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Improved Ductility in Tempered Martensitic Steels after Low Temperature Irradiation

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Outline

- **Advanced Fuels for Transmutation**
- **Cladding for High Burnup Fuels**
- **Tempered Martensitic Steels**
  - Previous work on irradiation hardening at low temperatures
  - ATR irradiation tensile data
  - Previous work on UE vs. dose at low temperatures
  - Elemental content of alloys
  - Effects of interstitial content on mech properties in Fe-Cr alloys
  - Annealing effects in Ferritic/martensitic alloys
- **Summary and Future Work**

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Advanced Fuels Campaign Mission & Objectives in the Fuel Cycle Research and Development Program

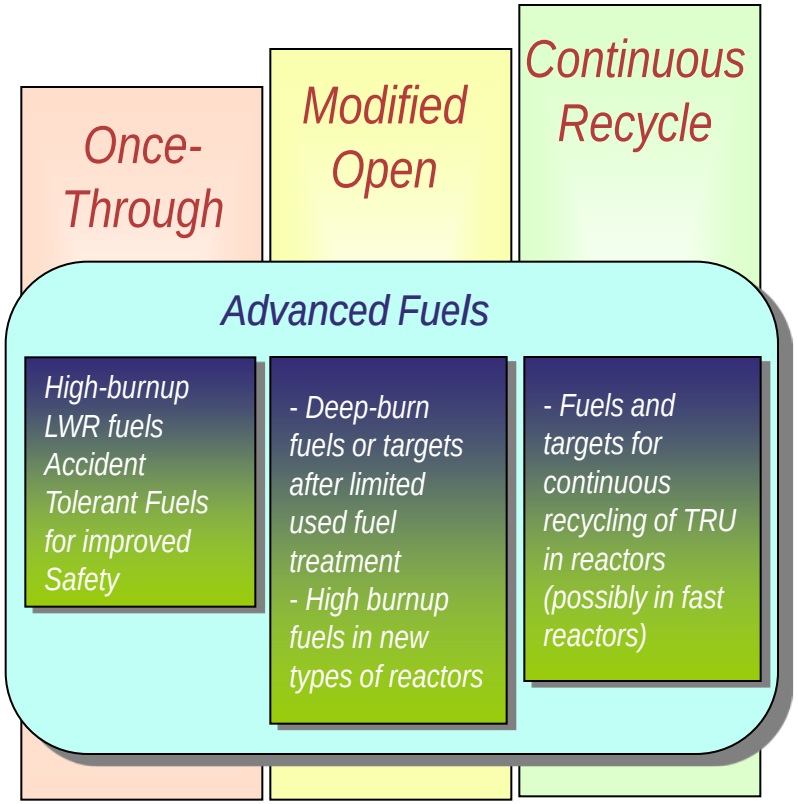
- Mission

  Develop and demonstrate fabrication processes and in-pile (reactor) performance of advanced fuels/targets (including the cladding) to support the different fuel cycle options defined in the NE roadmap.

- Objectives

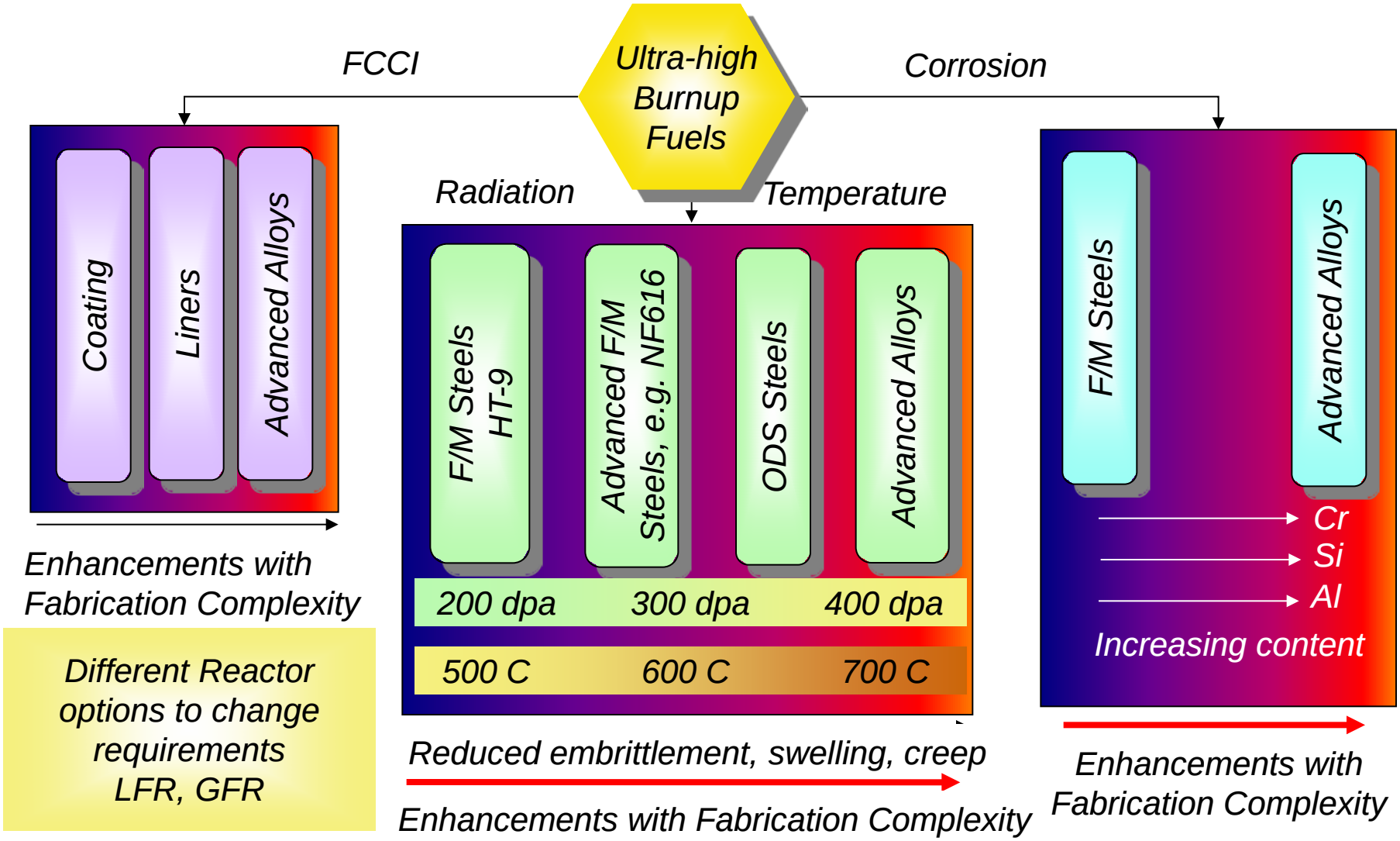
  Development of the fuels/targets that
  - Increases the efficiency of nuclear energy production
  - Maximize the utilization of natural resources (Uranium, Thorium)
  - Minimizes generation of high-level nuclear waste (spent fuel)
  - Minimize the risk of nuclear proliferation

