RADIATION-INDUCED FORMATION OF HARDENING SOLUTE CLUSTERS IN FERRITIC/MARTENSITIC ALLOYS: AN OBJECT KINETIC MONTE CARLO MODEL

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Operating temperature window of nuclear materials

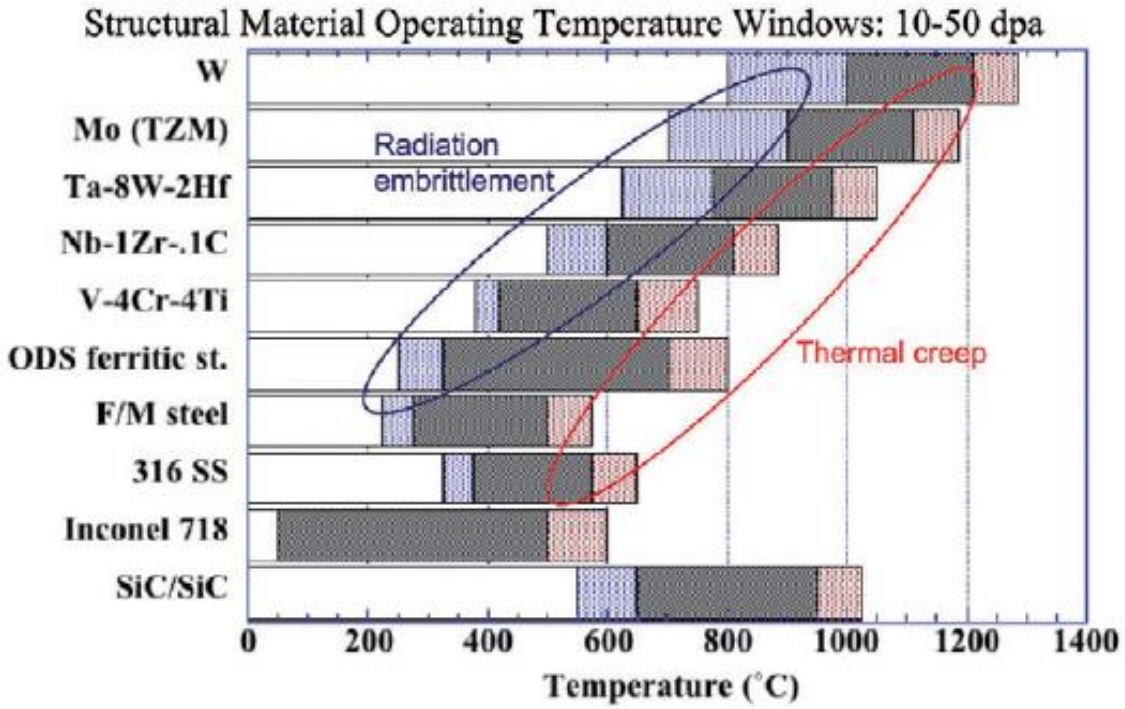
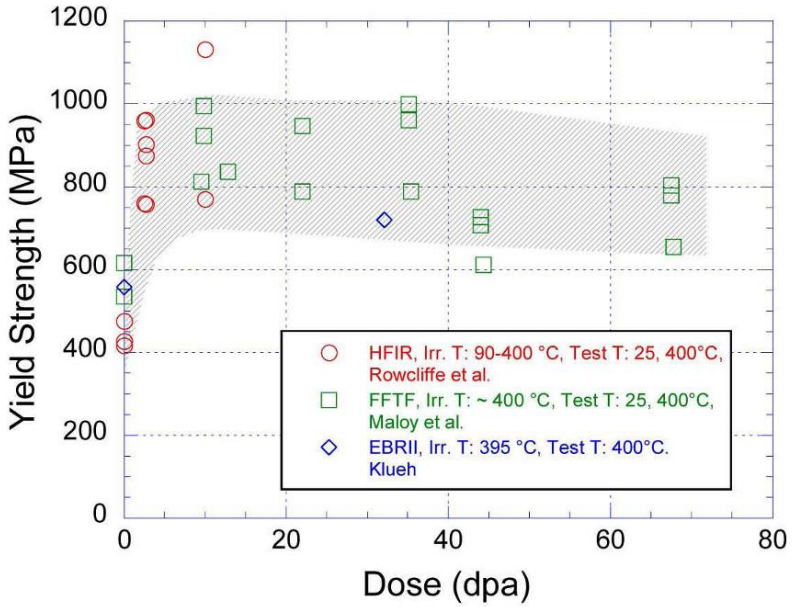


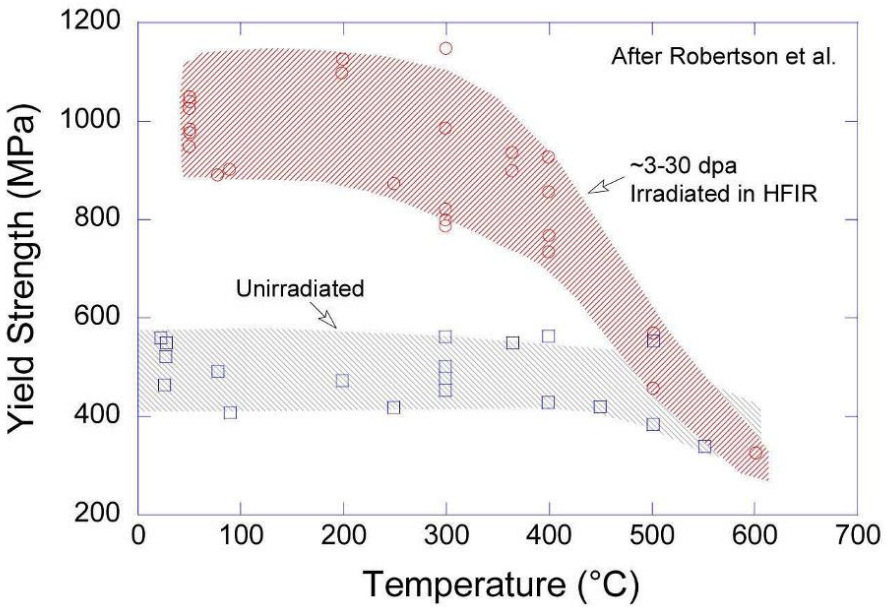
Fig. 5 Estimated operating temperature windows (dark shaded region)[30,79] for structural materials in nuclear energy systems for damage levels of 10 to 50 dpa. The light blue and red regions represent lower and upper temperature uncertainty bands.

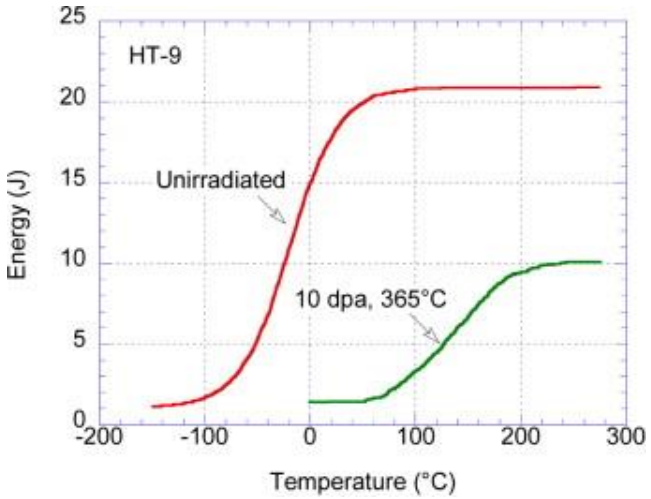
S.J. Zinkle, J.T. Busby, Materials Today 12 (2009) 12

- Lower bound:
  - Radiation induced embrittlement
    - (Low T)
- Upper bound
  - Thermal creep
- … although in reality environment effects are generally the real limiting factor

