Radiation damage in FeCr alloys for nuclear applications: Addressed problems and open issues

D. Terentyev, P. Olsson (EDF) et al.
### Structural materials requirements for nuclear applications

<table>
<thead>
<tr>
<th></th>
<th>Fission (Gen. I/II)</th>
<th>Fission (Gen. IV)</th>
<th>Fusion (Demo)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Structural alloy maximum temperature</td>
<td>&lt;300°C</td>
<td>500-1000°C</td>
<td>550-1000°C</td>
</tr>
<tr>
<td>Max dose for core internal structures</td>
<td>~1 dpa</td>
<td>~30-100 dpa</td>
<td>~150 dpa</td>
</tr>
<tr>
<td>Max transmutation helium concentration</td>
<td>~0.1 appm</td>
<td>~3-10 appm</td>
<td>~1500 appm</td>
</tr>
<tr>
<td>Coolants</td>
<td>H₂O</td>
<td>He, H₂O, Pb-Bi, Na</td>
<td>He, Pb-Li, Li</td>
</tr>
<tr>
<td>Structural Materials</td>
<td>Zircaloy, stainless steel</td>
<td>Ferritic steel, SS, superalloys, SiC-composite</td>
<td>Ferritic/ martensitic steel, V alloy, SiC composite</td>
</tr>
</tbody>
</table>

- Good swelling resistance
- Fracture toughness & acceptable shift of DBTT (lower temperature bound)
- Good corrosion properties
- Creep resistance (upper temperature bound)
Irradiation-induced hardening and related embrittlement in Ferritic/Martensitic Steels

- Irradiation below 425-450°C causes substantial hardening
  - observed as increase in strength
  - accompanied by decrease in ductility
  - can lead to flow localization, dislocation channel deformation

- Hardening causes a loss of impact toughness
  - observed as an increase in ductile-brittle transition temperature (DBTT) in a Charpy impact test
  - observed as a decrease in upper-shelf energy (USE)

- However, composition of steel and Cr content may seriously affect ΔDBTT

\[ \Delta \sigma = 210 \text{ MPa} \]
\[ \Delta \sigma = 60 \text{ MPa} \]

M.A. Sokolov, 2000

A number of early and recent experiments suggest that Cr content in FeCr alloys influences significantly radiation-induced microstructure, swelling, and DBTT. Interpretation of available results and understanding their interconnection is a challenge. FeCr alloys are therefore model materials to understand the mechanisms responsible for the non-linear dependence on Cr content of many properties of high Cr steels.
The multi-scale modelling ambition

- Models are simplified and idealized so they include the main driving mechanisms only.
- Each model is valid in its own scale, but overlap between the models is necessary for cross-check.
- Experiments are also needed to validate models.
- Building up a multi-scale pyramid we unavoidably accumulate an error.
- Predictions with multi-scale approach can only be made if: mechanisms are understood and correctly quantified.
- Modelling is first and foremost about identifying mechanisms.

Evolution of Radiation Damage in Fe-Cr alloys is addressed using:

\textit{Ab initio} – MD – kMC – Rate Theory
Issues addressed using different modelling techniques

A comparative study ‘Fe vs. Fe-xCr’ was performed to understand impact of Cr content on the production and accumulation of radiation damage

Relevant range of Cr content: 5-15%

<table>
<thead>
<tr>
<th>Characterization of:</th>
<th>Technique</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr distribution in Fe-Cr alloys</td>
<td>Ab initio</td>
</tr>
<tr>
<td>Primary Damage</td>
<td>MD</td>
</tr>
<tr>
<td>Migration of point defects and interaction with Cr solutes</td>
<td><em>Ab initio</em>, MD, kMC</td>
</tr>
<tr>
<td>Migration of interstitial defects</td>
<td>MD</td>
</tr>
<tr>
<td>Mutual interaction between 1D mobile interstitial clusters</td>
<td>MD</td>
</tr>
<tr>
<td>Interaction of defects and Cr-precipitates</td>
<td>MD</td>
</tr>
<tr>
<td>Isochronal annealing</td>
<td>Rate Theory</td>
</tr>
</tbody>
</table>
1.1 DFT. Heat of mixing (ferromagnetic FeCr)

1) 1983: Henion: Generalized perturbation c-ns
   “Effective pair potential changes its sign in Fe-Cr at 25%”