- Grand Challenges
  - Multi-fold increase in fuel burnup over the currently known technologies
  - Multi-fold decrease in fabrication losses with highly efficient predictable and repeatable processes



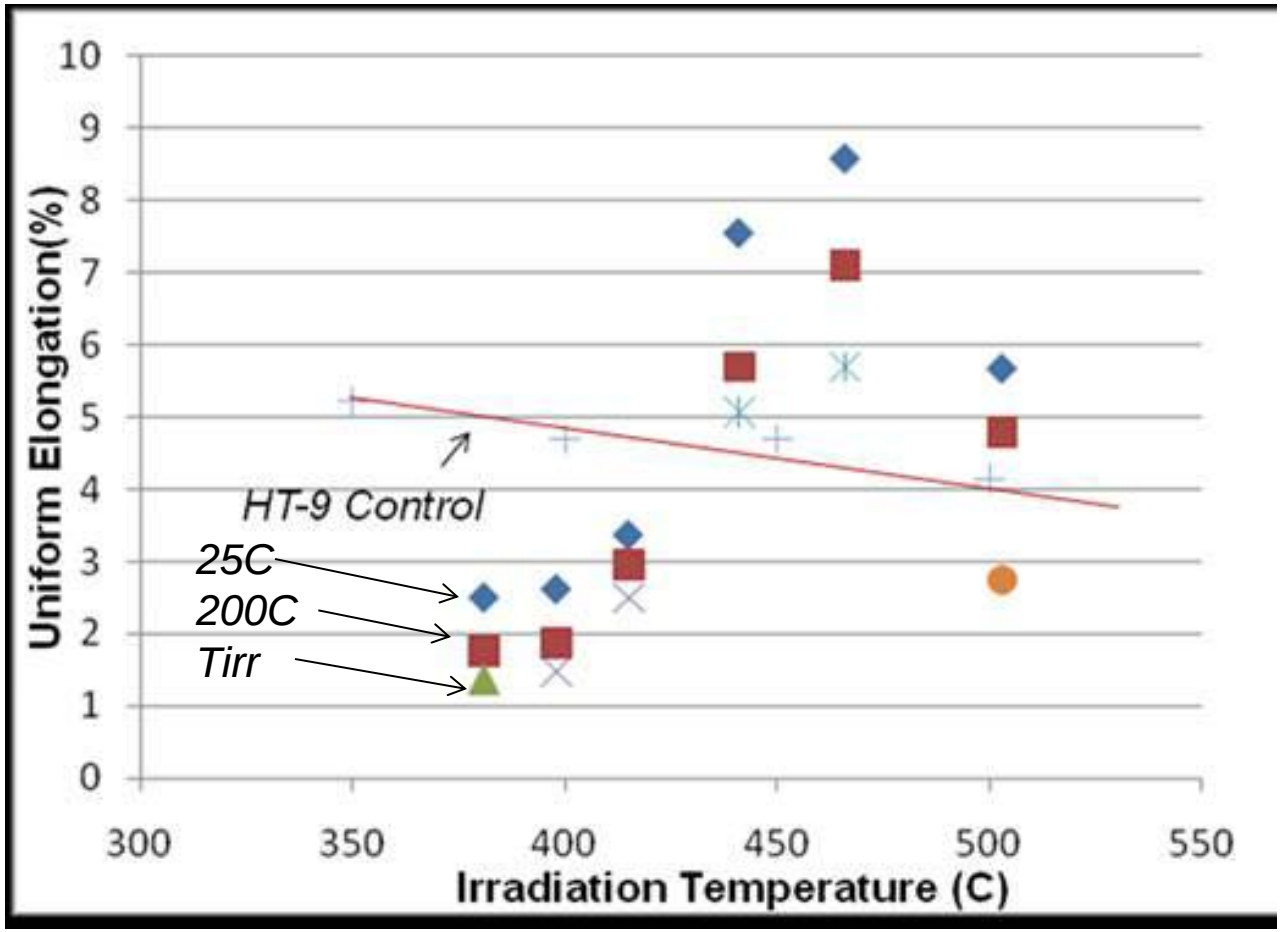
Approach to Enabling a Multi-fold Increase in Fuel Burnup over the Currently Known Technologies

Ultimate goal: Develop advanced materials immune to fuel, neutrons and coolant interactions under specific reactor environments



Loss of Strain Hardening Capacity needs to be Improved in Tempered Martensitic Steels

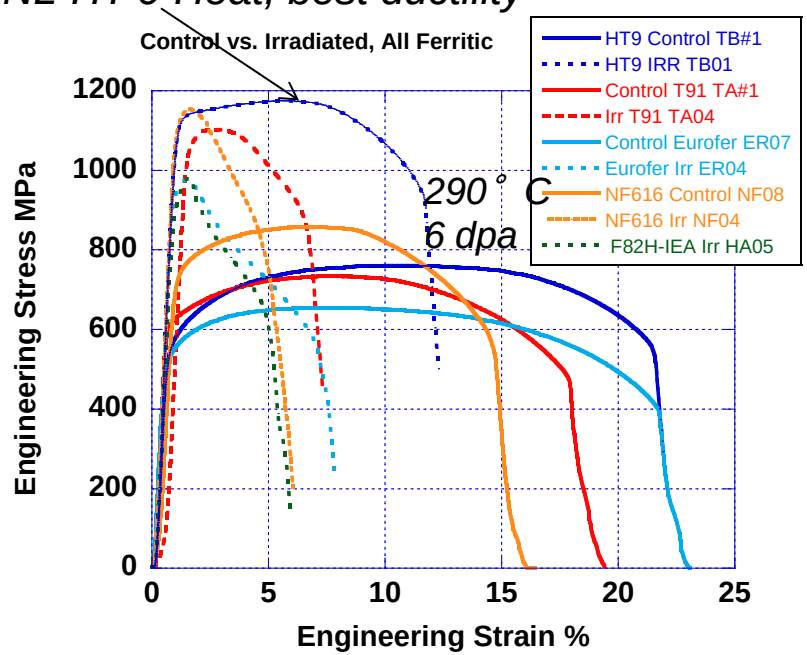
- Uniform elongation decreases with testing temperature
- Irradiation temperatures below 430 ° C result in lower uniform elongation
- UE are reduced to ~1% at Tirr