Low temperature radiation embrittlement in F/M alloys





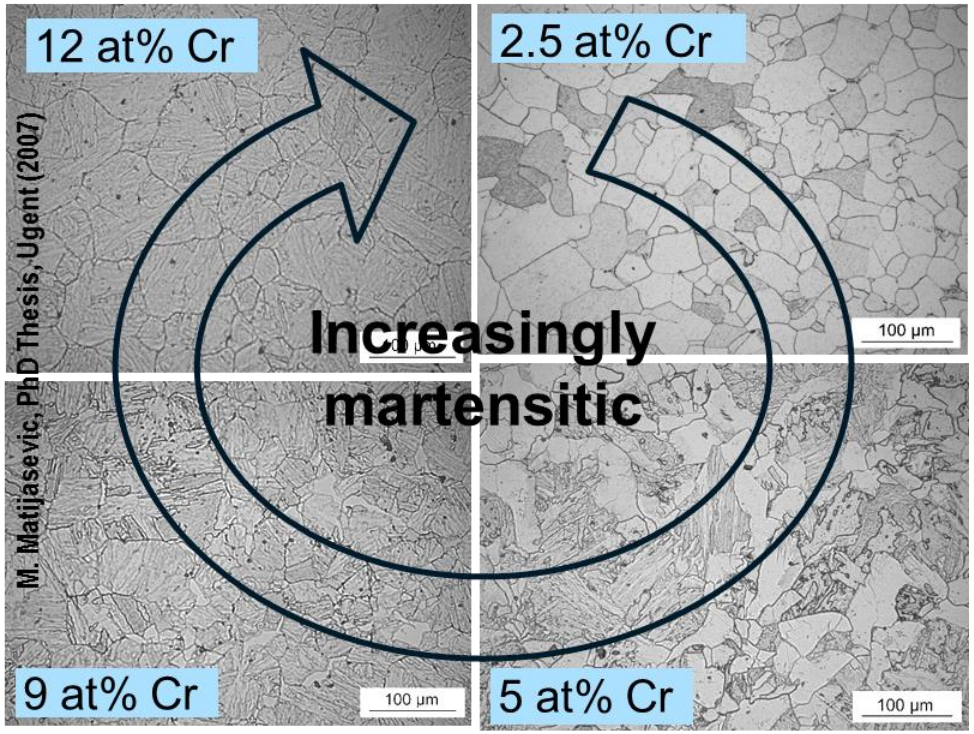


Y. Chen, Nucl. Eng. & Technol. 45 (2013) 311

- When irradiated below 400°C the yield strength increases significantly
- This leads to shifts of the DBTT of up to 120°C

Radiation hardening & embrittlement are caused by microstructural and microchemical changes

F/M model alloys neutron/ion irradiated to different dpa to correlate microstructural changes to radiation hardening



Neutrons (BR2, 2005)

T = 300°C
3 doses : 0.06 dpa – 0.6 dpa – 1.5 dpa

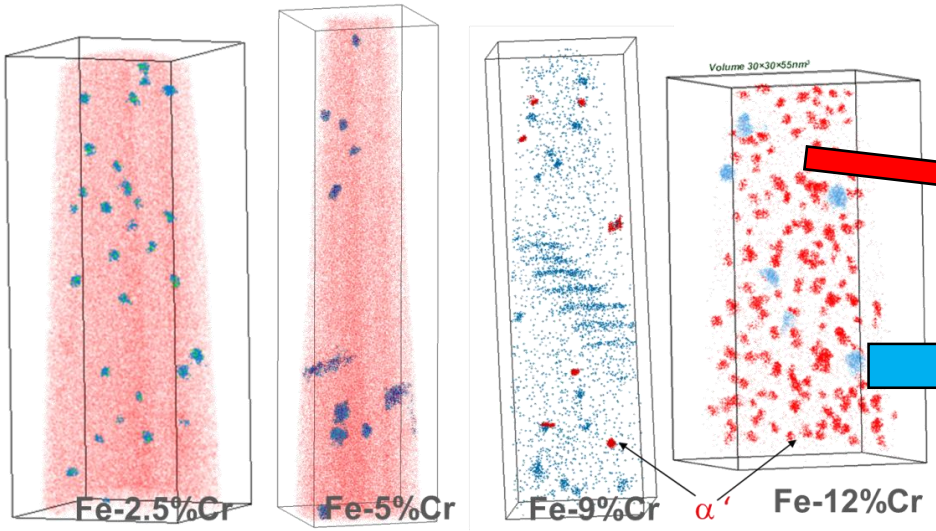
High energy Fe ions (HZDR, 2010)

3 temperatures: 100°C, 300°C and 420°C
2 doses : 1 dpa and 5 dpa

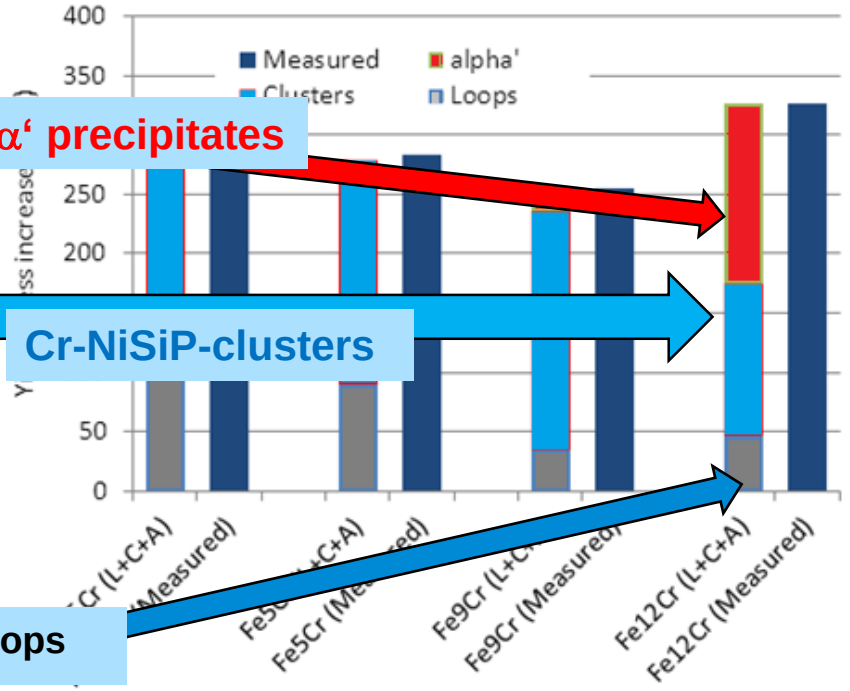
Materials	Composition in at.%											
	Cr	Si	P	Ni	C	Al	S	Ti	V	N	Mn	O
Fe-2.5% Cr	2.6	0.04	0.02	0.04	0.05	0.006	0.004	0.005	0.001	0.05	0.01	0.14
Fe-5% Cr	4.9	0.08	0.02	0.06	0.09	0.001	0.01	0.003	0.001	0.05	0.02	0.21
Fe-9% Cr	8.9	0.18	0.02	0.07	0.09	0.014	0.001	0.004	0.002	0.06	0.03	0.23
Fe-12% Cr	12.3	0.22	0.09	0.09	0.13	0.006	0.012	0.004	0.002	0.09	0.03	0.22

PIE identified several interesting microstructural features

V. Kuksenko et al. J. Nucl. Mater. 432 (2013) 160



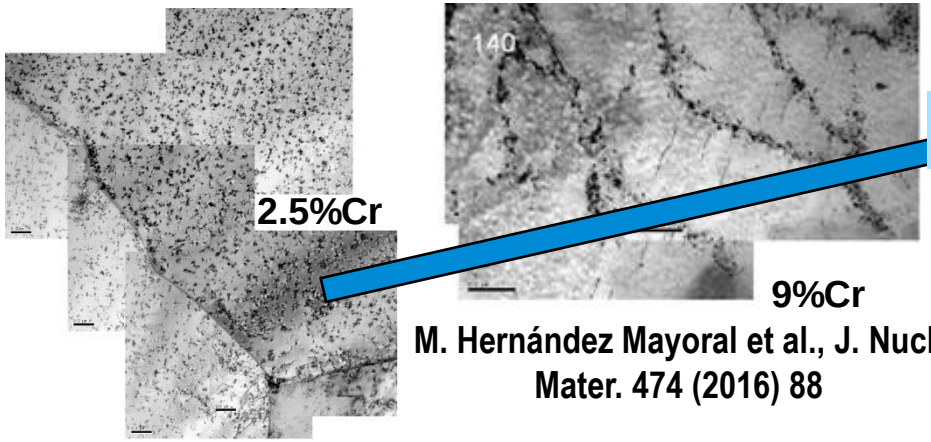
F. Bergner et al. J. Nucl. Mater. 448 (2014) 96



