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HIP

Review of development of interatomic potentials and MD calculations for Fe-H and beyond.

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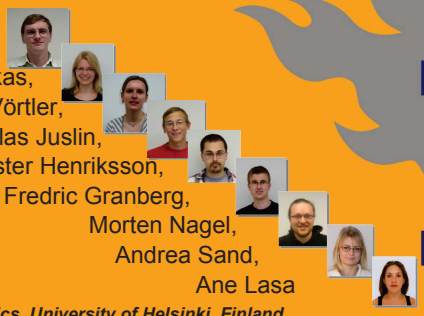


Contents

- Steel and stainless steel
- Radiation damage in nuclear reactors: why study?
- An interatomic potential for stainless steel
  - Test: metastability of cementite
- Brief overview of Fe radiation damage modelling efforts in Europe [IREMEV / EUROFusion]
- An interatomic potential for Fe-H and very first results for plasma-wall interactions
- Outlook: what to do next; possible new CRP topics?

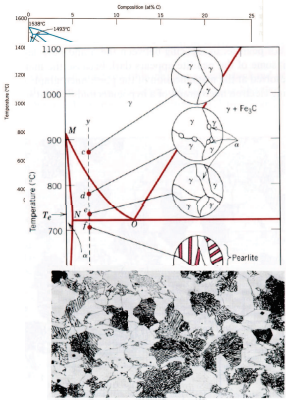
[Olkiluoto 3 pressure vessel, Finland]

Steel

- What is steel?
- Textbook definition: [e.g. Callister, Materials Science and Engineering]:
  - Fe with between 0.008 and 2.14 weight-% C
  - In practice usually < 1 weight-%
- Ferritic steel: BCC crystal structure, e.g. EUROFER
- Austenitic steel: FCC crystal, usually stabilized by Ni
- Can, and almost always does, contain also numerous other alloying elements; Cr, Ni, Mo, ...
- Detailed structure depends on C, alloying element content, and manufacturing conditions

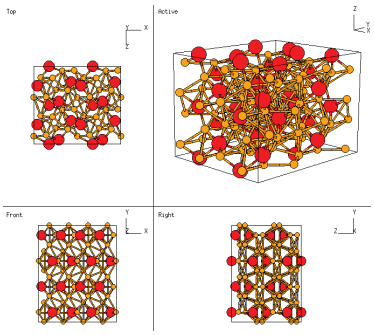
Steels: role of cementite

- The Fe-C phase diagram has a crucial role in understanding properties of steels
- For any C content above 0.022 w-%, formation of cementite inclusions (grains, lamellae) is expected from the phase diagram
- Also other carbides often present



Cementite

- Cementite Fe_3C is a *metastable* ceramic with a complex crystal structure
- Partly covalent bonding
- Hard, brittle

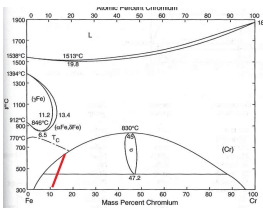


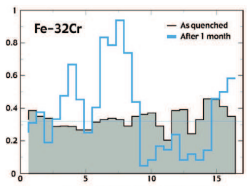
Other Carbides in Steels…

The Fe-Cr-C phase diagram contains the "pure" compounds Fe_3C (metastable), Cr_7C_3, Cr_3C_2, and $Cr_{23}C_6$ (Ref. 13). Chromium can be replaced by iron and *vice versa* (to some limits) in these carbides, so in general the "impure" carbides are M_3C, M_7C_3, M_3C_2, and $M_{23}C_6$, with M denoting a combination of iron and chromium. The carbides Cr_7C_3 and $Cr_{23}C_6$ are common in Cr steels, whereas Cr_3C_2 is not. This latter phase is present in some Fe-Cr-C phase diagrams but not in others[13,14]. In the mixed carbide $(Fe_xCr_{23-x})C_6$ one usually has $x \leq 8$, *i.e.* iron can replace chromium with up to 35 atomic percent[15]. For $(Fe_xCr_{7-x})C_3$ one similarly has $x \leq 4$, and in cementite chromium can replace one iron atom per every two cementite molecules. The chromium carbides can also be found in coating materials (see refs. in 16).