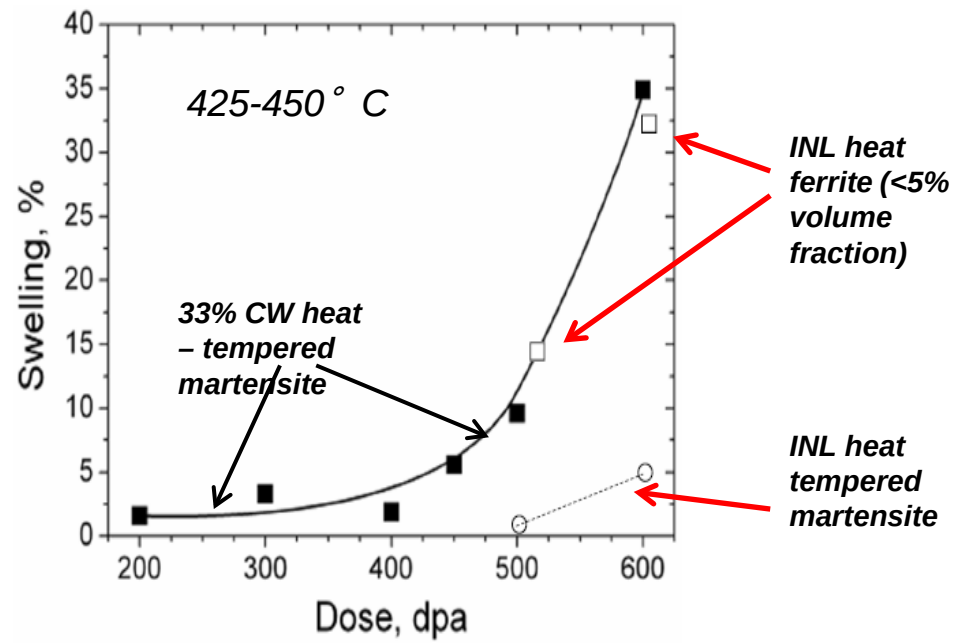
Improved Radiation Response of New NQA1 Heat of HT-9

- **300 lb heat of HT-9 produced by Metalwerks following NQA-1 quality control**
- **Tensile specimens irradiated in INL Advanced Test Reactor to 6 dpa at 290° C**
  - Hardening observed but excellent ductility retained after low temperature irradiation
- **Ion irradiations performed to 600 dpa at 425° C**
  - Minimal swelling observed in tempered martensitic grains after ion irradiation to >500 dpa.

INL-HT-9 Heat, best ductility



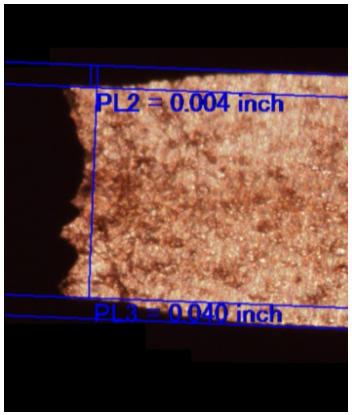
INL (NSUF), LANL, UCSB



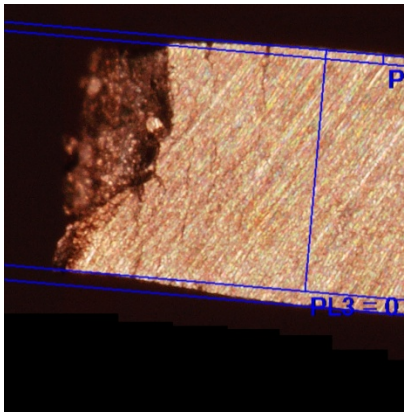
PNNL, REC, KIPT, LANL

Reduction of Area Measurements

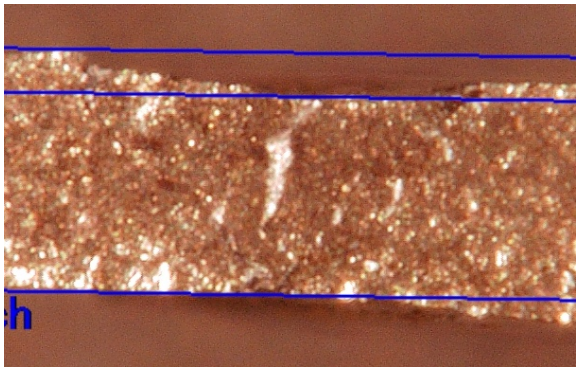
- **HT-9 heat retains UE and reduction of area after irradiation to 6 dpa at 290 C.**
- **In addition, less cracking observed near fracture surface compared to T91 and NF616.**