2) 1984: Mirebeau: Diffuse Neutron Scattering m-ts
   “Change of the sign of SRO parameter from Negative to Positive at 10%Cr”

3) 2000: Filippova: Mösbauer m-ts of irradiated alloys
   “Change of SRO sign at 9%”

4) 2003: Olsson: DFT c-ns
   “The heat of mixing changes sign at 6%”

- DFT data published within 2003-07 do scatter, as different approaches were used
- All agree on change of sign at ~6-10%
- Heat of mixing from CALPHAD does not include this feature!
1.2: DFT. Impact of the calculated heat of mixing

- **Green points** – observation of Cr ordering
- **Black points** – observation of Cr precipitation
- **Red points** – miscibility gap as suggested by investigators

Can we clarify these issues using experimental data?

Especially phase diagram!
2.1. Development of interatomic potential for Fe-Cr

\[
E = \sum_i^N \left[ F \left( \sum_{j \neq i} \rho(r_{ij}) \right) + \frac{1}{2} \sum_{j \neq i} V(r_{ij}) \right]
\]

- Standard EAM is not suitable to reproduce a mixing enthalpy whose shape deviates significantly from a skewed parabola

\[
E = \sum_i^N \left[ F^d \left( \sum_{j \neq i} \rho^d (r_{ij}) \right) + F^s \left( \sum_{j \neq i} \rho^s (r_{ij}) \right) + \frac{1}{2} \sum_{j \neq i} V (r_{ij}) \right]
\]

- Idea was to introduce a term perturbation the heat of mixing, thus the many-body part was split into two, so called: “d”- and “s”- bands.

\[
H_{mix}^d = 1 - \sqrt{C_B^2 + C_B (1 - C_B) + (1 - C_B)^2}
\]

“d” yields positive heat of mixing with parabolic shape

\[
H_{mix}^s = -\sqrt{0.01 C_B (1 - C_B)}
\]

“s” contributes the heat of mixing, providing change of sign

Besides \( H_{mix} \), potential includes:

1) Fe part from Ackland’04 : (good interstitial properties)
2) Solute - point defect interaction energies from DFT
3) Elastic constants & cohesive energy (pure metals)
2.2. Validation of Fe-Cr potential: phase diagram

- Metropolis Monte Carlo method can be used to estimate phase boundary with POTENTIAL

Main steps to build DIAGRAM:

1. Fit cluster expansion to be used in Metropolis Monte Carlo
2. Estimate vibrational entropy and correct cluster expansion
3. Estimation of phase boundary using MMC using transmutation ensemble (V, µ-is fixed)
2.3. Validation of Fe-Cr POTENTIAL: interaction of point defects with Cr

Cr-point defect interaction: POTENTIAL VS. DFT

<table>
<thead>
<tr>
<th>Configuration</th>
<th>DFT</th>
<th>Potential</th>
</tr>
</thead>
<tbody>
<tr>
<td>SIA – Cr Interaction energy [eV]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>#1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>#2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>#3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>#4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>#5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>#6</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

To be noted:
1. Strong variation of $E_{\text{INT}}$ of SIAs depending on local atomic environment (Cr arrangement around a defect)
2. Negligible interaction with vacancy
3. Effect of Cr on the <111> crowdion is much stronger than on <110> dumbbell

<table>
<thead>
<tr>
<th>Vacancy – Cr interaction energy [eV]</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>#7</td>
<td>0.057</td>
</tr>
<tr>
<td>#8</td>
<td>0.014</td>
</tr>
</tbody>
</table>

(negative sign denotes attraction)
2.4. Validation of Fe-Cr POTENTIAL: point defect migration energies

- Migration of point defects: POTENTIAL vs. DFT

Translation – rotation jump (e.g. [110] -> [011]) provides the lowest migration energy ($E_{MIG}$)

- Fe-Fe dumbbell
  - $E_{MIG}(DFT) = 0.35$ eV
  - $E_{MIG}(POT) = 0.35$ eV

- Fe-Cr dumbbell
  - $E_{MIG}(DFT) = 0.23$ eV
  - $E_{MIG}(POT) = 0.24$ eV

- Cr-Fe dumbbell
  - $E_{MIG}(DFT) = 0.33$ eV
  - $E_{MIG}(POT) = 0.32$ eV

- Cr-Vacancy exchange
  - $E_{MIG}(DFT) = 0.55$ eV
  - $E_{MIG}(POT) = 0.56$ eV

- Fe-Vacancy exchange
  - $E_{MIG}(DFT) = 0.65$ eV
  - $E_{MIG}(POT) = 0.65$ eV

To be noted:

1. Preferential migration of Fe-Cr dumbbells (i.e. long range Cr-SIA drag)
2. Preferential exchange of Cr-Vacancy (but no drag – since no $E_{BIND}$)
3. Both $E_{MIG}$ for vacancy and SIA are lower by about 0.1 eV if Cr atom migrates

Presented results are valid in dilute alloys

What about concentrated alloys?
3.1. Primary damage in FeCr vs Fe: defect production

- Scarce effect of Cr on damage production
- Defects produced are:
  - **Interstitials**: single $<110>$ dumbbells 70-40%, $<110>$ clusters with size: 2÷5; $<111>$ with size 5÷50;
  - **Vacancies**: single 50%, spherical clusters with size 2÷40, $<100>$ platelets with size 20÷40 – very rare
### 3.2. Primary damage in Fe-Cr vs. Fe: effect of Cr

**Observed effects:**
- SIA defects (especially <111> clusters) are enriched by Cr
- Cascades induce SRO in Fe-5 and Fe-15Cr

**Origin of observed effects:**
- High binding energy <111>-Cr (0.4 eV)
- Negative ΔSRO (5%Cr): breakup of Cr-Cr 1st pairs
- Positive ΔSRO (15%Cr) formation of Cr clusters during solidification

#### ΔSRO parameter in random Fe-Cr alloys

<table>
<thead>
<tr>
<th>5 KeV cascade</th>
<th>Fe-5Cr</th>
<th>Fe-10Cr</th>
<th>Fe-15Cr</th>
</tr>
</thead>
<tbody>
<tr>
<td>ΔSRO/dpa×10⁻³</td>
<td>-7 ± 2</td>
<td>-2 ± 4</td>
<td>12 ± 2</td>
</tr>
</tbody>
</table>

**Chemical composition of interstitials**

![Graph showing chemical composition of interstitials](image)
3. Primary damage Fe vs. Fe-Cr: Summary

Common features of primary damage in both Fe & Fe-Cr

1) Defect production efficiency saturates at 0.3
2) Fraction of clustered vacancies ~0.5, weakly depends on $E_{\text{PKA}}$
3) Fraction of clustered SIAs growth monotonically with $E_{\text{PKA}}$ from 0.3 to 0.5
4) SIAs in clusters are:
   - relatively large <111>, containing up to 50 defects
   - small <110>, containing up to 5 defects
5) Vacancies in clusters are:
   - 95% of vacancy clusters are nano-voids
   - <100> vacancy loops are produced only in dense cascades

Specific features of primary damage in Fe-Cr alloys:

1) Formed SIA and SIA clusters are preferentially bounded to Cr atoms in Fe-5,10 but not in Fe-15Cr !
2) Rearrangement of Cr atoms occurs. In Fe-5Cr, Cr atoms have tendency to order, whereas in Fe-15Cr to precipitate

Variation of Cr ‘enrichment’ at SIA defects is the only pronounced effect detected
4.1 Mobility of interstitial defects: pure Fe

MD allows to define migration mechanism and estimate corresponding diffusion coefficient \((D)\).

In pure Fe, SIA defects can be subdivided as:

1) 3D mobile: <110> dumbbells and small clusters
2) 3D-1D mobile: clusters of transition size, containing 4 and 5 defects
3) 1D mobile: <111> clusters with size > 5 SIAs

Note that 1D mobile SIA clusters move almost athermally with \(E_{MIG} \sim 0.05\) eV.
4.2 Interstitial defects: single SIA: Fe vs. FeCr

MD allows to study space of few hundreds of nm$^3$, for a period of time $\sim 100$ ns

1. Static calculations show that SIA can be trapped in some Cr configurations, $E_{TR} \sim 0.2-0.4$ eV

2. Concentration of these traps $\sim (1-100) \times 10^{-6}$

3. Escape from these traps requires time $\sim \exp\left(-\frac{E_{TR}+E_{MIG}}{k_B T}\right)$, which could easily exceed 100 ns