Stainless steel

- Textbook definition:
  - Stainless steel is a steel with > 12 weight-% Cr
- Fe-Cr phase diagram has a crucial role
  - Shows complex behaviour: miscible < 10% Cr, immiscible at 10 – 90% Cr (forms almost pure Cr α' precipitates)



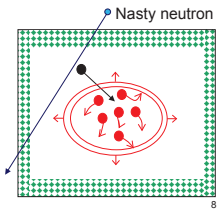


[Olsson et al, PRB **72**, 214119 2005]

Radiation damage in nuclear reactors: why study?

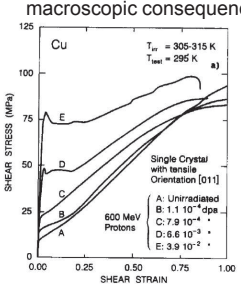
- Neutrons damage all materials in a nuclear reactor
  - The amount of primary defects produced, as well as whether they are isolated or clustered defects, will affect all longer time scale damage evolution
- The primary state of damage is formed when a MeV neutron occasionally hits a lattice atom nucleus and gives it a recoil energy
  - E range typically ~ 1 – 100 keV

[Olkiluoto 3 pressure vessel]

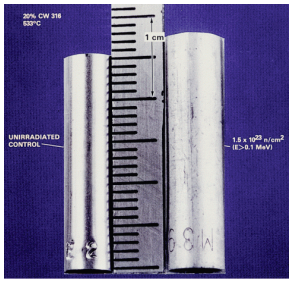


Radiation damage in nuclear reactors: long-term effects

- Via a complex set of additional processes (not well understood) the damage eventually can lead to major macroscopic consequences: swelling, embrittlement, …



[B.N. Singh, A.J.E. Foreman, H. Trinkaus, Journal of Nuclear Materials **249,** (1997) 103-115]



http://cmcsn.phys.washington.edu/book/export/html/467

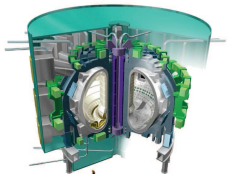
Fusion-relevant steels

- What steels will be used in fusion reactors, and under which conditions?

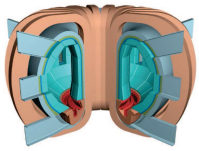
	First Wall	Dose (dpa)	Temperature	
ITER	Austenitic steel	<3 dpa	<300 °C	
DEMO	EUROFER	50-89 dpa	<550°C	
Power Plant	ODS Ferritic Steels	100-150 dpa	<750 °C	
Power Plant	SiC_f/SiC Composites	100-150 dpa	upto 1100°C	

[By Jean-Louis Boutard, ICFRM-14, 7th to 11th of September 2009, Sapporo (Japan)]

ITER



Fusion Power Plant



EFDA
EUROPEAN FUSION DEVELOPMENT AGREEMENT

Comparison: ITER, DEMO and Power Reactor

	ITER	DEMO	*Reactor*
Fusion Power	**0.5 GW**	**2 – 2.5 GW**	***3 - 4 GW***
Heat flux (FW)	**0.1-0.3 MW/m²**	**0.5 MW/m²**	***0.5 MW/m²***
Neutron Wall Load (First Wall)	**0.78 MW/m²**	**< 2 MW/m²**	***~ 2 MW/m²***
Integrated wall load (First Wall)	**0.07 MW.year/m² (3 years Inductive operation I)**	**5 - 8 MW.year/m²**	***10 - 15 MW.year/m²***
Displacement per atom (dpa)	**< 3 dpa**	**50 - 80 dpa**	***100 - 150 dpa***
	The challenge		
Transmutation product rates at first wall	**~10 appm Helium / dpa ~45 appm H / dpa**		

What is EUROFER?