HT-9



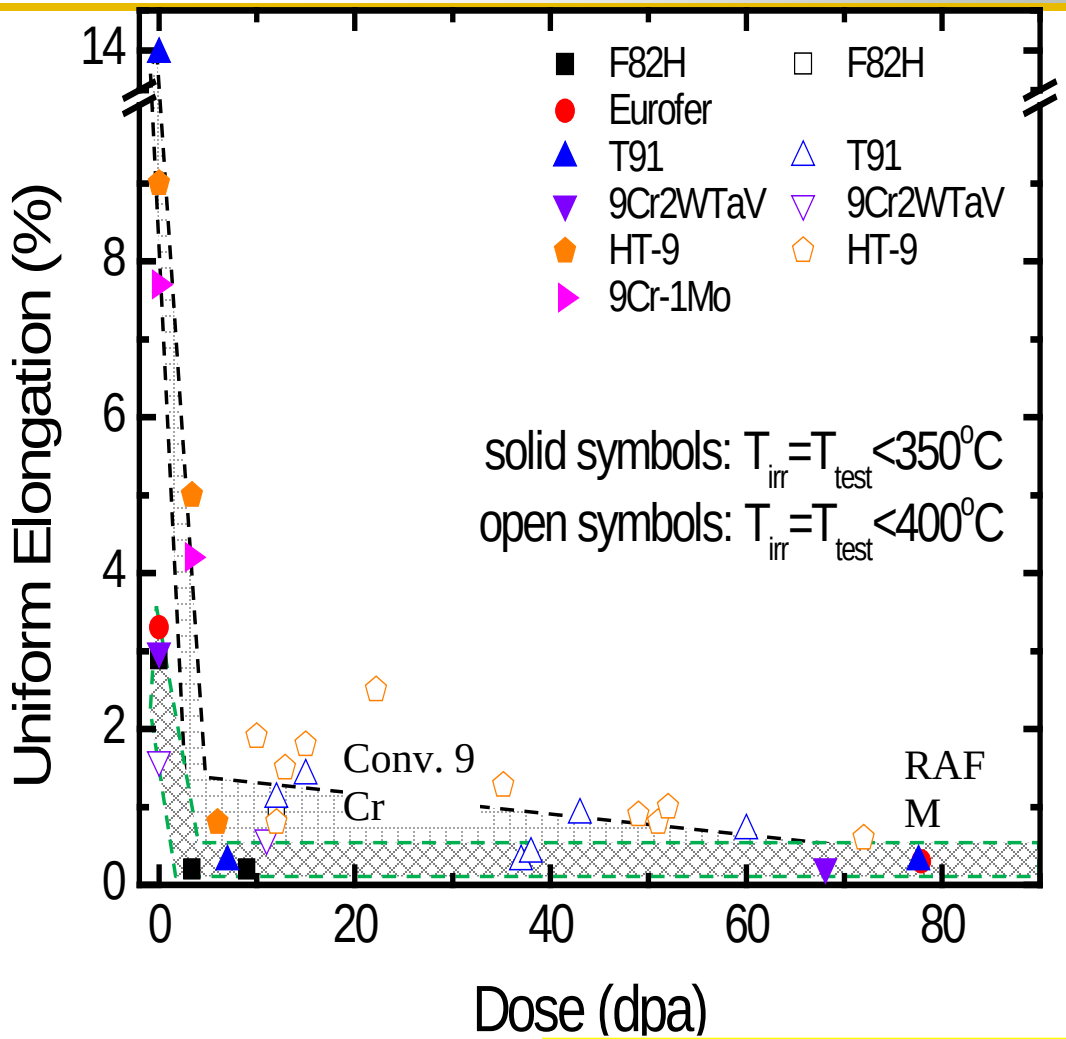
T91



NF616

Material	ID	Type	Yield MPa	UTS MPa	Uniform Elongation %	Total Elongation %	Reduction in Area %
HT9	TB#1c	Control	560	761	9.15	21	55.03
HT9	TB01	Irradiated	1100	1175	4.54	10.9	46.22
T91	TA04	Irradiated	1055	1102	1.07	5.7	39.03
NF616	NF04	Irradiated	1120	1154	0.65	4.7	23.72

Previous Results showing Reduction of Ductility in irradiated F/M steels



Anderoglu, O., Byun, T. S., Toloczko, M. and Maloy, S. A. Mechanical Performance of Ferritic Martensitic Steels for High Dose Applications in Advanced Nuclear Reactors. Metallurgical and Materials Transactions a-Physical Metallurgy and Materials Science, 44A(Jan 2013), 70-83.

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Exact Elemental Analysis on Control Materials

Alloy	C	Cr	Mn	Ni	Si	Mo	Nb	V	W	O	N	P	S	Al	Cu	Co	Ti	Fe
HT-9	.201	12.49	.41	.60	.28	1.07	<.002	.29	.52	.002	.001	.007	<.0005	.015	.034	-	-	Bal
Eurofer97	.117	8.69	.47	.024	.056	.005	<.002	.20	.82	.003	.023	.004	.002	.009	.023	.011	.006	Bal
F82H	.093	7.89	.16	.026	.12	.005	<.002	.16	1.21	.003	.008	.004	.002	.002	.028	.007	.002	Bal
NF616	.108	9.71	.46	.064	.056	.47	.043	.20	1.22	.003	.060	.007	.001	.003	.035	.015	.003	Bal
T91	.052	9.22	.46	.18	.24	.96	.063	.24	.013	.002	.057	.016	.001	.009	.087	.021	.002	Bal

LANL, UCSB

Questions

- *Why does HT-9 have improved ductility compared to other F/M steels with lower Cr content?*
  - *Is it related to interstitial content?*
    - *C and N are known to affect formability and localized deformation (Luder's band formation) in ferritic steels*
    - *Low Temperature irradiation embrittlement is generally linked to localized deformation.*
  - *Is it an experimental anomaly?*
    - *All samples still show high hardening*
    - *Irradiation of all samples were in same holder.*
  - *Other possible reasons?*
    - *HT-9 has higher initial strain hardening capacity*

Typical reasons for low UE after Low Temperature Irradiation: Localized flow

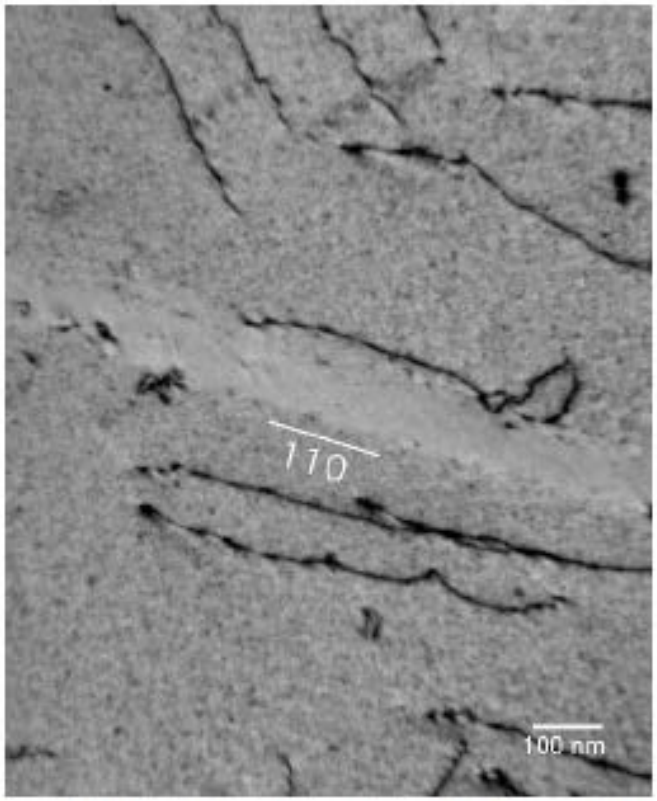


Fig. 5. Defect-free channel in the deformed proton-irradiated Fe–12Cr alloy at room temperature. Imaging conditions: $g = \langle 1\,1\,0 \rangle$, $z = \langle 1\,1\,1 \rangle$.