<table>
<thead>
<tr>
<th>Material</th>
<th>$E_{mig}$ (eV)</th>
<th>$D_0$ (cm$^2$/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>0.31</td>
<td>4.43 $\times$ 10$^{-3}$</td>
</tr>
<tr>
<td>Fe-0.2%Cr</td>
<td>0.29</td>
<td>1.60 $\times$ 10$^{-3}$</td>
</tr>
<tr>
<td>Fe-5%Cr</td>
<td>0.28</td>
<td>7.26 $\times$ 10$^{-4}$</td>
</tr>
<tr>
<td>Fe-7.5%Cr</td>
<td>0.26</td>
<td>4.02 $\times$ 10$^{-4}$</td>
</tr>
<tr>
<td>Fe-10%Cr</td>
<td>0.25</td>
<td>1.34 $\times$ 10$^{-4}$</td>
</tr>
<tr>
<td>Fe-15%Cr</td>
<td>0.23</td>
<td>9.33 $\times$ 10$^{-5}$</td>
</tr>
</tbody>
</table>
4.3. Mobility of interstitials in Fe-Cr vs. Fe: traps for small SIA clusters

All characteristic energies are functions of the local atomic configuration (LAC), thus in concentrated Fe-Cr alloys: 
There is no ‘migration’ or ‘binding’ energy’ but a distribution of such energies

1. Problem now can be formulated as: estimation of the LAC-dependent $E_{\text{MIG}}$ for a given migration mechanism

2. Whether SIA-Cr drag occurs in concentrated Fe-Cr alloys is a question
Formation of Cr-Cr dumbbells is unfavourable in both dilute and concentrated alloys
4.4. Mobility of interstitials in Fe-Cr vs. Fe:
1D migrating SIA clusters

First MD results in Random Fe-Cr alloys have shown that:

The origin of reduction in the diffusivity of SIA clusters stems from strong interaction of Cr with <111> crowdions!

1. Presence of Cr drastically reduces mobility of 1D-migrating SIA clusters

2. Migration energy (and slowing down) is an interplay of cluster size and Cr content

Assessment of SIA mobility (above MD scale) requires understanding of the migration mechanism i.e. characterization of cluster-Cr interaction
4.5. Mobility of interstitials: migration of $[111]$ clusters

![Graph showing binding energy vs. distance along [111] direction](image)

$$K_n = nn_I C_{Cr}$$

- $n$ – number of SIAs in cluster
- $n_I$ – length of interaction

![Diagram showing Cr-SIA cluster interaction](image)
4.6. Mobility of interstitials: estimation of diffusion coefficient in Fe-Cr alloys

Variation of the SIA-cluster free energy can be estimated as

\[ \Delta F \approx k_B T \ln \langle \exp\left(-\frac{E_B}{k_B T}\right) \rangle \]

Reduction of the diffusion coefficient is then related to \( \Delta F \)

1. MD confirms results of static calculations
2. Such approach can be applied to an alloy with any distribution of solutes, so far the mechanism of migration is independent motion crowdions
4.7. Summary: mobility of SIAs in Fe vs. Fe-Cr alloys

➢ In the presence of Cr:

- Single SIAs (as <110> Fe-Cr dumbbells) migrate slightly faster (with lower migration energy), but are expected to get trapped.
- Approximate trapping energy ($E_{TR}$) and concentration ($C_{TR}$) for single and 3D-mobile SIA clusters is estimated to be: $E_{TR} = 0.2-0.8$ eV $C_{TR} = 10^2 - 5 \times 10^3$ ppm, in alloys with 5-15%Cr.
- 1D – migrating <111>-SIA clusters are slowed down in Fe-Cr alloys.
- For each cluster size there is $MIN(D_{FeCr}/D_{Fe})$ which depends on:
  - Cr content
  - Temperature
  - Cr distribution

Important to note that:

1. $MIN(D_{FeCr}/D_{Fe})$ for small (in-cascade-created) SIA clusters stays at: 5-10%
2. Cr ordering (negative SRO parameter) makes $\Delta F$ independent on cluster size
3. $\alpha-\alpha'$ separation makes $D_{FeCr}/D_{Fe}$ (for clusters moving in the depleted matrix) independent on initial Cr content.
5. Isochronal defect recovery in Fe-Cr vs. Fe: Example of pure Fe