- Definitions presented by Rainer Laesser at SOFT conference in 2006 [Presentation available in web: laesser_materials_soft06.ppt]

"EUROFER is a RAFM steel developed on the basis of conventional 9%Cr-1%Mo steels used in fission, where
- Highly activating alloying elements (Mo, Nb) were replaced by those (W, Ta) offering lower activation.
- 8-10% Cr: optimized concentration for good fracture properties and corrosion resistance.
- 1-2% W: optimized for mechanical properties (ductility, strength, fracture properties).
- 0.07% Ta: stabilizes grain size and improves strength"

Specification of the chemical composition of EUROFER 97

	Element	Minimum Value (wt%)	Maximum Value (wt%)	Remarks Target
A)	Cr	8.50	9.50	9.0
	C	0.090	0.120	0.11
	Mn	0.20	0.60	0.4
	P		0.005	
	S		0.005	
	V	0.15	0.25	
	B			ALAP, <0.001
	N_2	0.015	0.045	0.03
	O_2		0.01	
B)	W	1.0	1.2	1.1
	Ta	0.05	0.09	
	Ti		0.01	
C)	Nb			ALAP, <0.001
	Mo			ALAP, <0.005
	Ni			ALAP, < 0.005
	Cu			ALAP, <0.005
	Al			ALAP, < 0,01
	Si		0.050	
	Co			ALAP*), < 0.005
	As+Sn+Sb+Zr			<0.05

A) = basic composition
B) = varied substitution elements
C) = radiological undesired tramp-elements according FZKA 5624

*) ALAP = as low as possible, no addition

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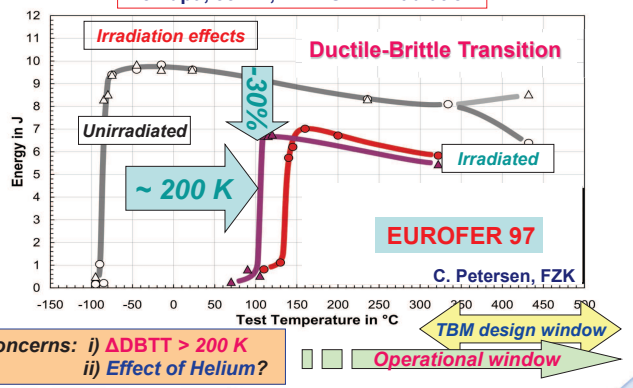
Degradation of Impact Properties under neutron irradiation

~32 dpa, 332°C, ARBOR 1 irradiation



Concerns: i) ΔDBTT > 200 K
ii) Effect of Helium?

Interatomic potential for stainless steel + He

- To enable modelling stainless steels we made in 2009-2012 an interatomic potential for the Fe-Cr-C system

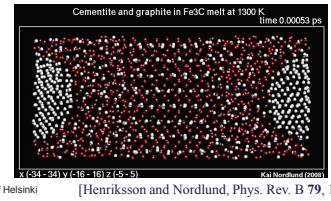
	Fe	Cr	C	He
Fe	✓			
Cr	✓	✓		
C	✓	✓	✓	
He	✓	✓	✓	✓

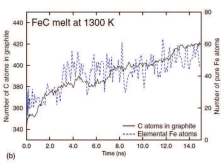
✓ < 2002, several
✓ 2005: Olsson / Caro
✓ 2009: Henriksson / Hepburn
✓ 2012: Henriksson, Juslin