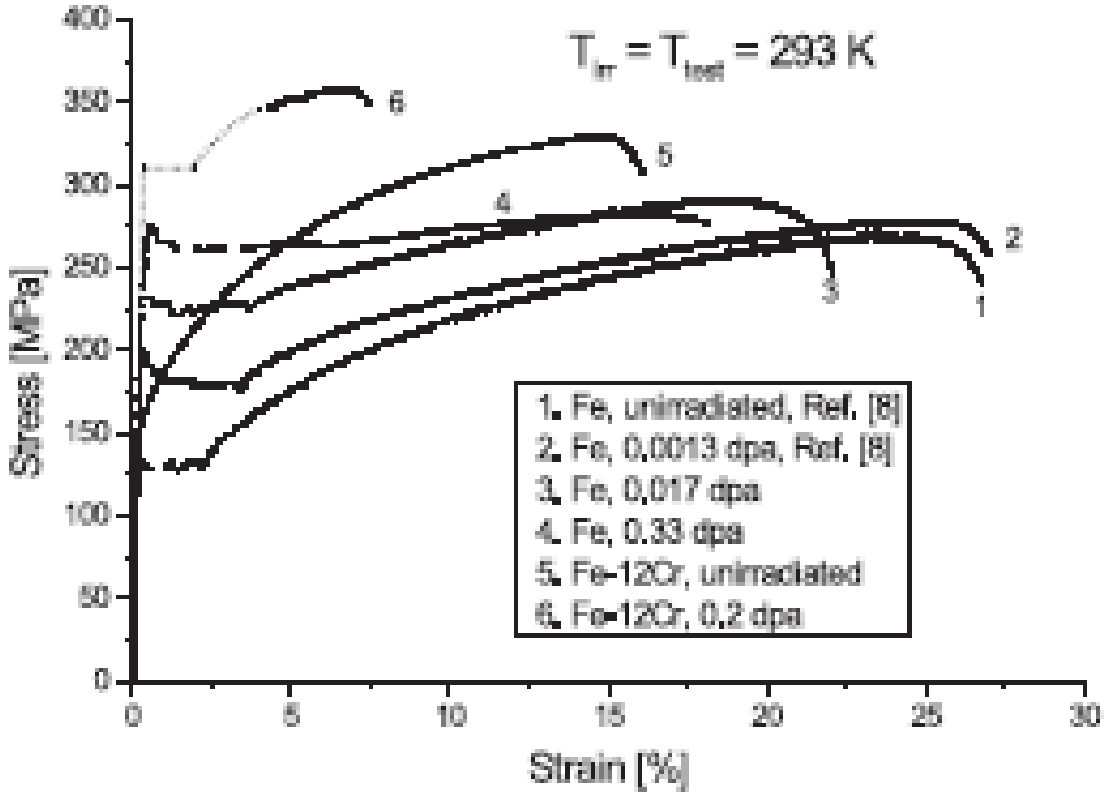


Fig. 1. Tensile stress–strain curves of unirradiated and proton-irradiated specimens of Fe and Fe–12Cr alloy at room temperature.

M.I. Luppo et al. / Journal of Nuclear Materials 283–287 (2000) 483–487

Effects of Interstitial content on Luder's band formation in Ferritic steels

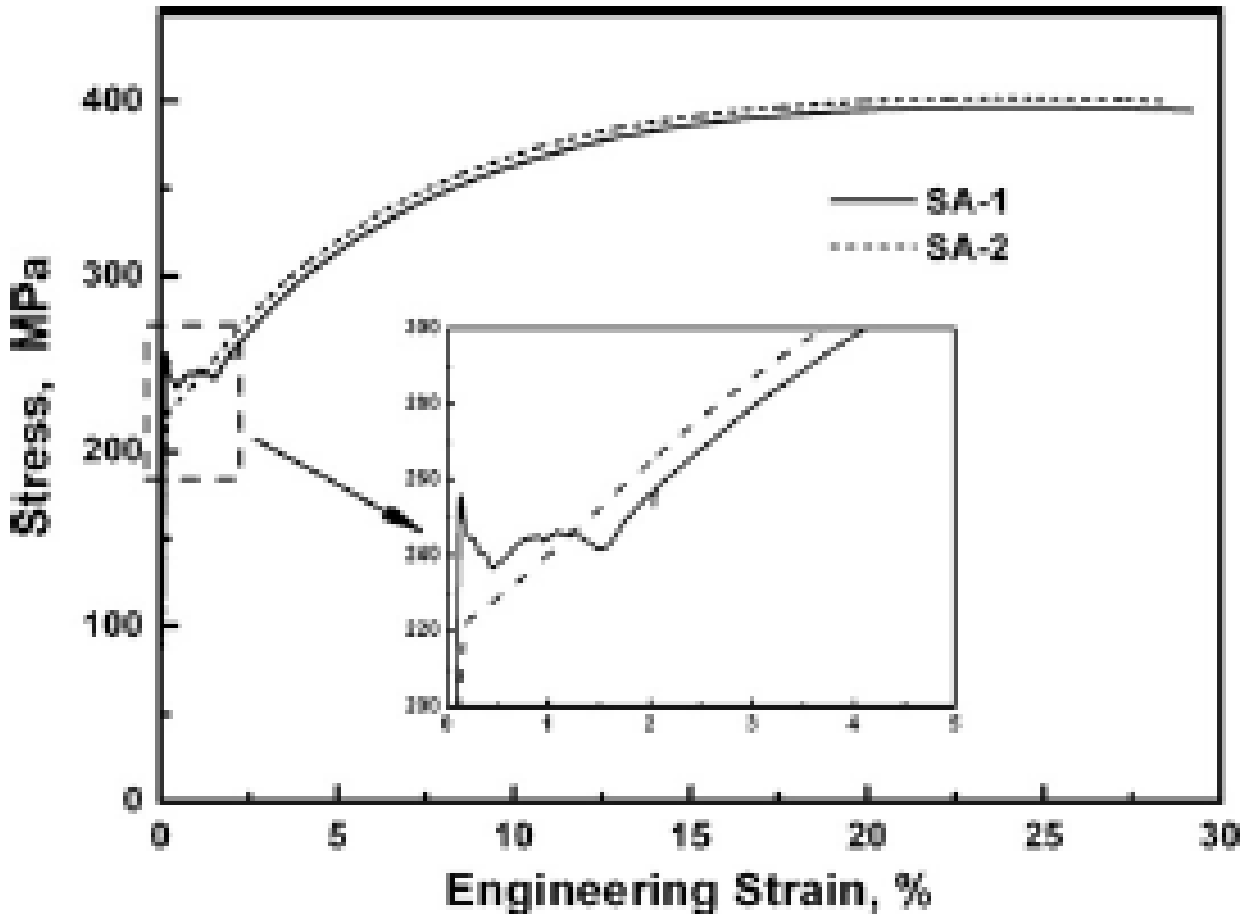


Fig. 3. The stress–strain curve for the specimen along the rolling direction of the experimental steel after different annealing treatments at the strain rate of 0.001 s^{-1}.