α' precipitates

Cr-NiSiP-clusters

Loops

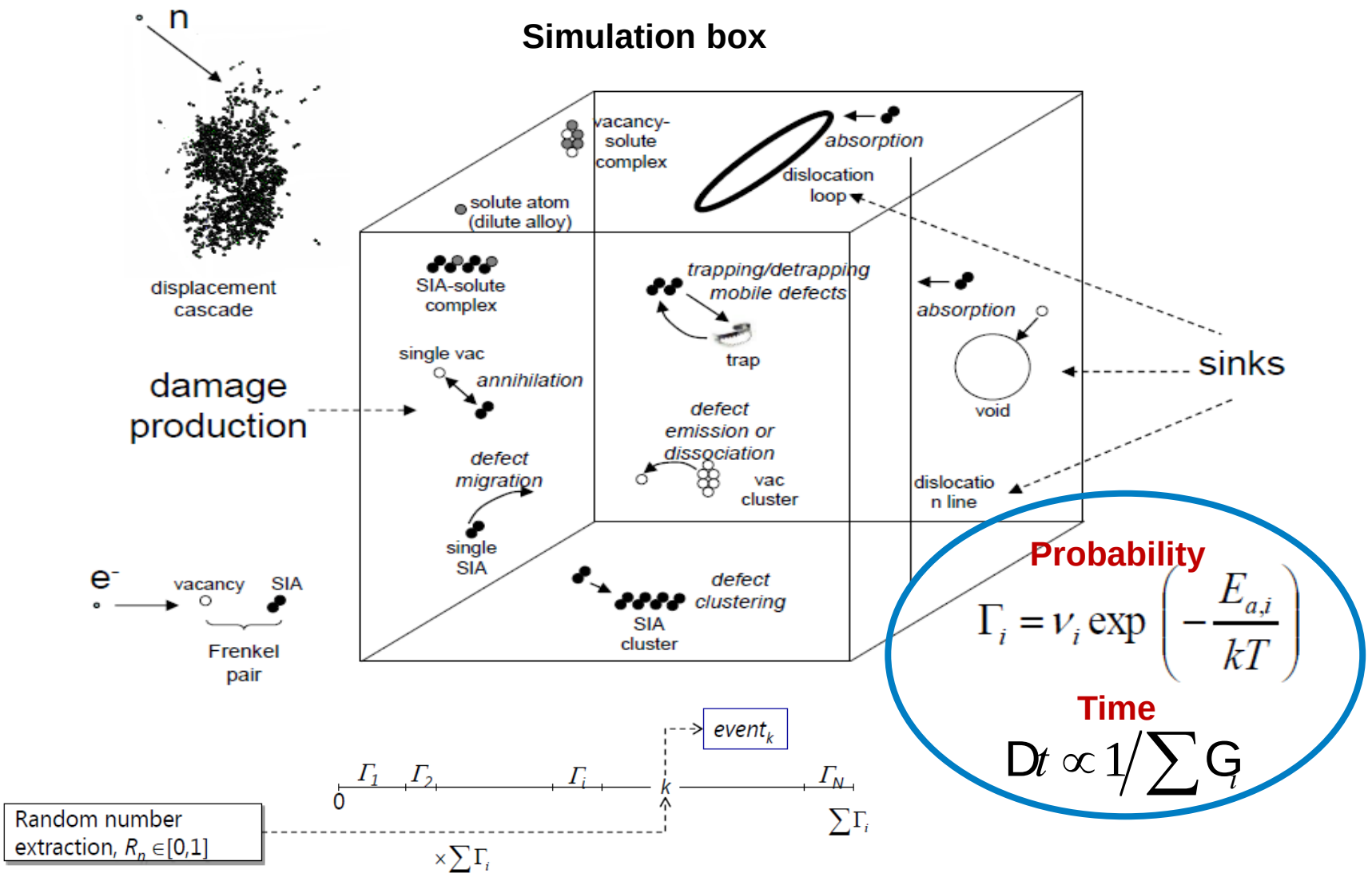


M. Hernández Mayoral et al., J. Nucl. Mater. 474 (2016) 88

Visible dislocation loops, solute clusters and α' explain hardening of F/M Fe-Cr alloys vs Cr content

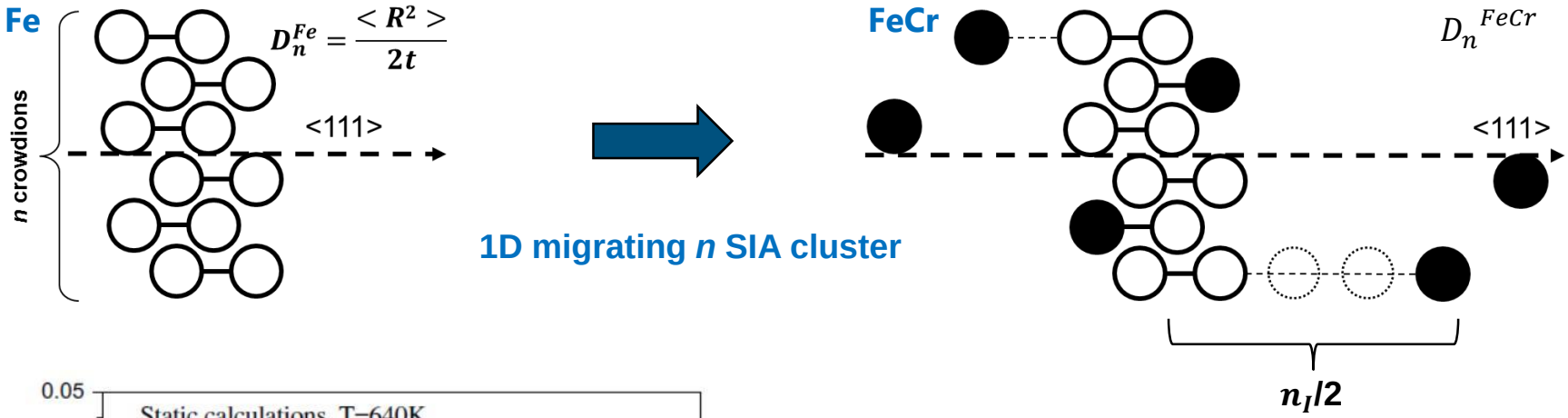
Objective: develop microstructure evolution models capable of describing this whole complexity