[FeC: Henriksson and Nordlund, Phys. Rev. B **79**, 144107 (2009)]
[FeHe: Juslin and Nordlund, J. Nucl. Mater **382**, 143 (2008)]
[CrHe: Terentyev, Juslin and Nordlund, J.Appl. Phys. **105**, 103509 (2009)]
[FeCrC: K. O. E. Henriksson, C. Björkas, and K. Nordlund, J. Phys. Condens. Matt. 25, 445401 (2013)]

Test of FeC part: metastability of cementite

- As one of the first dynamic tests, we examined the melting properties of cementite
- Experimentally cementite is metastable, and decomposes into graphite around 1100-1200 K
  - We found decomposition around 1200 K – 1300 K!
- In a cell with both cementite and graphite inclusion, the graphite grows – excellent agreement with experiments!





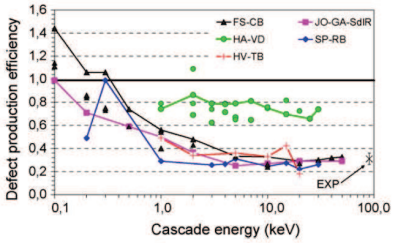
[Henriksson and Nordlund, Phys. Rev. B **79**, 144107 (2009)]

Alternative interatomic potentials: EAM family

- For metals the embedded-atom-method formalism is more commonly used, and Fe and steels have a large number of alternatives:
  - Pure Fe: tens of EAM potentials, important ones: Ackland-Mendelev, Dudarev-Derlet ("magnetic")
  - Fe-Cr: several potentials, e.g. Olsson, Caro, Marinica
  - Fe-W, Fe-Ni: recent potentials by Bonny-Malerba et al
  - Fe-C: Hepburn potential for C in Fe, does not work for pure C though
  - Fe-He, Cr-He: Juslin potential
  - Fe-H: At least by Carter group
- Not all combinations of these possible, though

Radiation damage in Fe:
Status in 2006

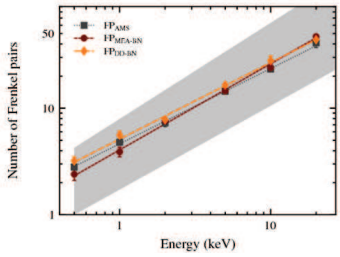
- In 2006 Lorenzo Malerba reviewed the status of damage production simulations in Fe
- Factor of 2 difference between results! ☹



[L. Malerba, J. Nucl. Mater. 351 (2006) 28]

Radiation damage in Fe:
Status in 2007: quantitative reliability!?

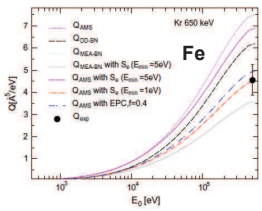
- However, in 2007 we restricted the comparison to modern potentials and ran new simulations in a consistent manner
- It appeared that quantitative reliability was achieved
  - Gray area shows variation of previous potentials, lines 3 recent potentials with good i properties tested by us



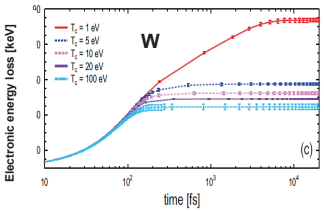
[Björkas and Nordlund, NIM B 259 (2007) 853]

Radiation damage in Fe:
2009: Back to the drawingboard…

- However, slightly after that we realized that depending on how the low-energy limit of electronic stopping is treated, one can get a major (~factor of 2) variation in damage production and ion beam mixing
  - Even primary damage production not reliably predictable – in spite of 5 decades of studies in Fe …



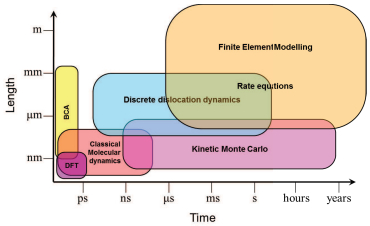
[C. Björkas and K. Nordlund, Nucl. Instr. Meth. Phys. Res. B 267, 1830 (2009)]



