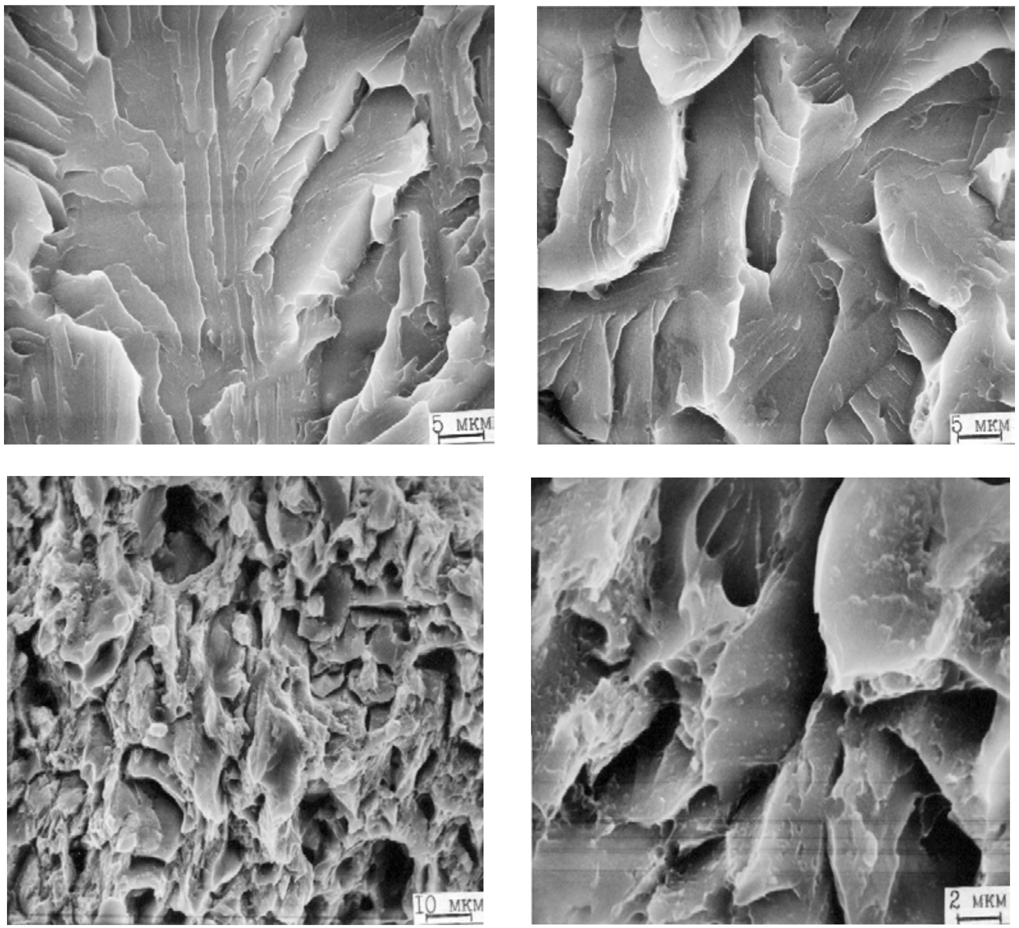
EP-852, 100 MPa

Ductile-to-brittle transition at low temperatures has been a major concern for F/M steels.

During post-irradiation puncturing of pressurized creep tubes to release the fill gas, a number of tubes failed in very brittle manner.

Failure surfaces of specimens cut from pressurized tubes irradiated in BN-350

Shulepin, Dvoriashin, Ivanov, Konobeev, Porollo, Budylkin, Mironova, Garner, 2004