Interstitial mobility by *ab initio*:
- Single SIA: 0.34 eV
- Di-SIA: 0.42 eV
- Tri-SIA 0.43 eV, larger - immobile

Vacancy mobility by *ab initio*:
- Single Vacancy: 0.67 eV
- Di-Vacancy: 0.62 eV
- Tri-Vacancy: 0.35 eV
- 4-Vacancy: 0.48, larger - immobile

C.C Fu et al., Nature Materials 2004
5. Isochronal defect recovery in Fe-Cr vs. Fe: Application of rate theory

- **Rate theory**: basic ideas
  
  \[ A + B \xrightleftharpoons[k_C^-]{k_{A+B}^+} C \]

  \[
  \frac{\partial C_C}{\partial t} = G_C + D_C \nabla C_C - \left( k_{A+B}^+ C_A C_B - k_C^- C_C \right)
  \]

  \[
  k_{A+B}^+ = 4\pi (r_A + r_B) (D_A + D_B)
  \]

  \[
  k_C^- \propto k_{A+B}^+ \exp\left(-\frac{E_b}{kT}\right)
  \]

- Such studies allow effective parameters to be obtained, the local chemistry (atoms) are not explicitly allowed for

  - Capture radii
  - Diffusion coefficients
  - Binding energies

  **Problem of describing 1D-1D and 1D-3D reactions is to be addressed !!!**
Gathering data about SIA and vacancy mobility presented above, we recalculate defect recovery spectra for Fe-Cr alloys.

1. Traps for SIA defects were introduced explicitly (i.e. as objects pinning defects for a time $\sim \exp(-E_{TR}/kT)$)
2. No 1D motion of clusters is allowed (i.e. clusters with size >5 are immobile)
3. Possible phase separation or ordering effects are not treated.

With increase of Cr content:
1. $T$ stage $I_E$ ↓
2. $T$ stage II ↑
3. $T$ Stage III ↔

But we can’t hope to reproduce the full complexity of configurational traps.
5. Isochronal defect recovery: Summary of defect properties

- In pure Fe
  - Good agreement with experiment can be obtained providing that:
    - $E_{\text{MIG}}(\text{SIA})=0.33 \text{ eV}$, $E_{\text{MIG}}(\text{Di-SIA})=0.42 \text{ eV}$
    - Assumption about immobility of SIA clusters (>5-SIAs) is necessary
    - Ab initio calculations predict $E_{\text{MIG}}(\text{VAC})=0.65 \text{ eV}$, whereas agreement with experiment is achieved only if $E_{\text{MIG}}(\text{VAC})=0.55 \text{ eV}$

- In Fe-Cr alloys, with 5-15% Cr
  - Trends as seen in experiments can be obtained if:
    - $E_{\text{MIG}}(\text{SIA})\approx 0.25 \text{ eV}$, and two classes of traps are introduced:
      - Shallow: $E_{\text{TR}}\approx 0.2 \text{ eV}$
      - Deep: $E_{\text{TR}}\approx 0.4 \text{ eV}$
    - Concentration of deep traps increases with Cr content
    - $E_{\text{MIG}}(\text{VAC})\approx 0.55$, i.e. unaffected in the presence of Cr

- Comparison of RT study and experimental recovery spectra, obtained in concentrated alloys, suggest:
  - Mobility of single vacancy is not affected by Cr
  - Mobility of single SIAs is reduced progressively by increasing Cr content
  - Onset of long-range SIA migration occurs above stage III (after vacancy migration !)
  - Stage II has different origin in Fe and Fe-Cr alloys
  - Stage III is vacancy migration and release of SIAs from deep traps

Main problem of RT approach is to include spatial correlations and specific reactions with 1D mobile objects

Kinetic Monte Carlo techniques wherein atoms or defects are considered as objects solves this problem, in principle...
6.1. Basics of MC

- **Object kinetic Monte Carlo**: basic ideas
  - Each object defined by:
    - type
    - centre-of-mass position
    - reaction radius
    - possible reactions
    - diffusion coefficient
  - All possible events must be known in advance
    - A probability associated to each event
    - \( \sum_i P_i = 1 \)
    - Monte Carlo uses the same ingredients as RT;
      - However, Object kMC is not suitable to study concentrated alloys on a large scale