Z.Y. Liu et al. / Materials Science and Engineering A 527 (2010) 3800–3806

Impact of C + N on properties of Fe-Cr Steels

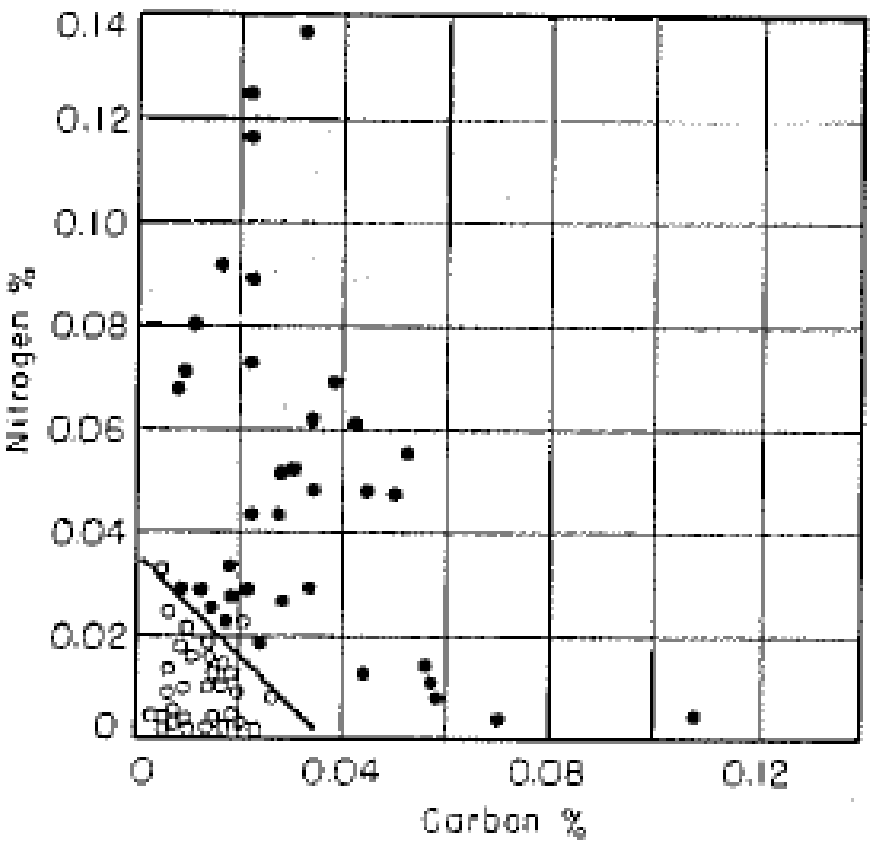


Fig. 6—Effect of Carbon and Nitrogen on the Toughness of 24 to 26% Chromium-Iron Alloys. Open circles—high impact strength alloys; solid circles—low impact strength alloys.

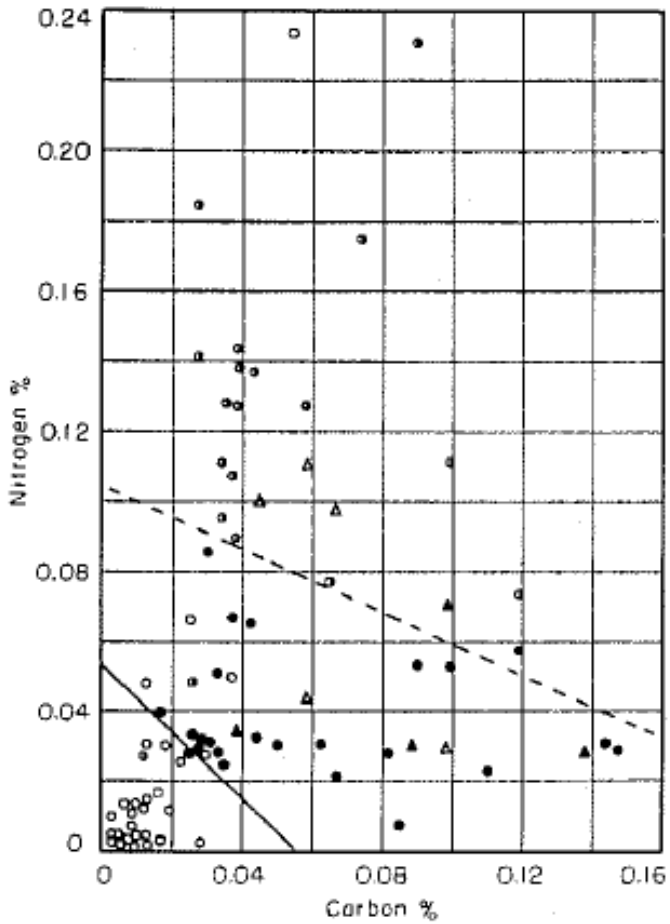
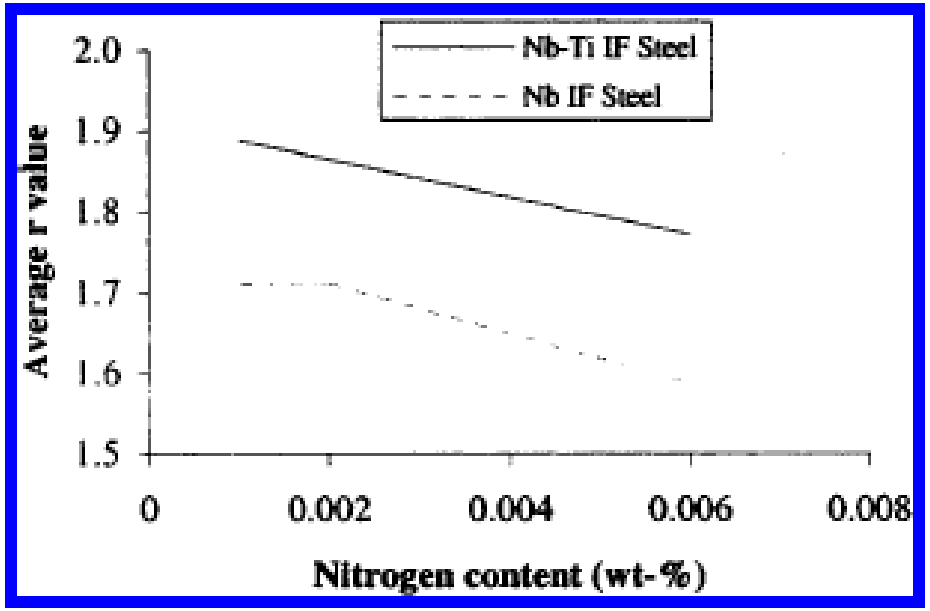


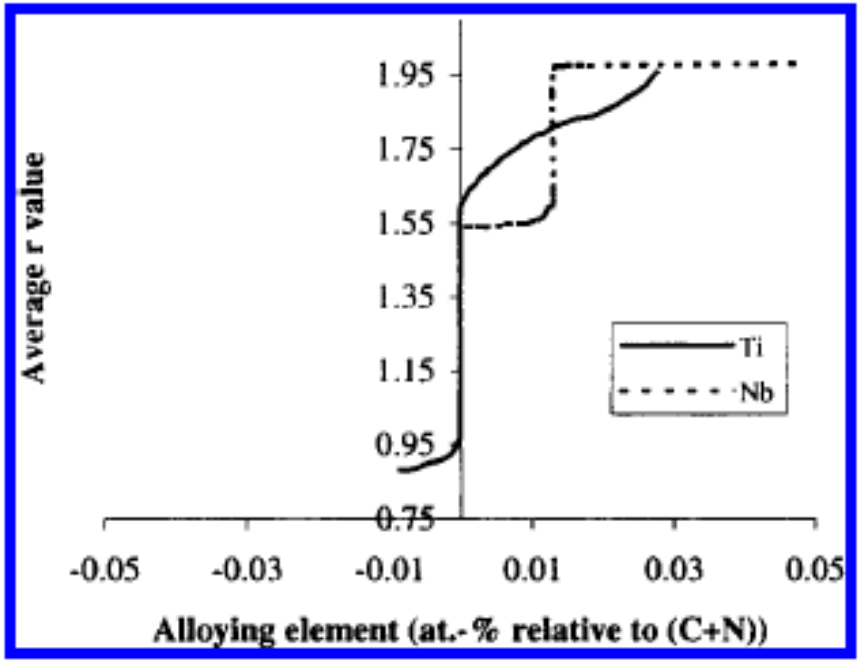
Fig. 7—Effect of Carbon and Nitrogen on the Toughness of 17 to 19% Chromium-Iron Alloys. Open circles—high impact strength alloys; solid circles—low impact strength alloys; semi-solid circles—intermediate impact strength alloys; triangles represent commercial arc-melted steels.