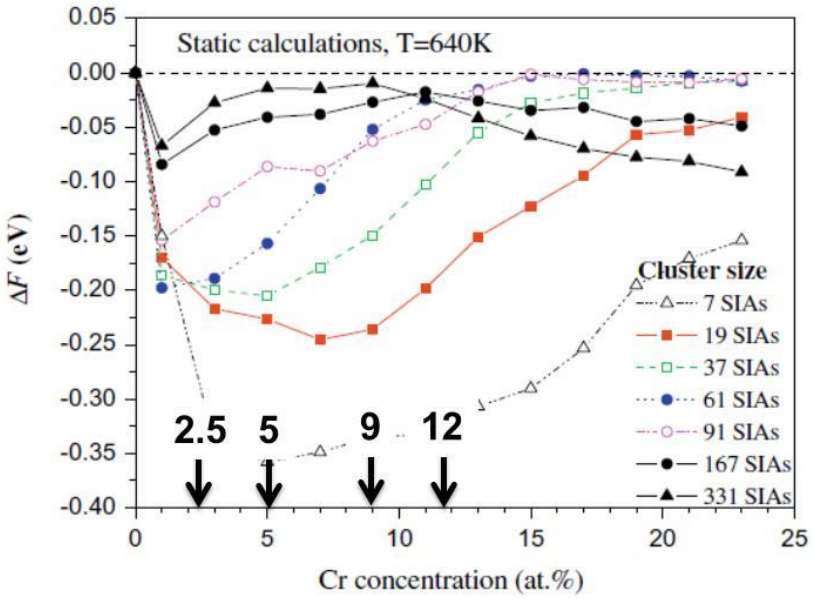
Object kinetic Monte Carlo method



Grey alloy – **key assumption**: slowing down of SIA clusters

- Interaction with Cr reduces SIA defect mobility



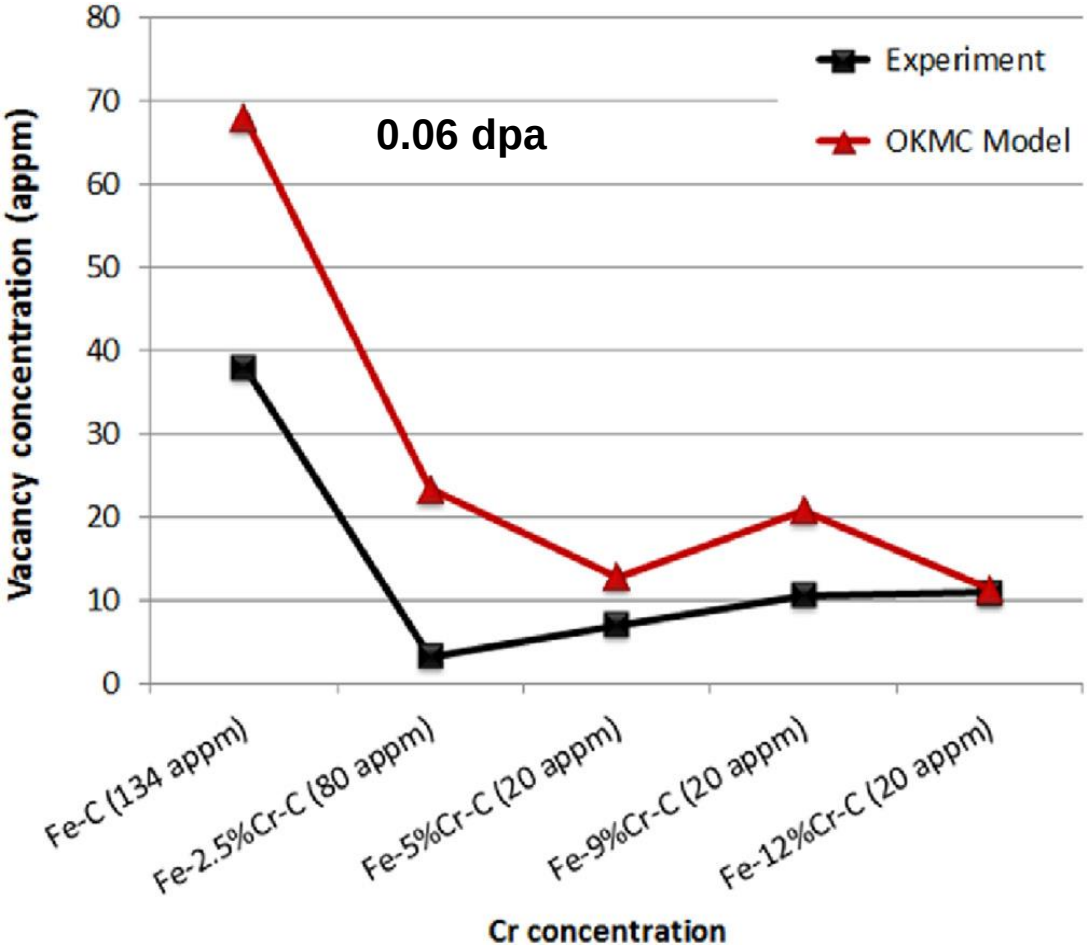


$$D^{\mathrm{FeCr}} = D^{\mathrm{Fe}} \exp(\Delta F / k_B T)$$

$$\frac{D^{FeCr}}{D^{Fe}}$$

$$\Gamma_{X,V} = \nu \exp\left(-\frac{E_a}{k_B T}\right)$$

Cr addition suppresses formation of vacancy clusters

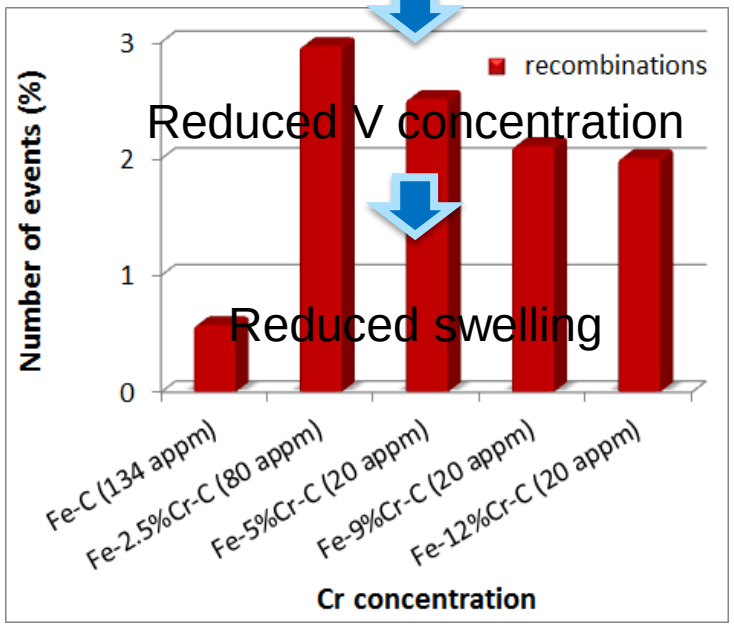


Slow SIA clusters are "easier" sinks for V

Enhanced recombinations

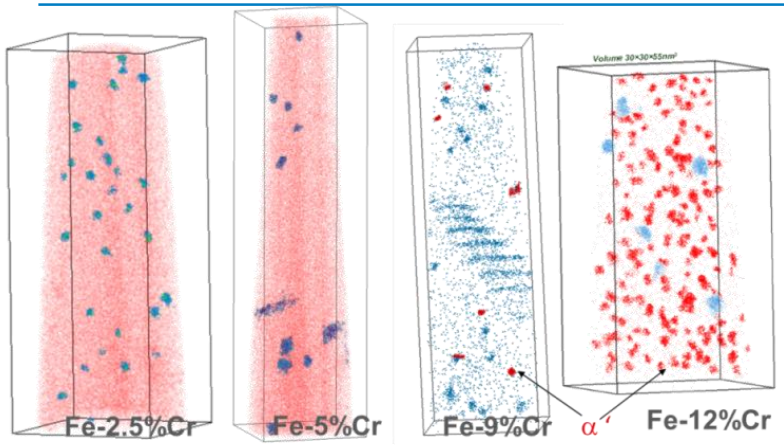
Reduced V concentration

Reduced swelling

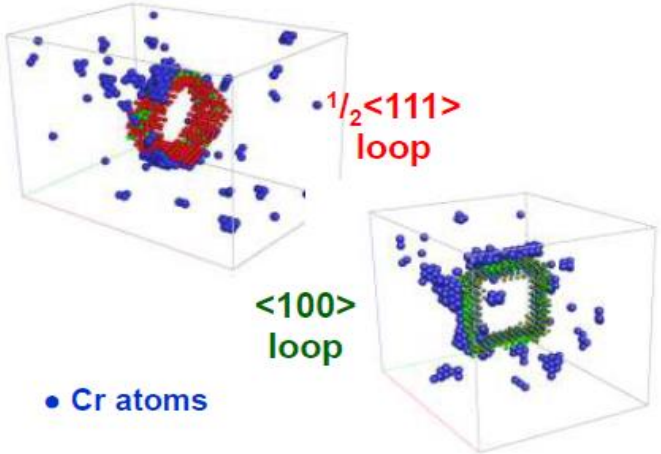


M. Chiapetto, L. Malerba, C.S. Becquart, Journal of Nuclear Materials 465 (2015) 326–336

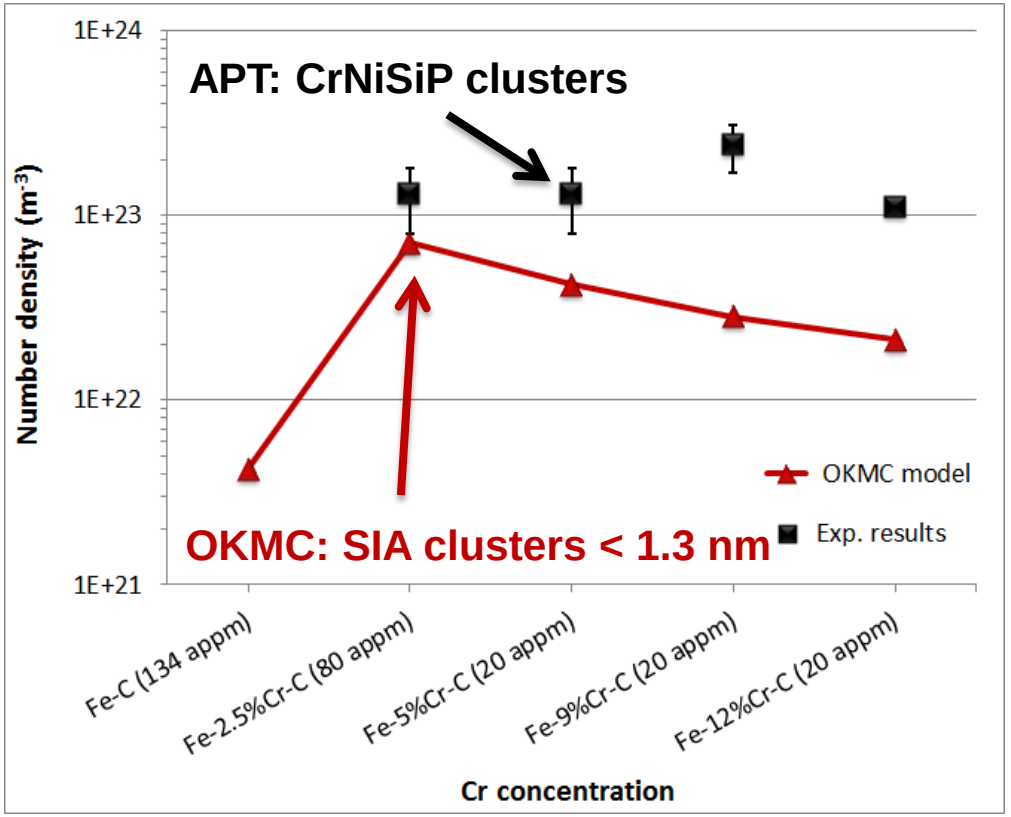
Invisible loops associated with CrSiNiP clusters



Metropolis Monte Carlo
T=600-1200K, Fe10-12%Cr



E.E. Zurkin et al., JNM 417 (2011) 1082



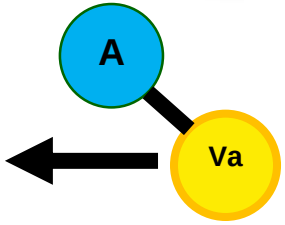
