Application of a three-feature dispersed-barrier hardening model to neutron-irradiated FeCr alloys

F. Bergner ¹, M. Hernández Mayoral ², L. Malerba ³, C. Pareige ⁴

¹ HZDR Dresden, Germany
² CIEMAT Madrid, Spain
³ SCK·CEN Mol, Belgium
⁴ GPM, Université et INSA de Rouen, CNRS, France

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Outline

1 Introduction

2 Model

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   - Tensile tests
   - TEM
   - APT
   - SANS

4 Best rms fit, results and discussion

5 Conclusion and outlook
Introduction

Industrial purity Fe-Cr alloys fabricated by SCK-CEN, irradiated in the BR2 reactor

Tensile tests performed at SCK-CEN → Yield stress as function of neutron exposure, Cr and test temperature and first TEM.

EU FP7 project GETMAT → SANS performed by HZDR at GKSS Geesthacht
APT performed by and at Uni de Rouen/CNRS
TEM performed by and at CIEMAT … + others

Three kinds of irradiation-induced nanofeatures detected and quantified:
   a‘ phase particles,
   NiSiPCr enriched clusters,
   interstitial dislocation loops.

Resulting question: Are the observed features potentially sufficient to explain the observed hardening?

Application of a three-feature dispersed-barrier hardening model.
Work in progress !!!
Model

Random distribution of barriers of type $x$, number density $N_x$ and diameter $d_x$.

BKS model (Bacon, Kocks, Scattergood, Phil. Mag. 1973):

$$\Delta \sigma_{yx} = f(N_x, d_x) \equiv \alpha_x \frac{M \mu b}{2 \pi l} \left[ \ln \left( \frac{b}{l} \right) \right]^{-1/2} \left[ \ln \left( \frac{d'}{b} \right) \right]^{3/2}$$

$$l = \frac{1}{\sqrt{N_x d_x}}$$

$$d' = \frac{d_x l}{d_x + l}$$

$M = 3.06$ Taylor factor.
$\mu = 84000$ MPa shear modulus.
$b = 0.248$ nm Burgers vector.
$\alpha_x$ unknown dimensionless obstacle strength.

Simplified version (periodic array of obstacles, no self-stress of the dislocation):

$$\Delta \sigma_{yx} = f_s(N_x, d_x) \equiv \alpha_s M \mu b \sqrt{N_x d_x}$$
Model

Superposition rules:

(1) Linear superposition of pre-existing barriers and irradiation-induced barriers:

\[ \Delta \sigma_y = \sigma_{y,i} - \sigma_{y,u} \]

(2a) Linear superposition of barriers of different kinds:

\[ \Delta \sigma_y = \alpha_A f(N_A, d_A) + \alpha_L f(N_L, d_L) + \alpha_C f(N_C, d_C) \]

(2b) Square superposition of barriers of different kinds:

\[ (\Delta \sigma_y)^2 = [\alpha_A f(N_A, d_A)]^2 + [\alpha_L f(N_L, d_L)]^2 + [\alpha_C f(N_C, d_C)]^2 \]
Model

List of assumptions (incomplete):

(1) Uniform and random distributions of barriers (observed non-uniformity smeared out).
(2) A number of three distinguished families of barriers.
(3) Families represented by two parameters: Number density and mean size (size distributions ignored).
(4) Values of $M, \mu$ and $b$ (crystallographic anisotropy and variety of slip systems ignored).
(5) Underlying superposition law.
...

(far too many)
Input parameters

Materials and irradiations:

Composition (wt%)

<table>
<thead>
<tr>
<th>Material</th>
<th>Mn</th>
<th>Si</th>
<th>P</th>
<th>S</th>
<th>Al</th>
<th>Ti</th>
<th>Cr</th>
<th>Ni</th>
<th>O</th>
<th>C</th>
<th>N</th>
<th>V</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe2.5Cr</td>
<td>0.09</td>
<td>0.02</td>
<td>0.013</td>
<td>0.002</td>
<td>0.003</td>
<td>0.004</td>
<td>2.4</td>
<td>0.044</td>
<td>0.035</td>
<td>0.008</td>
<td>0.0117</td>
<td>0.001</td>
</tr>
<tr>
<td>Fe5Cr</td>
<td>0.02</td>
<td>0.04</td>
<td>0.011</td>
<td>0.006</td>
<td>0.0033</td>
<td>0.0028</td>
<td>4.6</td>
<td>0.06</td>
<td>0.065</td>
<td>0.02</td>
<td>0.0127</td>
<td>0.001</td>
</tr>
<tr>
<td>Fe9Cr</td>
<td>0.03</td>
<td>0.09</td>
<td>0.012</td>
<td>0.00066</td>
<td>0.0069</td>
<td>0.0034</td>
<td>8.4</td>
<td>0.07</td>
<td>0.066</td>
<td>0.02</td>
<td>0.0148</td>
<td>0.002</td>
</tr>
<tr>
<td>Fe12Cr</td>
<td>0.03</td>
<td>0.11</td>
<td>0.05</td>
<td>0.006</td>
<td>0.003</td>
<td>0.0037</td>
<td>11.6</td>
<td>0.09</td>
<td>0.03</td>
<td>0.027</td>
<td>0.0237</td>
<td>0.002</td>
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</table>