Kinetic Monte Carlo uses the same ingredients as RT;
But, the outcome includes spatial correlations
6.2 Problems of kMC applied to concentrated alloy

1. Evolution of the system is governed by thermodynamics: minimization of the free energy

2. Kinetics is governed by: migration mechanisms and corresponding migration barriers

3. In concentrated alloys there is no unique migration barrier for a given defect, but there is a spectrum

4. Conventional way to treat alloys is to estimate $E_{\text{MIG}}$ as:

$$E_{\text{MIG}} = E_0 + \frac{\Delta E}{2}$$

$E_0$(Cr-V) = 0.55 eV

$E_0$(Fe-V) = 0.66 eV

More precise estimation can be done with Nudged Elastic Band method

The conventional way to estimate migration energy is not appropriate!
6.3. Artificial Intelligence in the kMC framework

The systematic on-the-fly use of the NEB method within the AKMC is absolutely unfeasible in practice ...

We propose the use of an Artificial Neural Network (ANN) instead.

One can describe Local Atomic Configuration (LAC) by digital code containing information about positions of different chemical species of surrounding a defect

Function of ANN: is to obtain a correlation between

1. Establishment of the training set
2. Benchmarking of the generated set

Example: 1Vac in Fe-10%Cr
39 Variables
Mean error = 0.49%

Speed up factor = $10^6$
Combination of standard kMC and Artificial Intelligence can be used

- as engine generating $E_{\text{MIG}}$ for kMC
- or to study:
  1. Migration of point defects in concentrated alloys (e.g. removing problem of SIA-trapping)
  2. Migration of slow defects clusters (such vacancy clusters, or vacancy-solute clusters, surface diffusion), which is inaccessible on MD scale

The ANN approaches allows to speed-up the kMC by a factor $10^6$, but some limitations also exit:

- Large number of input variables makes **training a long operation.** => parallelization allows to reduce the problem
- NEB database must be carefully chosen to be representative of the Local Atomic Environments that are encountered during the AKMC simulation. => feedback from kMC.

Study of phase separation accelerated by irradiation is in progress ...

However, the problem of Cr precipitate defect interaction can be already addressed...
7. Effect of phase separation on defect mobility

- Cr precipitation is usually observed in binary FeCr and Ferritic/Martensitic steels as:
  - Nearly pure Cr spherical precipitates
  - Onset occurs with high density (up to $10^{24} \text{ m}^{-3}$) and small mean size (few nm)

- Regarding evolution of $\mu$-structure it is important to envisage:
  - Whether Cr precipitates may act sinks for 3D mobile defects
  - How Cr precipitates interaction with 1D mobile SIA clusters

- Main characteristics of pure Cr vs. Fe
  - $\Delta a_0 = 2.8845 - 2.8665 = +0.018 \text{ A}$
  - $\Delta E_{coh} = 4.10 - 4.29 = -0.2 \text{ eV}$
  - $\Delta \mu = 110 - 80 = +30 \text{ GPa}$
    (point defect energies from ab initio calculations)
  - $\Delta E(Vacancy) = 2.64 - 2.15 = +0.5 \text{ eV}$
  - $\Delta E(<110> - \text{dumbbell}) = 5.68 - 3.75 = +1.93 \text{ eV}$
  - $\Delta E(<111> - \text{crowdion}) = 5.76 - 4.45 = +1.31 \text{ eV}$
7.1. Effect of phase separation on defect mobility: Main observations

- **SIAs and small SIA clusters:**
  - Are repelled by Cr precipitates, formation of interstitial defects inside precipitates is unfavourable.

- **Vacancies:**
  - Are attracted at the precipitate-matrix interface, but do not enter inside; formation of voids attached to Cr precipitates is not favourable.

- **1D SIA clusters:**
  - Exhibit long-range repulsive interaction.

- **Cascades near Cr precipitates:**
  - No effect on the defect production, but cascade splitting is enhanced.

