Microstructure Evolution in Fe and Fe-Cr Alloys with OKMC Methods

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The type of damage observed under transmission electron microscopy (TEM) in b.c.c. Fe and FeCr alloys under irradiation is very dependent on the irradiation conditions. At high doses the damage consists mostly of $\frac{1}{2}$ $\langle 111 \rangle$ and $\langle 100 \rangle$ loops, which are considered to be of interstitial type [1, 2]. Depending on the type of irradiation and the irradiation conditions, the ratio of one type of loop with respect to the other changes, with situations in which only one of the two types of loops are observed. The formation of $\langle 100 \rangle$ loops is still a controversial issue, with different models arising in the last few years. Experimentally, even the nature of these loops (vacancy or interstitial type) is still under debate under certain situations. Moreover, the type of irradiation (ion implantation vs. neutron irradiation) also changes the type and distribution of these defects.

In an attempt to shed some light into this complex system, we have developed models to study the damage of Fe and FeCr under irradiation. We combine results from density function theory (DFT), molecular dynamics (MD) and object kinetic Monte Carlo (OKMC) to study the production and evolution of the damage to time scales that can be directly compared to experiments. In this work we will discuss one particular issue: the differences between ion implantation and neutron irradiation. Currently, ion implantation is commonly used to obtain information about defect production and defect evolution, in an attempt to build models for neutron damage. We will point out the differences and commonalities between the two types of irradiation in the particular case of Fe, and those aspects that should be taken into consideration when building models for neutron irradiation from information obtained from ion implantation.

We will describe results of damage evolution in Fe using two different models for $\langle 100 \rangle$ loop formation: one where $\langle 100 \rangle$ loops are formed from the interaction between $\frac{1}{2}$ $\langle 111 \rangle$ loops (based on simulations by Marian [3] and Terentyev [4]) and a second one where two populations of loops are formed directly in the collision cascade and evolve independently. This last model is based on the observation of stable immobile clusters from DFT calculations [5]. The population of loops formed as a function of dose for different irradiation conditions is explored and results compared to experiments. The validity of the models and the influence of the different parameters are discussed. In particular, we will study how the mobility of $\langle 111 \rangle$ interstitial loops changes both the total concentration of visible defects as well as the ratio of $\langle 111 \rangle$ vs. $\langle 100 \rangle$ loops and compare the results with experimental data available, as shown in the Fig. 1. In this case the experiments are from reference [1].

Figure 1: OKMC simulations (solid lines) compared to experiments from ref. [1]

Finally we will discuss how object kinetic Monte Carlo can be modified to study high concentrated alloys for the particular case of FeCr.

References

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- [3] J. Marian, B. D. Wirth and J. Manuel Perlado, *Mechanism of formation and growth of <100> interstitial loops in ferritic materials*, Phys. Rev. Lett. 88, 25 (2002).
- [4] Haixuan Xu, Roger E. Stoller, Yuri N. Osetsky and Dmitry Terentyev, *Solving the puzzle of <100> interstitial loop formation in bcc iron*, Phys. Rev. Lett. 110, 265503 (2013).
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Microstructure Evolution in Fe and FeCr alloys with OKMC methods

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Motivation:

validate models with ion implantation - extrapolate to neutron irradiation

Reproduce and explain ion and neutron implantation experiments in Fe and FeCr

Z. Yao, et al., Phil. Mag. 88, 2851 (2008)

<http://jannus.in2p3.fr/>

Intermediate Voltage
Electron Microscope
(IVEM) Tandem
facility at ANL (USA)

:
<http://www.ne.anl.gov/ivem/>

MD database + DFT data → OKMC → damage in bulk and thin films

MD

+

DFT

OKMC

Exp

100 keV Fe recoil in bulk

70 nm

100 keV Fe recoil in thin film

- **Interstitials**
- **Vacancies**

Marinica PRL (2012)

New methods/models are emerging at all levels

Primary damage: influence on microstructure evolution

bcc Fe

Fe 30keV, 66 defects

fcc Cu

Cu 30keV, 69 defects

**Similar number of defects but more vacancy clustering in Cu than in Fe
This has important consequences in the subsequent damage evolution**

Damage accumulation in Cu and Fe

Damage accumulation in Cu and Fe compared to experiments

Fe calculations – N. Soneda

Simulations explain the basic differences observed experimentally in Cu and Fe: nucleation of vacancy clusters in Cu together with fast migration of self-interstitials, the presence of traps in Fe and the visibility under TEM

Influence of initial cascade damage distribution on damage accumulation (Carolina Björkas, Univ. Helsinki)

Question addressed: Is the long term evolution of defects affected by the picosecond cascade damage distribution or does it only depend on migration and binding energies?