- **Binder and Spendelow, Trans. of ASM, vol. 43, 1951, P. 759**

Properties of Interstitial Free Steels



Effect of nitrogen content on $\bar{r}$ value of range of Ti and Nb–Ti IF steels: after Ref. 7



Effect of alloying addition on $\bar{r}$ value for Ti and Nb additions to IF steels[9]

- Hoile, "Processing and Properties of mild IF steels," Materials Science and Technology, October 2000, vol. 16

Similar Results of Reduced Low Temperature Embrittlement

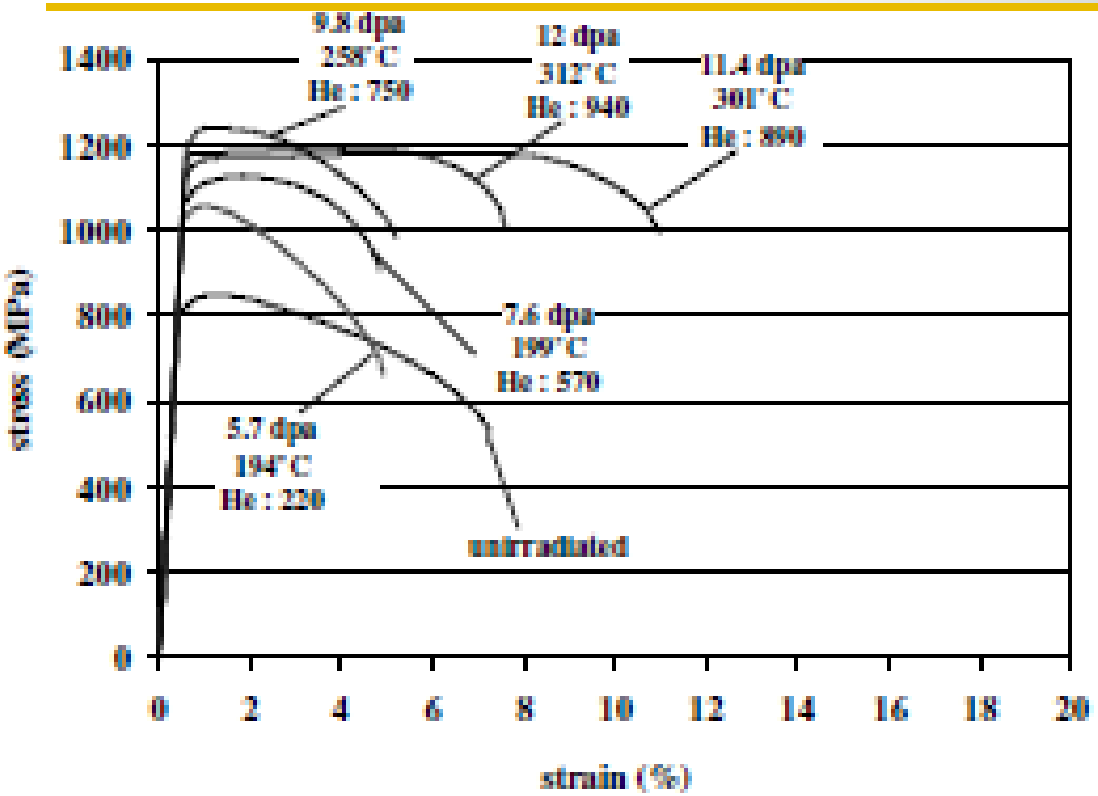


Fig. 3. Selected engineering tensile curves for 20% cold-worked EM10, tested at room temperature. As in Fig. 1, irradiation dose, average irradiation temperature and helium concentrations are

J. Henry et al. / Journal of Nuclear Materials 318 (2003) 215–227

- Note that the highest dose sample shows hardening but retains good elongation.
- During this irradiation, the temperature was increased to over 600C during the middle of the irradiation. It annealed out damage but the resulting microstructure had improved radiation tolerance
- It has also been observed that high purity Ta shows reduced low temperature embrittlement in high purity alloys.

Can annealing sweep out or lock up nitrogen impurities?

- **Modeling studies show that nitrogen strongly couples with vacancy complexes –binding energy of 0.71-0.86 eV (**C. Domain, C.S. Becquart, J. Foct, Phys. Rev. B 69 (2004) 144112, T. Ohnuma, N. Soneda, M. Iwasawa, Acta Mater. 57 (2009) 5947–5955.)
- **Previous results on low temperature irradiated iron with nitrogen impurities show that annealing at >250C reduces the Snoek peak.**

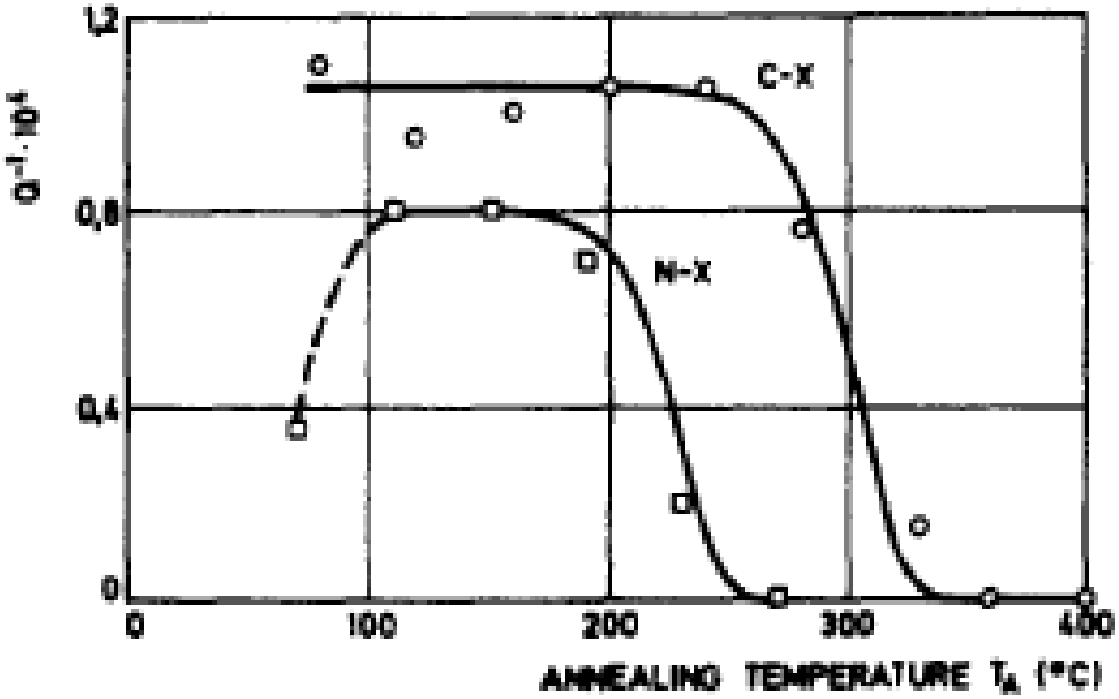


FIG. 4

Isochronal annealing of the heights of peaks C-X and N-X.