Austenitization: 1050 °C/1 h, tempering: 730 °C/4 h, air cooling

Neutron irradiation in the BR2 reactor:
- Irradiation temperature 300 °C.
- Neutron flux was $9 \times 10^{13}$ cm$^{-2}$ s$^{-1}$.
- Neutron exposure 0.06 dpa, 0.6 dpa, 1.5 dpa (nominal).

More details:
## Input parameters

Overview of methods applied so far (PAS not included):

<table>
<thead>
<tr>
<th>wt% Cr</th>
<th>dpa</th>
<th>TEM</th>
<th>APT</th>
<th>SANS</th>
<th>Tensile tests</th>
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</thead>
<tbody>
<tr>
<td>2.4</td>
<td>0.06</td>
<td>X</td>
<td></td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td></td>
<td>0.6</td>
<td>XX</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td></td>
<td>1.5</td>
<td>X</td>
<td></td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>4.6</td>
<td>0.06</td>
<td>X</td>
<td></td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td></td>
<td>0.6</td>
<td>XX</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td></td>
<td>1.5</td>
<td>X</td>
<td></td>
<td>X</td>
<td>X</td>
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<tr>
<td>8.4</td>
<td>0.06</td>
<td>XX</td>
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<td>XX</td>
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<td></td>
<td>1.5</td>
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<td>X</td>
<td>X</td>
</tr>
<tr>
<td></td>
<td>0.6</td>
<td>XX</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td></td>
<td>1.5</td>
<td>X</td>
<td></td>
<td>X</td>
<td>X</td>
</tr>
</tbody>
</table>

Work in progress
Input parameters

TEM

Performed at CIEMAT Madrid
TEM: JEOL model JEM-2010 operating at 200 keV (see talk by M. Hernández Mayoral, this workshop)

Findings:

• Non-uniform spatial distributions of loops observed for Fe5Cr, Fe9Cr and Fe12Cr.

→ Average density of loops calculated, treated as if they were uniformly distributed.

• Different families of loops not distinguished here.

• Mean diameter of loops measured from both BF images and WBDF images.

• Reasonable agreement between both measures and with earlier results reported by Matijasevic et al., JNM 377 (2008) 147 (within a factor of 2).

→ Size obtained in BF contrast taken as reference case here.
Input parameters

**APT**
- Energy Compensated Tomographic Atom Probe (ECoTAP)
- Energy Compensated Wide-Angle TAP (ECoWATAP)
- Laser Assisted Wide-Angle TAP (LAWATAP)

Performed at Université et INSA de Rouen/CNRS (see e.g. Kuksenko et al., *JNM 432 (2013) 160*)

**Findings:**
- Cr-rich clusters identified as $\alpha'$ for Fe9Cr and Fe12Cr.
- NiSiPCr-enriched clusters for each Cr content.
- Total number density and average size of NiSiPCr-enriched clusters
- Narrow zones free of clusters around grain boundaries.

…

(many more)
Input parameters

SANS

Performed by HZDR at SANS-2 facility of GKSS Geesthacht, $\lambda = 0.58$ nm, $^3$He-detector, 128 x 128 cells, saturation magnetic field (see e.g. Heintze et al., JNM 409 (2011) 106)

Findings:

• $\alpha'$-phase nanoparticles clearly identified for Fe9Cr and Fe12Cr.

→ Total volume fraction, mean size and number density calculated, statistically reliable average over a macroscopic volume, reasonable agreement with APT (within a factor of 2).

• Other minor detections in Fe2.5Cr, Fe5Cr and Fe9Cr might be attributed to NiSiPCr-enriched clusters and/or Cr-C-enriched loops.
Input parameters

Tensile tests

Performed at SCK·CEN using miniaturized round tensile bars. (see Matijasevic et al., JNM 377 (2008) 147)

Findings:

→ Yield stress increase measured as a function of Cr and neutron exposure.
• In some cases yield point phenomenon observed.
## Input parameters

### Summary

<table>
<thead>
<tr>
<th>Alloy specification</th>
<th>$\alpha'$ phase particles</th>
<th>Dislocation loops</th>
<th>NiSiPCr-rich clusters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr (wt %)</td>
<td>Exposure (dpa)</td>
<td>$\Delta\sigma_y$ (MPa)</td>
<td>$d_A$ (nm)</td>
</tr>
<tr>
<td>2.4</td>
<td>0.6</td>
<td>277</td>
<td>-</td>
</tr>
<tr>
<td>4.6</td>
<td>0.6</td>
<td>283</td>
<td>-</td>
</tr>
<tr>
<td>8.4</td>
<td>0.6</td>
<td>255</td>
<td>1.82</td>
</tr>
<tr>
<td>11.6</td>
<td>0.6</td>
<td>327</td>
<td>1.66</td>
</tr>
</tbody>
</table>