30keV Fe in bcc Fe

OKMC calculations using cascade damage distributions from 3 different interatomic potentials, AMS [1], DD-BN [2,3] and MEA-BN [3, 4]

[1] G. J. Ackland, M. I. Mendelev, et al. J. Physics: Condens. Matter, 16 (2004) [2] S. L. Dudarev and P. M. Derlet. J. Phys.: Condens. Matter, 17 (2005) [3] C. Bjorkas and K. Nordlund, Nucl. Instrum. & Meth. B 259 (2007) [4] M. Muller, P. Erhart, and K. Albe, J. Phys.: Condens. Matter, 19 (2007)

NO EXPERIMENTAL VALIDATION OF MD RESULTS ON SINGLE CASCADE DAMAGE

Influence of initial cascade damage distribution on damage accumulation (Carolina Björkas, Univ. Helsinki)

OKMC simulations

TOTAL DEFECT CONCENTRATION:

no significant difference between the three potentials

$I > 5$ mobile $\langle 111 \rangle$ + traps (0.9 eV)

Influence of initial cascade damage distribution on damage accumulation (Carolina Björkas, Univ. Helsinki)

VISIBLE DEFECT CONCENTRATION:

only those clusters of interstitials > 55 (loop of 1nm radius)

only those clusters of vacancies > 350 (void of 1nm radius)

$I > 5$ mobile $\langle 111 \rangle$
+ traps (0.9 eV)

Large differences are now observed between the three potentials

Differences in cluster size distribution with int. potential (Carolina Björkas, Univ. Helsinki)

SIA MEA Potential

SIA AMS Potential

AMS potential predicts significantly larger self-interstitial clusters at 50keV cascades

Experimental validation of models

Multiscale modeling can only work with the proper experimental validation

Z. Yao, et al., Phil. Mag. 88, 2851 (2008)

Intermediate Voltage
Electron Microscope
(IVEM) Tandem
facility at ANL (USA)

:
<http://www.ne.anl.gov/ivem/>

In-situ TEM analysis of
irradiation

What is the role of
surfaces?

MD simulations of irradiation in Fe thin films

Enhanced production of vacancy loops, mostly $\langle 100 \rangle$

(a)

(b)

Irradiation of thin films at low energies (50-150keV) show the formation of large ($\sim 1\text{nm}$ radius) $\langle 100 \rangle$ vacancy loops. Mostly square loops as expected from energy considerations (Gilbert et al J.Phys:Cond.Mat. 2008)

Simulating damage in metals: surface vs. bulk irradiation

Bulk irradiation with 100keV PKA in Fe

- Bulk irradiation shows the formation of self-interstitial loops
- Some large vacancy loops are also observed.
- Note the sub-cascade formation

Statistical analysis of MD cascades

Cluster size differences between bulk and thin films

(a)

(b)

- Significant difference between the size of the clusters in bulk and thin films for the same energy
- Vacancy clusters much larger in thin films (> 100 defects)
- Self-interstitial clusters significantly larger in bulk irradiation

Damage produced by Ga ions in Fe at low energies (30keV)

Heavier ion (Ga) produces much larger vacancy clusters also of $\langle 100 \rangle$ type even at lower energies (30 keV), in agreement with experiments, M. L. Jenkins Nature (1976) & Z. Yao Phil. Mag. 2008

Modeling damage accumulation in Fe: OKMC

MMonCa – OKMC code
All individual defects can be resolved

OKMC code – MMonCa from IMDEA-Materials

I. Martin-Bragado, et. al. Computer Physics Communications (2013). /
www.materials.imdea.org/MMonCa

Small defects

Vacancies and Self-interstitials up to 4
Energetics from C. C. Fu, Nat. Mat. **4** (2005) 68

<100> vacancy loops

Energetics from
M R Gilbert et al, J. Phys.:
Condens. Matter **20** (2008)
345214

Defect-defect interaction in OKMC

$\langle 100 \rangle$ vs. $\langle 111 \rangle$ self-interstitial loops

100 keV Fe implantation in thin film

Model A

J. Marian [1]

D. Terentyev [2]

Formation of $\langle 100 \rangle$ by
interaction between $\langle 111 \rangle$ loops
of the same size

Model B

Marinica [3]

Growth of two independent loop
populations

5% $\langle 100 \rangle$ - 95% $\langle 111 \rangle$

[1] J. Marian et al. PRL 88, 25 (2002).