M. Chiapetto et al., JNM 465 (2015) 326–336

Probably there are also solute clusters not associated with loops, but the model gives a reasonable order of magnitude

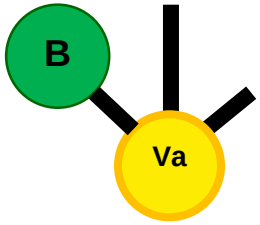
Further step: Introduce explicitly the transport of solutes to describe loop enrichment/solute cluster formation

Key atomistic mechanism: solute dragging by point defects

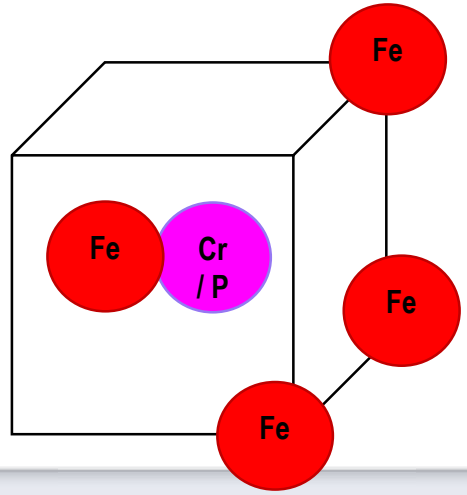
In general, one may expect that a solute atom will move via vacancy in the opposite direction to the vacancy



Instead, Ni, Si, and P, follow the vacancy during its migration



Finally, any solute forming a mixed dumbbell (Cr & P) wil be dragged by it

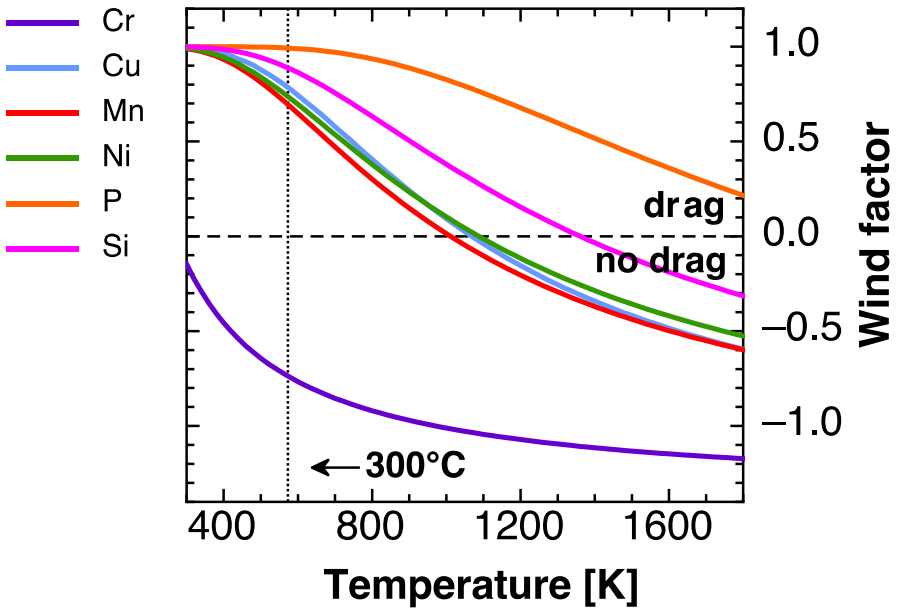


Dragging has solid physical basis rooted in DFT

L. Messina, P. Olsson, M. Nastar, T. Garnier, C. Domain, PRB 90, 104203 (2014)

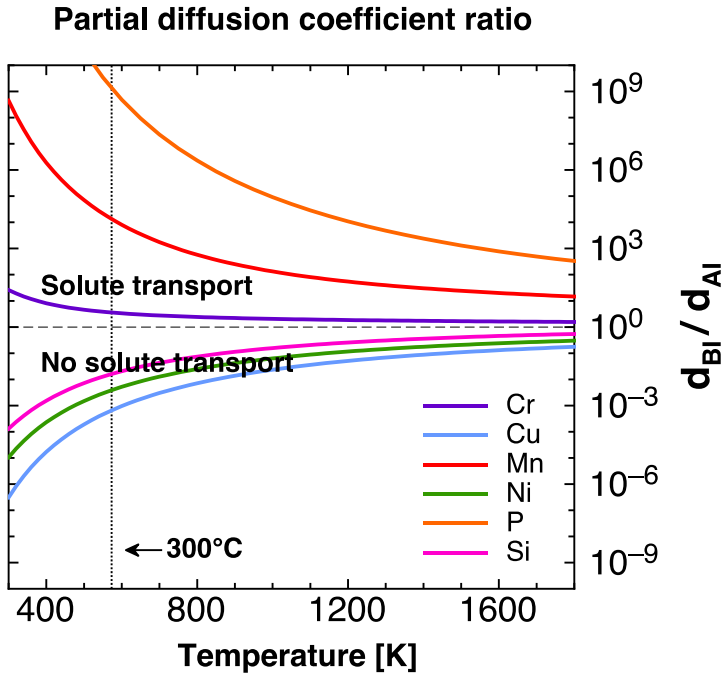
VACANCY DRAG

- Vacancy can carry solute atoms along.
- Occurring for **manganese, nickel, copper, phosphorus, silicon.**