Over-determined system of linear equations (four equations, three unknowns, 16 measured quantities)

Reference case: (BKS model, linear superpos., BF contrast)

\[
\begin{pmatrix}
0 & 236 & 402 \\
0 & 167 & 496 \\
258 & 64 & 506 \\
1564 & 87 & 325 \\
\end{pmatrix}
\begin{pmatrix}
\alpha_A \\
\alpha_L \\
\alpha_C \\
\end{pmatrix}
= 
\begin{pmatrix}
277 \\
283 \\
255 \\
327 \\
\end{pmatrix}
\]
Best rms fit

Results: Reference case

<table>
<thead>
<tr>
<th>Superposition / contrast</th>
<th>Obstacle strength</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\alpha_A$</td>
<td>$\alpha_L$</td>
</tr>
<tr>
<td>Linear / BF</td>
<td>0.099 ± 0.003</td>
<td>0.513 ± 0.026</td>
</tr>
</tbody>
</table>

![Graph showing yield stress increase (MPa) for different Fe-Cr alloys.](image)

**F. Bergner et al., 4th nFAME and 22nd Workshop on Fe-Cr Alloys, Edinburgh, 4-5 June 2013**
Discussion

Reference case:

- The three-feature dispersed-barrier hardening model is sufficient to reproduce the measured yield stress increase within the range of measuring errors.
- The fitted model suggests that the dislocation loops are the strongest obstacles and the $\alpha'$-phase particles are the weakest obstacles to dislocation glide.
- However, each of the families of obstacles does contribute significantly to the yield stress increase of the set of Fe-Cr alloys.
- Absolute values of the obstacle strength must not be overstressed.
## Best rms fit

Results: Reference case and selected options

<table>
<thead>
<tr>
<th>Superposition / contrast</th>
<th>Obstacle strength</th>
<th>( R^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( \alpha_A )</td>
<td>( \alpha_L )</td>
</tr>
<tr>
<td>Linear / BF (Reference case)</td>
<td>0.099 ± 0.003</td>
<td>0.513 ± 0.026</td>
</tr>
<tr>
<td>Square / BF</td>
<td>0.177 ± 0.001</td>
<td>0.835 ± 0.032</td>
</tr>
<tr>
<td>Linear / WBDF</td>
<td>0.096</td>
<td>0.599</td>
</tr>
<tr>
<td><strong>Old set of data</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Linear / BKS</td>
<td>0.103 ± 0.004</td>
<td>0.755 ± 0.058</td>
</tr>
<tr>
<td>Square / BKS</td>
<td>0.178 ± 0.001</td>
<td>1.085 ± 0.063</td>
</tr>
<tr>
<td>Linear / Orowan</td>
<td>0.018 ± 0.003</td>
<td>0.425 ± 0.084</td>
</tr>
<tr>
<td>Square / Orowan</td>
<td>0.033 ± 0.001</td>
<td>0.533 ± 0.059</td>
</tr>
</tbody>
</table>
Discussion

Comparison of selected options (work in progress):

• The quality of fit does not depend significantly on the rule of superposition, i.e. the present set of data and the applied method cannot distinguish between linear and square superposition.

• The use of loop sizes derived from BF images gives much better „predictions“ than WBDF. This might be an indication for the TEM analysis but must not be overstressed, because the result may be biased by the assumptions.

• The BKS model gives much better fits than the Orowan model, whichever superposition rule is used. This might be an indication that dislocation self-interaction and randomness of obstacles (both not included in the Orowan model) do matter.
Conclusion

(1) The three-feature dispersed-barrier BKS-type hardening model is sufficient to reproduce the measured yield stress increase within error for neutron-irradiated Fe2.5Cr, Fe5Cr, Fe9Cr and Fe12Cr.

(2) TEM, APT, SANS and tensile tests are suitable to provide the required set of input data.

(3) However, the goodness of fit does not mean that a fourth family of obstacles could not exist and be operative.

(4) All model options tested so far agree in the ranking of obstacle strengths according to: Dislocation loops > NiSiPCr-enriched clusters > \( \alpha' \) particles. This ranking corresponds to expectation. However, the absolute values of the obstacle strength must not be overstressed.
Outlook

An increase of the degree of over-determination of the system of equations can be reached by adding the irradiation conditions (0.06 dpa, 1.5 dpa) not yet fully analyzed (work in progress).

The number of available materials and irradiation conditions is sufficient to evaluate a four-feature model.

The propagation of measuring errors is not yet included in the analysis, the given errors are just fit errors (work in progress).

Consideration of the source hardening model versus (or in combination with) the dispersed-barrier hardening model.