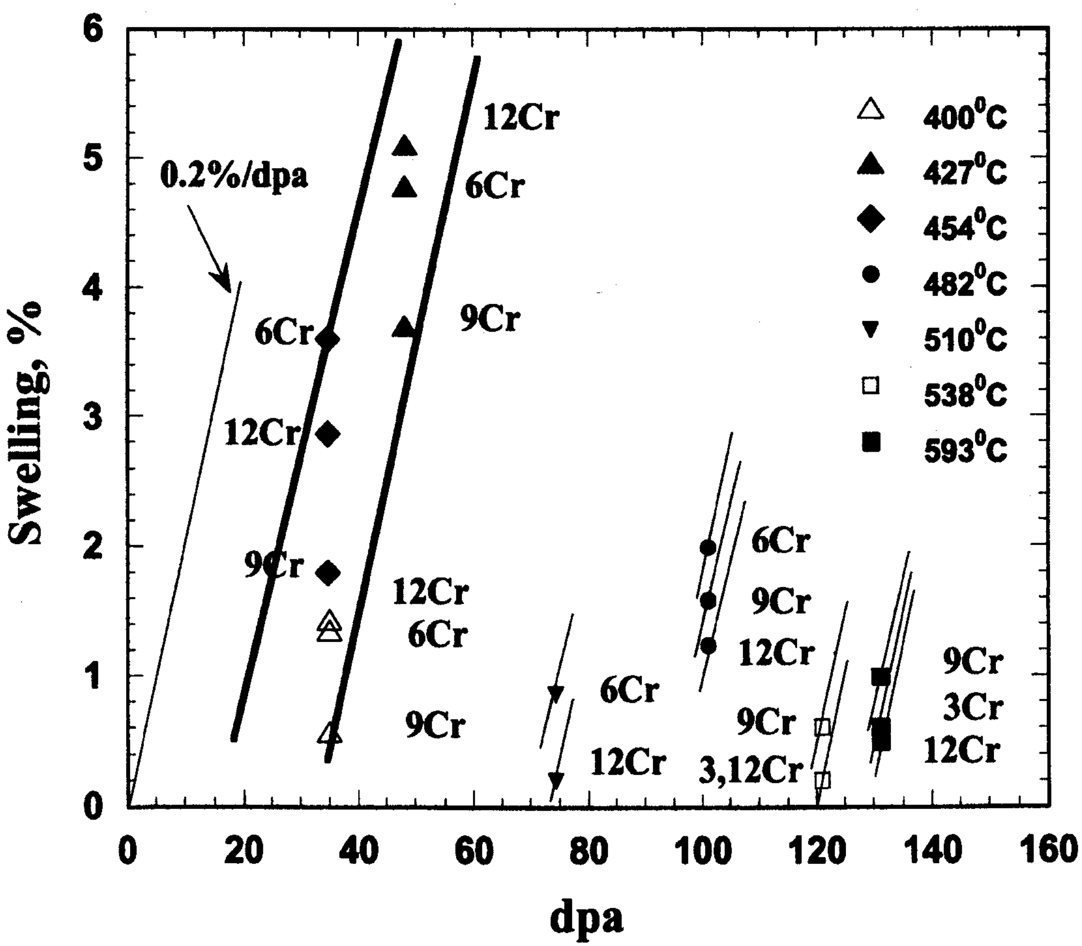
Ring-pull tensile test

EP-852 at 305°C and 69 dpa, at hoop stresses of 0 and 100 MPa.

EP-450 at 310°C and 61 dpa at hoop stress of 60 MPa.

Swelling of binary Fe-Cr alloys irradiated in EBR-II

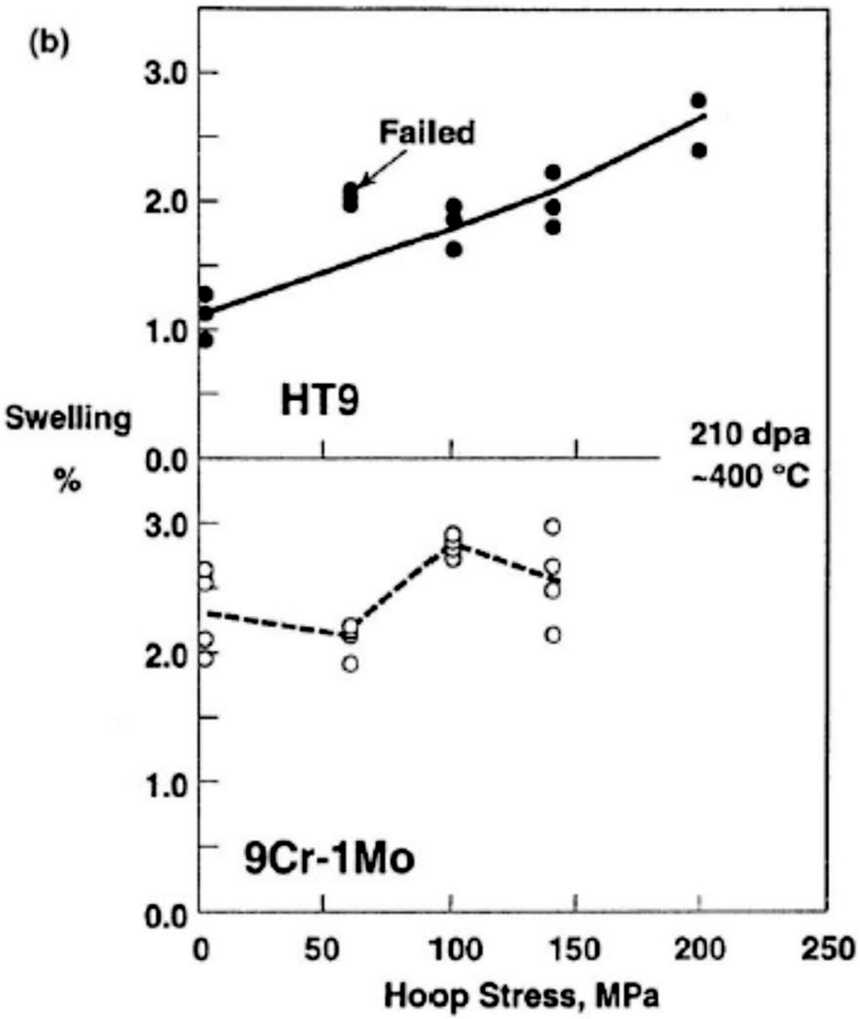
Sencer and Garner, 1999



At 400-457°C the post-transient swelling rate is ~0.2%/dpa.

The transient duration becomes longer with increasing temperature or with increasing dpa rate.

Swelling measured by density change in HT9 and 9Cr-1Mo pressurized tubes at 208 dpa and ~400° C



Toloczko, Garner and Eiholzer, 1994

Swelling of HT9 increases with stress at 400°C.

Similar stress-affected behavior is almost always observed in austenitic alloys .

Stress operates on incubation period only.

Strong effect of stress signals the approaching end of the transient swelling regime.