INTERSTITIAL TRANSPORT

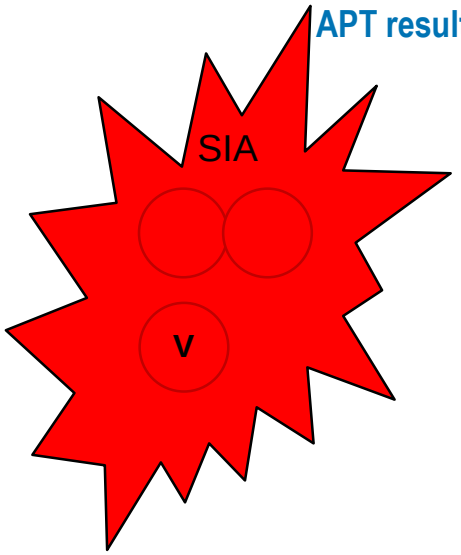
- Interstital atoms can couple with solute atoms and move together.
- Occurring for **manganese, phosphorus, chromium**.

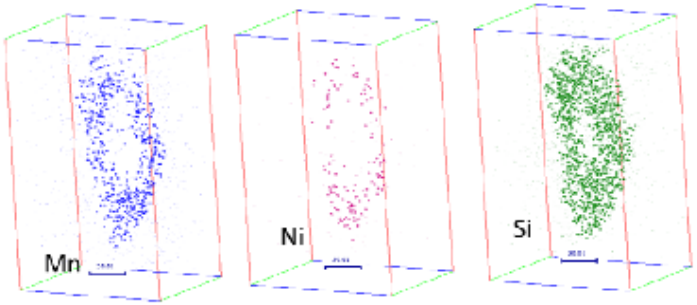


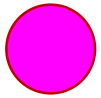
NB: Cr atoms are essentially the only ones NOT dragged by vacancies (though dragged by SIA)

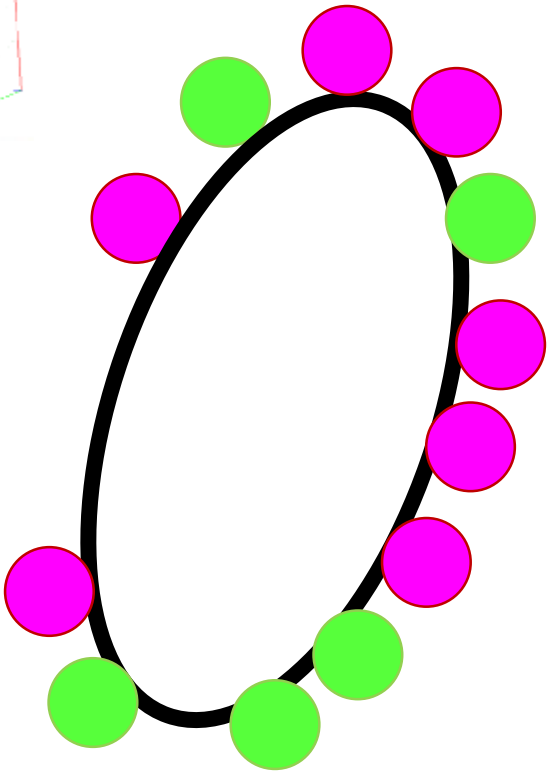
Solutes will segregate at dislocation loops!

APT results - Courtesy of Radiguet , Huang, Cammelli & Pareige, GPM – FP7/Longlife









Atomistic models provided diffusion data for Va-X pairs

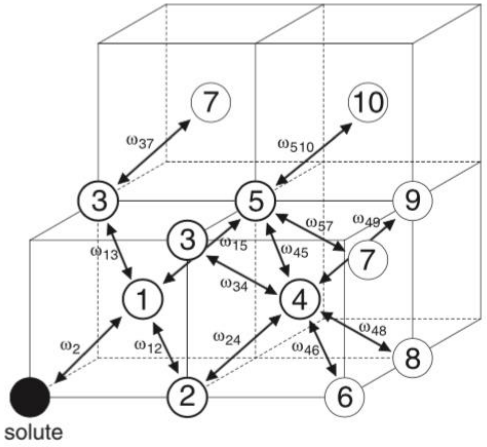
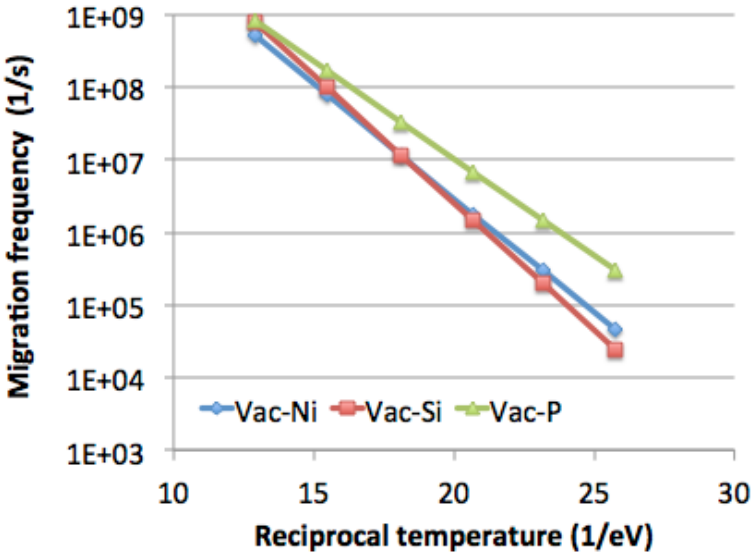
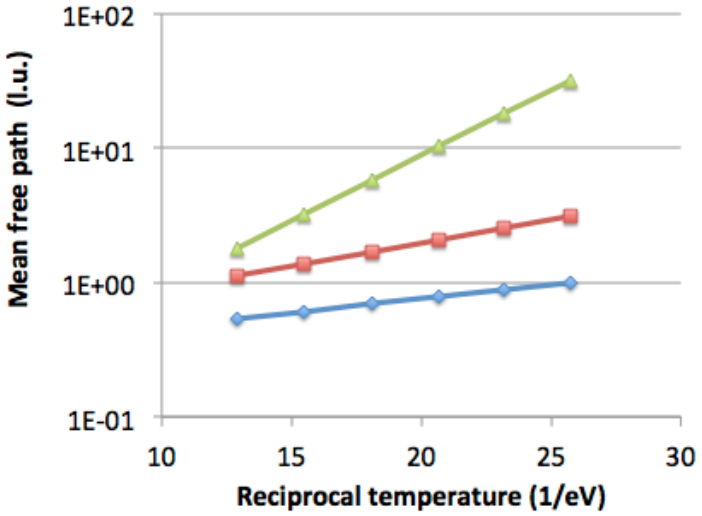
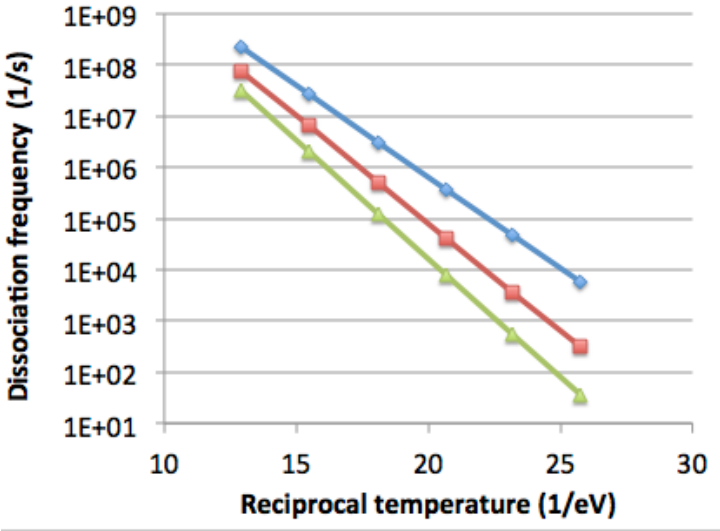


FIG. 1. Network of the 12 Fe-V jump frequencies affected by the presence of a solute atom, for solute-vacancy interactions extending to the 5nn distance. The solute-vacancy exchange is labeled with ω_2, while ω_0 represents the unaffected Fe-V jump frequency (not shown).