[2] H. Xu, R. Stoller, Y. Osetsky, D. Terentyev, PRL 110, 265503 (2013).

[3] M.-C. Marinica, F. Willaime, J.-P. Crocombette, PRL 108, 025501 (2012).

[4] N. Soneda, T. Diaz de la Rubia, Phil. Mag. A 81 (2001) 331

- $\langle 111 \rangle$ loop mobility from Soneda [4]
- $\langle 100 \rangle$ loops immobile

OKMC of 100keV Fe implantation in Fe and FeCr

Reference experiments

- Thickness ~ 50nm
- Pure Fe: mostly <100> loops **at low dose**
- FeCr: mostly <111> loops

Evaluate the defects obtained from the two models

Effect of different parameters:

- Mobility of <100> loops
- Model A: minimum <111> loops to <100>
- Model B: ratio of <100> vs. <111> loops
- Presence of traps (binding of 0.8 eV)
- Mobility of <111> loops

OKMC of 100keV Fe implantation in Fe and FeCr <100> Vacancy loops

<100> Vacancy loops immobile

Concentrations too high compared to experimental measurements

<100> Vacancy loops mobile

Considering a mobility of 0.5 eV
low concentration of <100> V loops

OKMC of 100keV Fe implantation in Fe and FeCr

Model A vs. Model B – no Carbon

Model A: $\langle 111 \rangle + \langle 111 \rangle \rightarrow \langle 100 \rangle$
Model B: 5% $\langle 100 \rangle$, 95% $\langle 111 \rangle$

Without Carbon all loops are of $\langle 100 \rangle$ type with both models

Concentrations are too high compared to experimental measurements

Dose dependence closer to experiments in the case of Model B

OKMC of 100keV Fe implantation in Fe and FeCr

Model B – effect of initial distribution of loops

Model B:

- 5.0% <100>, 95.0% <111>
- 0.1% <100>, 99.9% <111>

Distribution of only 0.1% of the small Interstitial clusters (< 4) as seeds for <100> loops gives loops concentrations in closer agreement to experimental values for (100) orientation

Rate of growth of loops with dose in agreement with experimental measurements

OKMC of 100keV Fe implantation in Fe and FeCr

Model B – effect of $\langle 111 \rangle$ mobility

Model B:

- $\langle 111 \rangle E_m \sim 0.1 \text{ eV}$
- $\langle 111 \rangle E_m = 1 \text{ eV}$

Two population of loops:

$\langle 111 \rangle$ and $\langle 100 \rangle$

Densities close to values for FeCr

More $\langle 111 \rangle$ than $\langle 100 \rangle$ loops

Problems with the saturation of loops at high doses:
loop-loop interaction capture radius
or long range interactions must be reviewed

OKMC model for concentrated alloys

1. The alloying element is not treated discretely but in terms of concentration
2. Jump rates of particles are not fixed: will depend on the location of the particle and the environment.

1 C_1	2 C_2	...
...	i C_i	$i+1$ C_{i+1}
	...	N C_N

L. MALERBA & M. J. CATURLA

Do all these mechanisms just add up? $1 + 1 = 2$?

500 kev cascade in Fe

E. Zarkadoula et al,
J. Phys. Condense. Matter
25 (2013) 125402

Simulating interfaces and complex systems

Fréchard et. al. JNM (2009)
TEM image of a martensitic steel
after He implantation

What we want to achieve:

Modeling concentrated
alloys, interfaces (grain
boundaries) and
precipitates (ODS)

Conclusions

- Primary damage plays a major role in the subsequent microstructure evolution
- Surface effects must be taken into account when comparing to in-situ TEM ion implantation:
 - Damage production could be affected by the presence of surfaces (low energy irradiation)
 - Damage evolution will be affected due to preferential trapping by surfaces or image forces.
- More complex and detail simulations: loop size and type.
- New methods emerging at all levels to study complex systems: alloys, electronic effects...

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