[A. E. Sand, S. L. Dudarev, and K. Nordlund, EPL **103**, 46003 (2013)]

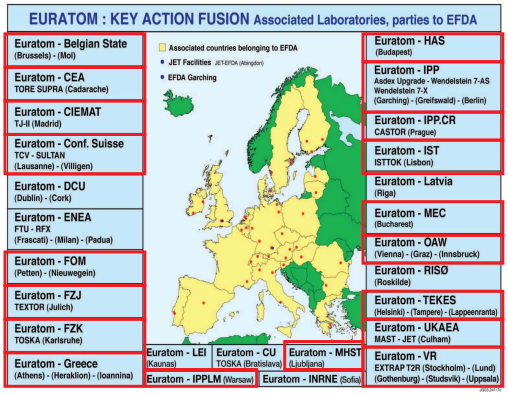
Radiation damage in Fe:
Overall Fe activities in Europe: IREMEV

- EFDA / EUROFUSION is coordinating a major EU-effort where all levels of multiscale modelling are used to attempt to finally understand radiation damage in steels
  - Now Sergei Dudarev task coordinator, Sehila Gonzalez, …



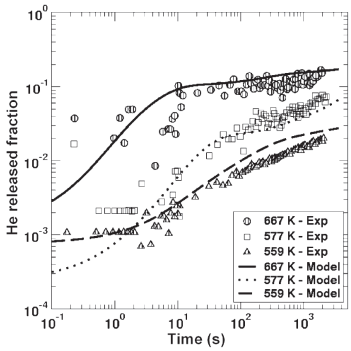
[Review: S. Dudarev, J.-L. Boutard, R. Lässer, M. Caturla, P.M.Derlet, M. Fivel, C.-C. Fu, M. Lavrentiev, L. Malerba, M. Mrovec, D. Nguyen-Manh, **K. Nordlund**, M. Perlado, R. Schäublin, H. V. Swygenhoven, D. Terentyev, J. Wallenius, D. Weygand, and F. Willaime, J. Nucl. Mater. **386-388**, 1 (2009)]

Partners in materials: almost all EFDA partners



Radiation damage in Fe:
Highlight of numerous IREMEV results

- DFT + rate theory modelling of He desorption from Fe obtains very good agreement with experiments – provided vacancy trapping with C is included in model!



[M. Victoria, S. Dudarev, J. L. Boutard, E. Diegele, R. Lässer, A. Almazouzi, M. J. Caturla, C. C. Fu, J. Källne, L. Malerba, **K. Nordlund**, M. Perlado, M. Rieth, M. Samaras, R. Schaeublin, B. Singh, and F. Willaime, Fusion and engineering design **82**, 2413 (2007).]

Interatomic potential for FeH

- Since steels became a plasma-facing material in latest ITER redesign, we have now developed an interatomic potential for H-Fe (H-C parts for 'free' from Brenner potential)
  - Extension of previous Fe-Cr-Cr potential but without Cr-H part

	Fe	Cr	C	He	H
Fe	✓				
Cr	✓	✓			
C	✓	✓	✓		
He	✓	✓	✓	✓	
H	✓		✓	✓	✓

✓ < 2002, several

✓ 2005: Olsson / Caro

✓ 2009: Henriksson / Hepburn

✓ 2012: Henriksson, Juslin

✓ 2014: Kuopanportti

Interatomic potential for FeH

- DFT-based, DFT calcs. by Erin Hayward and Chu-Chun Fu [CEA]:
- Fit to several crystal structures, defect properties tested