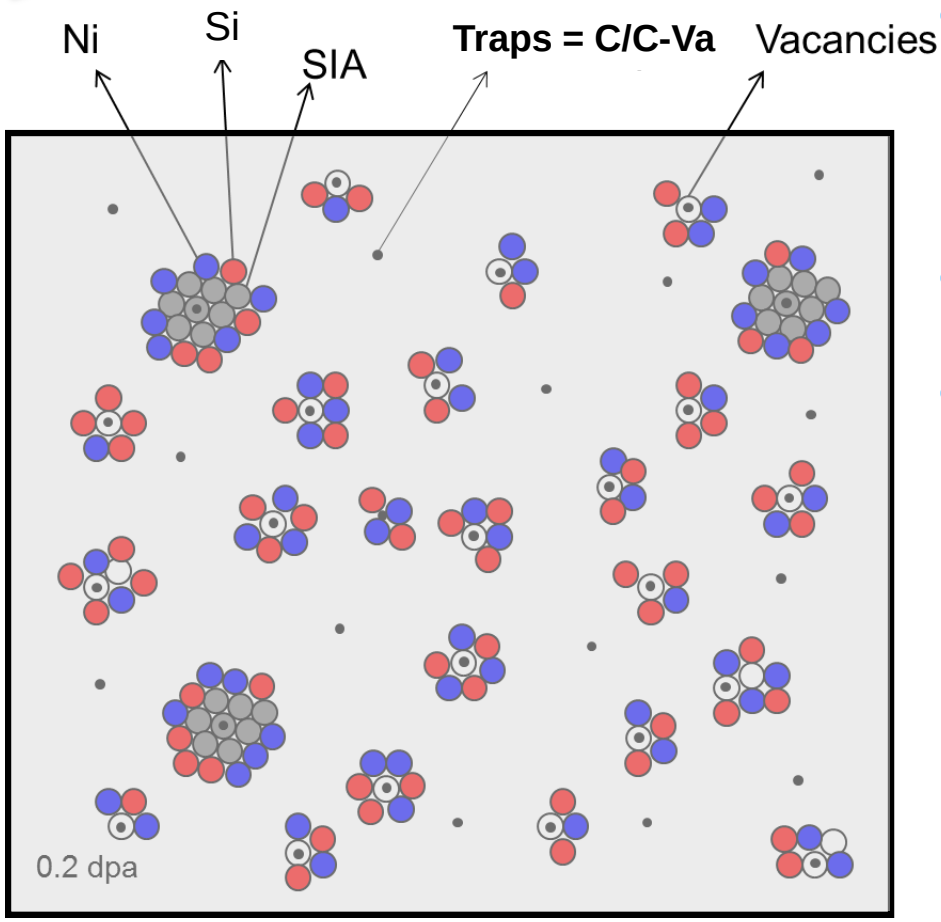






Mateo (SCK•CEN)

Hybrid A/OKMC model: OKMC for microstructure, AKMC for solute transport



Current stage: Cr atoms, present in high concentration, remain as "grey background" and uniformly distributed!

- Most parameters are the same as in the "grey alloy" model
  - In particular SIA clusters are slowed down by Cr
- C atoms are simulated as traps
- Each time a point-defect finds a (*dilute*) solute, a new object (solute-Va pair, mixed dumbbell, …) is created
  - The binding energy defines for how long the point-defect drags the solute, i.e. until dissociation
  - Correlation effects with surrounding solutes are disregarded

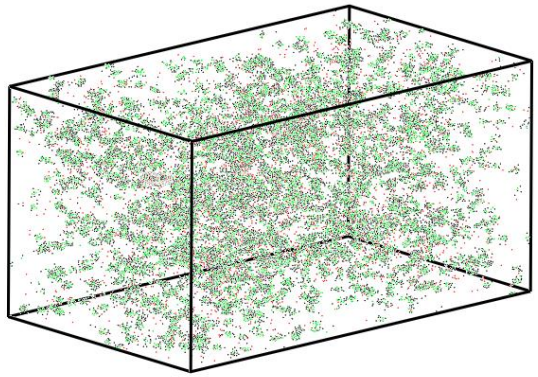
Only single defects transport solutes: SIA transport P, Va transport Si, P & Ni

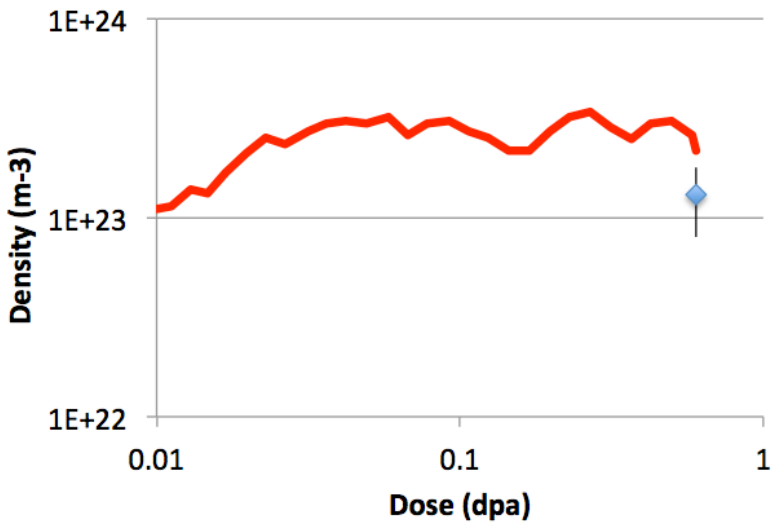
- Ingredient to be further studied: loops can be also trapped by *(dilute)* solutes: Ni, Si, **P**

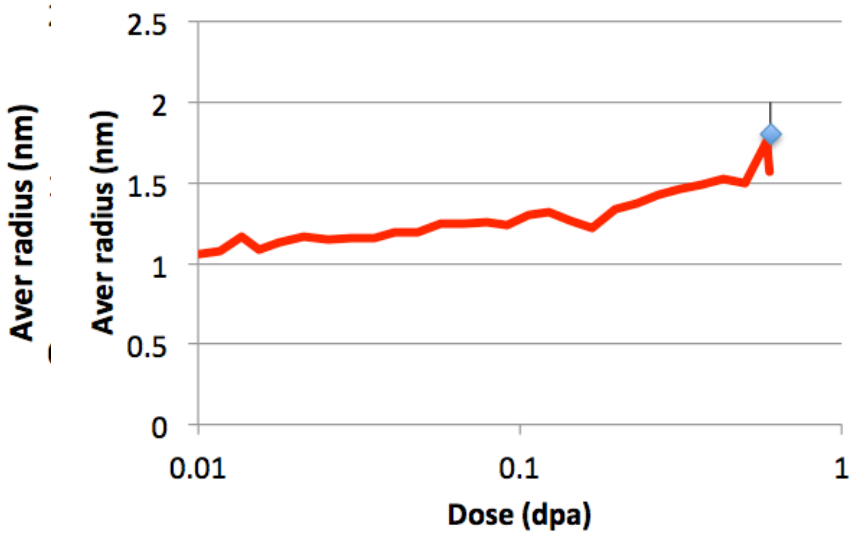
First results: Fe-2.5%Cr-0.1%Si-0.02%P

After processing the simulation box with a procedure that emulates the APT:

- The density of solutes-rich clusters compares well with experimental values.
- The radius is underestimated: explained by the absence of Cr.