Weller, M. and Diehl, J., "INTERNAL FRICTION STUDIES ON REACTIONS OF CARBON AND NITROGEN WITH LATTICE DEFECTS IN NEUTRON IRRADIATED IRON" Scripta Met, V. 10, pp. 101-105, 1976.

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Proposed Hypothesis and Future Research

- **Proposed Hypotheses:**
  - 1. Nitrogen attracts point defects under irradiation., This creates stronger pinning centers in ferritic alloys. Under stress, when the pinning centers are overcome, defect free channels are formed leading to localized deformation.
  - 2. Annealing at >500C after irradiation removes nitrogen from solution either through precipitation or diffusion creating an interstitial free alloy.
- **Next steps**
  - Perform free C/N measurements on alloys. Look at effects of heat treatment
  - Develop ion irradiation technique to detect localized flow after low temperature irradiation
  - Perform a confirmatory irradiation in HFIR
  - Compare to other alloys with low free C/N Super-ferritic alloys (S44627, S44635, S44660)- Ti/Nb added to react with C/N.
  - Investigate differences in strain hardening on different heats of HT-9. Test on different heats of HT-9 and vary the tempering time. Perform TEM analysis on INL heat of HT-9.

Summary

- *Tempered Martensitic Steel Development*
  - *Results from new heat of HT-9 show improved uniform elongation over other TMS after irradiation to 6 dpa at 290C*
  - *Void swelling measurements with high dose ion irradiation also show improve swelling resistance to >500 dpa.*
  - *Reduction of area measurements show more uniform deformation compared to other F/M steels.*
  - *Elemental analysis on this heat of HT-9 reveal a very low nitrogen content compared to other alloys.*
  - *Previous results show that C and N can contribute to localized deformation in ferritic steels.*
  - *Samples that were annealed under irradiation showed improved retention of uniform elongation. Could this be related to reduction in free nitrogen?*

Irradiation Testing on High Purity Fe-Cr alloys

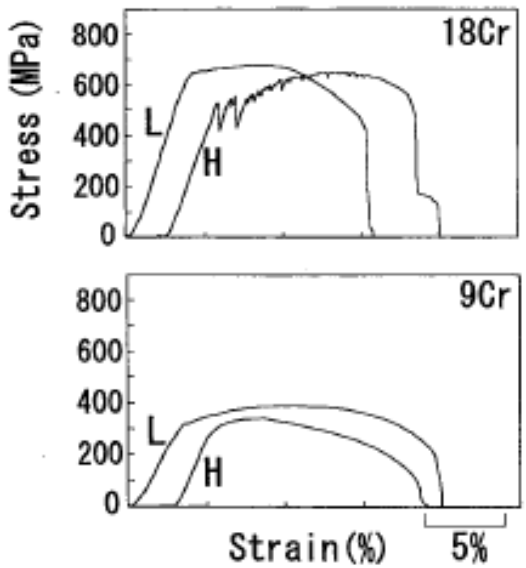

Fig. 1. Dependence of stress–strain curves of Fe–Cr alloys neutron-irradiated and tested at 673 K on the chromium and impurity content. The irradiation dose is 5.2×10^{24} n/m^2. Letters "L" and "H" represent low and high purity, respectively

Table 2

Summarized results of tensile tests measured at 673 K in unirradiated and irradiated Fe–Cr alloys. YS, UTS, UF, and TE stand for yield stress, ultimate tensile stress, uniform elongation, and total elongation

	unirradiated specimens				irradiated specimens			
	YS (MPa)	UTS (MPa)	UE (%)	TE (%)	YS (MPa)	UTS (MPa)	UE (%)	TE (%)
9Cr-H	71	206	13.8	24.5	281	339	2.9	15.9
9Cr-L	180	434	17.8	25.2	324	385	6.0	19.4
18Cr-H	85	233	20.7	34.5	509	648	7.1	17.5
18Cr-L	100	247	34.6	39.0	646	672	5.0	15.1
30Cr-H	282	401	13.4	23.5	589	852	3.9	13.3
30Cr-L	295	464	13.0	22.6	788	879	6.9	10.0

Table 1

Chemical compositions of the low- and high-purity Fe–Cr alloys used. The symbol • represents concentrations less than 0.01 wt%

	C	N	O	S	Cr	Si	Mn	P
	(wt ppm)				(wt%)			
low purity								
Fe–9Cr	100	75	21	50	9.07	0.01	0.07	0.03
Fe–18Cr	110	85	28	50	18.16	0.12	0.13	0.03
Fe–30Cr	120	84	34	60	30.26	0.11	0.10	0.03
high purity								
Fe–9Cr	3	3	51	2	9.11	•	•	•
Fe–18Cr	5	5	63	3	18.21	•	•	•
Fe–30Cr	57	10	66	2	30.20	•	•	•

- **Irradiation to a dose of 5.2 x 10^{24} n/m^2 E> 1.0 MeV (0.3 dpa)**

E. Wakai et al.: Tensile Properties in High-Purity Fe–Cr Alloys

phys. stat. sol. (a) **160**, 441 (1997)

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Irradiation effects on High Purity Fe-Cr alloys

Table 2 Yield strength, ultimate tensile strength, uniform elongation, and total elongation in high- and low-purity Fe–9Cr alloys irradiated at 255°C to 0.3 dpa.

	Temp. (°C)	YS (MPa)	UTS (MPa)	UE (%)	TE (%)
High-purity	20	375	378	3.3	13.3
After	400	273	342	2.9	15.9
Irradiation	500	255	284	2.1	17.6
Before	20	108	233	37.6	58.7
Irradiation	400	71	206	13.8	24.5
	500	69	166	18.6	29.9
Low-purity	20	414	497	11.2	29.3
After	400	330	385	6.0	19.4
irradiation	500	230	288	10.1	35.2
Before	20	212	335	30.0	50.2
irradiation	400	180	434	17.8	25.2
	500	168	418	13.6	25.6

Table 1 Chemical compositions of high- and low-purity Fe–9Cr alloys used in this study. Symbol* represents the concentration less than 0.01 mass%.

	Cr	Si	Mn	P	C	N	O	S
	(mass%)				(mass ppm)			
High-purity	9.11	*	*	*	3	3	51	2
Low-purity	9.07	0.01	0.07	0.03	100	75	21	50

- **Irradiated to 0.3 dpa at 255 or 290 C.**
- **Not sure why they tested at 400C after irradiation.**

E. Wakai *et al.*
Damage Structures and Mechanical Properties of High-Purity Fe–9Cr Alloys Irradiated by Neutrons
Materials Transactions, JIM, Vol. 41, No. 9 (2000) pp. 1180 to 1183