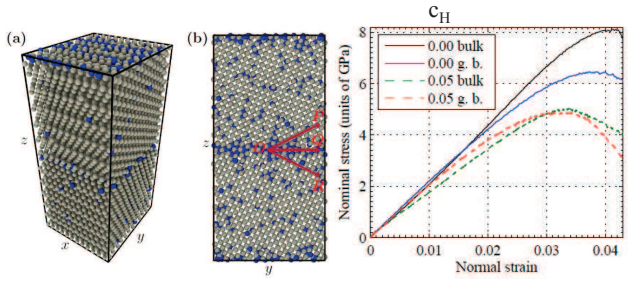
	Experiment		*Ab initio* calculations					AP
FeH	Ref. [47]	Ref. [48]	Ref. [49]	CI/ECP [50]	CASSCF/MRCI [51]	MRCPA(4) [52]	MRSDCI+Q [52]	
r_b (Å)	1.589			1.578	1.588	1.596	1.582	1.589
E_c/atom (eV)		−0.815		−0.71	−0.965	−0.90	−0.94	−0.815
k (cm^{-1})			1774	1701	1643	1735	1778	1774
FeH_2 linear	Ref. [45]	Ref. [46]		CASSCF [53]	CI [53]	B3LYP [54]	B3LYP/ECP [55]	
r_b (Å)	1.6484	1.665		1.746	1.689	1.645	1.647	1.630
E_c/atom (eV)								
FeH_3 planar D_{3h}								
r_b (Å)								
E_c/atom (eV)								
FeH rock salt								
a (Å)								
E_c/atom (eV)								
B (GPa)								
B'				4.26	3.7	3.7		4.749

Defect	Experiment Ref. [59]	DFT calculations PAW-GGA [60]	USPP-GGA [61]	This work	AP This work
Tetrahedral interstitial, unrelaxed		0.29		0.484	0.515
Tetrahedral interstitial, relaxed	0.296	0.20	0.30	0.234	0.240
Octahedral interstitial, unrelaxed		0.76		0.822	1.186
Octahedral interstitial, relaxed		0.33		0.259	0.256
Substitutional defect, unrelaxed				2.855	4.027
Substitutional defect, relaxed				2.526	3.145

[Pekko Kuopanportti, Erin Hayward, Chu-Chun Fu, Antti Kuronen, and Kai Nordlund, submittedish (2014)]

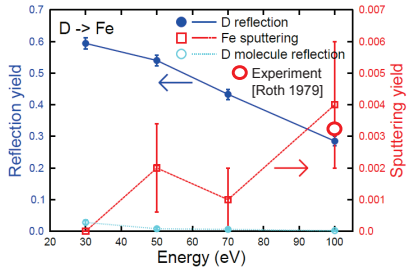
Interatomic potential for FeH: test

- We tested the potential wrt. Tensile testing of a Fe bicrystal with a tilt grain boundary



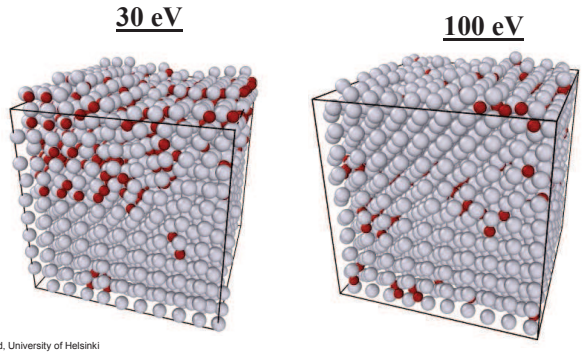
Plasma-wall interactions of H-Fe

- We just initiated simulations of H-Fe plasma wall interactions
  - 30 – 100 eV cumulative H bombardment of pure BCC Fe
- First results (from a week ago):



Plasma-wall interactions of H-Fe

- Sample modification after 1000 incoming H



Issues to address…

- All the usual sputtering yield, T retention and D/T reflection issues etc. need to be determined
  - See W. Jacob – TRIDYN useful for a lot
- Difference between sputtering and reflection from austenitic [ITER?] and ferritic [DEMO] steels?
- Steels always have C – will it get enriched or depleted at surface
  - Will steels after all introduce hydrocarbons into the reactor??