**Interaction of nanometric Cr precipitates with 1D-SIA clusters**
7.2. Effect of phase separation on defect mobility: Summary

- Effect of phase separation on defect mobility can be summarized as follows:
  - Depletion of matrix by Cr down to ~10% changes distribution and strength of traps for 3D-migrating interstitials as well as \( \Delta F \) for 1D-migrating loops
  - 3D-migrating SIA and their clusters will be scattered at Cr precipitates, vacancies could be bound at the precipitate-matrix interface (with binding energy ~0.1 eV)
  - Cr precipitates with size >1nm, act as insurmountable obstacles for 1D-migrating SIA clusters and small dislocation loops (e.g. 4nm)

- Main effect of \( \alpha-\alpha' \) separation on the defect mobility is:
  - Blockage of 1D migrating clusters

- As a consequence of that (indirect effect):
  - Increase in the density of loops
  - Increase of interstitial sites for vacancy to recombine
  - Faster growth of dislocation network

Note that while \( \alpha-\alpha' \) separation advances (coarsening stage), its effect on blockage of 1D clusters vanishes
Formulation of the basic principles

What atomistic modelling provided us:

- Cr negligibly interacts with vacancies, but progressively reduces mobility of interstitials
- Below 8-10%, Cr trend to order, above to precipitate. Both processes are expected to be accelerated by irradiation
- In-cascade created 1D-migrating SIA clusters are effectively slowed down by Cr.

Factor of slowing $S = D_{FeCr}/D_{Fe}$ behaves as:

$$\Delta F \approx k_B T \ln < \exp(\Delta E^B / k_B T) >$$

What can be explained using these hints provided by modelling…
Swelling in Fe-Cr alloys

Swelling dependence on %Cr, from Little & Stow
JNM, 87 (1979) 25, as in Garner, JNM 276 (2000)

Lower T:
Minimum 5 -10%Cr increase of size clusters?

Higher T:
Minimum at 15%
accelerated \(\alpha\)’ formation
locking SIA clusters?

Decrease of SIA diffusivity
In random alloys

If phase separation occurs

Phase diagram

Dislocation network

<100> dislocation loops

Ordering/precipitation
1D-SIAs

S=\(\frac{D_{FeCr}}{D_{Fe}}\)

Temperature (K)

Cr content (%)

Number density (m^{-3}) * 10^{21}

Mean radius

Dose 140 dpa

Ordering/precipitation
1D-SIAs

Swelling dependence on %Cr, from Little & Stow
JNM, 87 (1979) 25, as in Garner, JNM 276 (2000)

Lower T:
Minimum 5 -10%Cr increase of size clusters?

Higher T:
Minimum at 15%
accelerated \(\alpha\)’ formation
locking SIA clusters?

Decrease of SIA diffusivity
In random alloys

If phase separation occurs

Phase diagram

Dislocation network

<100> dislocation loops

Ordering/precipitation
1D-SIAs

S=\(\frac{D_{FeCr}}{D_{Fe}}\)

Temperature (K)

Cr content (%)

Number density (m^{-3}) * 10^{21}

Mean radius

Dose 140 dpa

Ordering/precipitation
1D-SIAs
Outstanding and primary issues in modelling of radiation damage in FeCr alloys

- **Interatomic potentials + Molecular Dynamics:**
  - Origin and properties of <100> SIA loops (ratio of <111>/<100>)
  - Development of MD including spin term, which is believed to be enough to allow for α-γ transition (so effect of magnetism could be treated)

- **Atomistic Kinetic Monte Carlo:**
  - Correct treatment of defect mobility in concentrated alloys + effect of vibrational entropy

- **Coarse-grain models (such as Object Kinetic Monte Carlo):**
  - Introduction of α-α’ phase separation accelerated in the presence of irradiation: formation of Cr precipitates (and coarsening), induced negative SRO, Cr mass transport
  - Dependence of defect mobility/stability on α-α’ phase separation/ordering
The study of the behaviour of FeCr alloys under irradiation has shown that three basic properties of Cr in Fe, namely:

- Repulsion between Cr-Cr pairs in dilute Fe-Cr alloys (until screened by large concentration)
- Attraction between dispersed Cr atoms and self-interstitial defects in Fe
- Repulsion between Cr precipitates and self-interstitial defects in Fe

... seem to explain a large amount of phenomena

As it often happens the gap between stating the basic principles and applying them to obtain quantitative prediction remains large...

Yet, understanding of the basic principles allows to interpret and gather different experimental results into “one story”