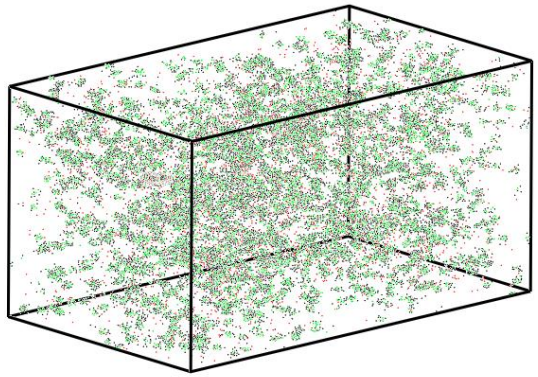


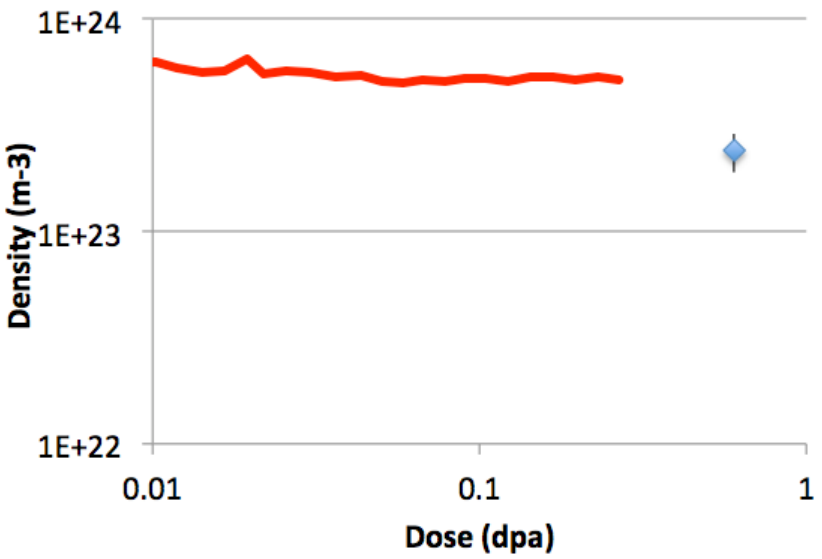


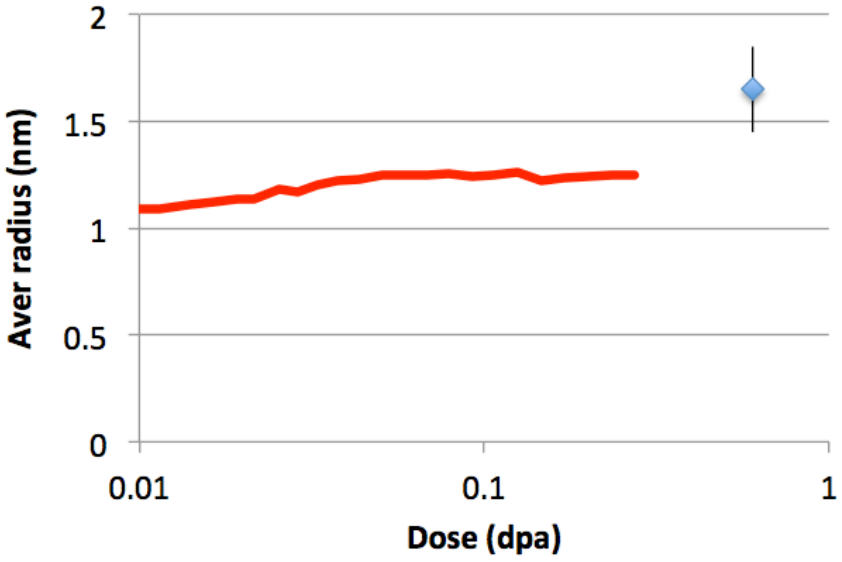
First results: Fe-9%Cr-0.06%Ni-0.07%Si-0.01%P

After processing the simulation box with a procedure that emulates the APT:

- The Density of solutes-rich clusters compares well with experimental values.
- The radius is underestimated: explained by the absence of Cr.







First results: Chemical composition

The chemical composition of the solute-rich cluste
however, is not in agreement with APT:

- Absence of Cr by construction of the model
- Minor solutes in different proportion:
  - Revise compositions?
  - Inaccurate description of solute transport?
  - Needs for accurate description of cluster free energy and cluster dissolution mechanisms?

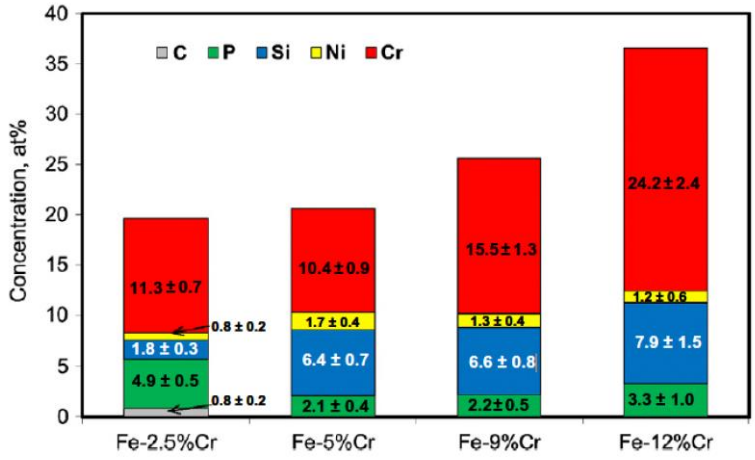
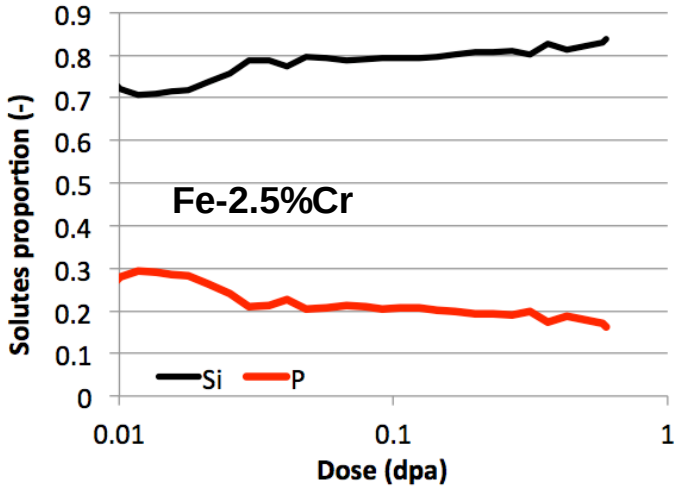
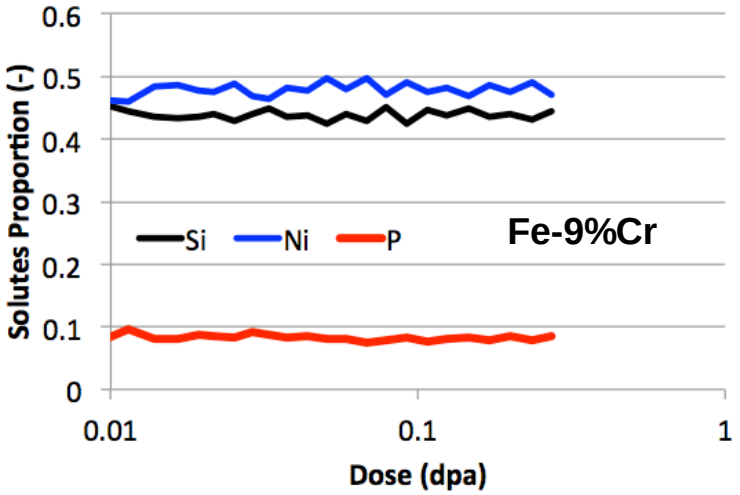


Fig. 3. Composition of the NiSiPCr-enriched clusters (at.%) in the different model alloys.





Summary and Outlook

Model of microstructural evolution under irradiation of F/M steels

- Grey alloy model:
  - Cr affects mainly mobility of SIA clusters
  - Correct trends reproduced / solute clusters assessed as invisible loops
- Introduction of (dilute) solute transport:
  - Ni, Si, P dragged by point defects, accumulate on point-def clusters
  - Quantitative assessment of (dilute) solute enrichment is possible
- Future perspectives
  - Introduce cellular model for Cr precipitation and redistribution