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Workshop

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**Multiscale Modelling of Microstructure Evolution under
Radiation Damage of Steels Based on Atomistic to
Mesoscale Methods**

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In order to simulate the microstructure evolution with the objective to predict it, multiscale modelling tools are developed (Fig. 1). For this purpose different simulation methods are used and developed in order to treat the different physical phenomena occurring at different time scales and length scales: *ab initio*, classical molecular dynamics, kinetic Monte Carlo, dislocation dynamics, phase field [1]. These simulations are very CPU demanding and take advantage of the development of High Performance Computing machines.

Fig. 1: Multiscale modelling scheme applied within the PERFORM-60 project to the pressure vessel and internal material microstructure.

The microstructure evolution under irradiation is obtained starting from the neutron spectrum to obtain the primary damage (displacement cascades), followed by the evolution of the point defects formed and their accumulation (Fig. 2).

Fig. 2: Microstructure modelling under irradiation.

The point defects created (vacancy and self interstitials) under irradiation often interact with the solute elements present in the materials. Solute elements can precipitate and/or segregate on point defect clusters (loops or voids) or extended defects (dislocations, grain boundaries). These modifications of the microstructure affect directly the mechanical properties of the materials. Thus, modelling should take into account the most important solute elements in the chemical composition of the industrial alloys.

For the pressure vessel steels (for which an important international effort is done in particular thanks to the PERFECT [2] and PERFORM-60 European projects) the evolution of the microstructure of dilute Fe alloys as complex as Fe-CuNiMnSiP-C under irradiation are modelled using a multiscale approach based on *ab initio*, molecular dynamics and kinetic Monte Carlo (KMC) simulations. In these atomic KMC simulations, both self interstitials and vacancies, isolated or in clusters, as well as carbon atoms are modelled [3]. The short term evolution of the microstructure is simulated. The medium to long term evolution of the microstructure is obtained by object KMC and cluster dynamics, considering a “grey” material. The interaction of some of these defects with dislocations are characterised by molecular dynamics in order to be used in mesoscopic dislocation dynamics. A similar approach is developed for austenitic materials modelled by a concentrated FeCrNi alloy (γ -Fe₇₀Cr₂₀Ni₁₀). The thermal ageing (without irradiation) of FeCr alloys will also be presented.

In the framework of the European projects dedicated to the pressure vessel steels and the austenitic steels, the multiscale modelling methods of the microstructure have been capitalised within two tools (RPV and INTERN) [4].

References

- [1] C. S. Becquart, C. Domain, Metallurgical and Materials Transactions A 42 (2011) 852.
- [2] Special Issue: PERFECT project. Journal of Nuclear Materials, 406 (2010).
- [3] C.S. Becquart, C. Domain, Phys. Status Solidi B 247 (2010) 9.
- [4] G. Adjanor et al., J. Nucl. Mater 406 (2010) 175.

PERFORM 60
FP7 Project

Multiscale Modelling of Microstructure Evolution under Radiation Damage of Steels Based on Atomistic to Mesoscale Methods

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Lifetime extension: Materials ageing prediction

- To improve quantitative predictions of ageing of irradiated structural materials in nuclear power plants in order to gain margins.
- Challenge: to predict the evolution of hundred of tons over more than 40 years based on physical phenomena occurring at the nanometer scale and picosecond times (10^{-12} s)
- Construction and improvement of multiscale modelling methods allowing to better take into account the material composition and radiation damage

Microstructure evolution of Fe alloys under irradiation

RPV Fe ferritic alloys

- microstructure modelling short & long term evolution
- plasticity

Austenitic alloys

- FeNiCr AKMC RIS modelling

[A. Volgin PhD
SAT@GPM Rouen]

1nm^3

$0 - \text{ps}$

$(10-30\text{nm})^3$

ns

ab initio

Molecular dynamics

Multi-scale
modelling

Finite elements

cm^3

Micro-macro

μm^3

Dislocation
dynamics

Mesoscopic

+ experimental validation

Barbu, CEA

Pareige, U. Rouen

PERFORM 60
FP7 Project

$s - h$

KMC
cohesive model
& parameterisation

$(30-100\text{nm})^3$

$h\text{-year}$

Simulation tools

Atomic Kinetic Monte Carlo of microstructure evolution

Objective:

Simulation formation of solute rich complexes (observed by TAP) under irradiation

Ab initio

Solute interactions (Cu, Ni, Mn, Si)
(interface energies,
mixing energies ...)

Experimental data and
Thermodynamical data

Parameterisation
cohesive model

AKMC

Solute diffusion by
- vacancy mechanisms
- interstitial mechanisms

$$E_{\text{mixing}} = -4\varepsilon_{(Fe-Fe)}^{(1)} - 3\varepsilon_{(Fe-Fe)}^{(2)} + 8\varepsilon_{(Fe-X)}^{(1)} + 6\varepsilon_{(Fe-X)}^{(2)} - 4\varepsilon_{(X-X)}^{(1)} - 3\varepsilon_{(X-X)}^{(2)}$$

$$E_{\text{formation}}(V^Z) = 8\varepsilon_{(V-Z)}^{(1)} + 6\varepsilon_{(V-Z)}^{(2)} - 4\varepsilon_{(Z-Z)}^{(1)} - 3\varepsilon_{(Z-Z)}^{(2)}$$

$$E_{\text{binding}(V-X)}^{(1)} = \varepsilon_{(Fe-V)}^{(1)} + \varepsilon_{(Fe-X)}^{(1)} - \varepsilon_{(Fe-Fe)}^{(1)} - \varepsilon_{(V-X)}^{(1)}$$

Experimental validation: TAP, SANS, SAXS, PA, TEP

Atomistic Kinetic Monte Carlo (AKMC)

Vincent et al. NIMB 255 (2007) 78
Vincent et al. JNM 382 (2008) 154

- ⊙ Treatment of multi-component systems on a rigid lattice
  - Substitutional elements
  - Interstitial elements

Code: LAKIMOCA

- ⊙ Diffusion by 1nn jumps
  - Via vacancies
  - Via interstitials

Jump Probability: $\Gamma_x = \nu_x \exp\left(-\frac{Ea}{kT}\right)$ $\nu_x = \text{attempt frequency}$

- ⊙ Residence Time Algorithm applied to all events
  - Vacancy and Interstitial jumps
  - Frenkel Pairs and Cascade flux for irradiation

Average time step: $\Delta t = \frac{1}{\sum_{j,k} \Gamma_{jk}}$

→ Environment dependant form of activation energy Ea

$$Ea = Ea(X_i) + \frac{Ef - Ei}{2}$$

AKMC irradiation simulation conditions

For electron irradiation: Frenkel Pair (FP) flux

For neutron irradiation: flux of

- 20 keV and 100 keV cascades debris obtained by Molecular Dynamics
(R. Stoller, J. Nucl. Mater. 307-311 (2002) 935)
- Frenkel Pairs

Typical simulation box:

$1.01 \times 10^{-17} \text{ cm}^3$ boxes

$8.65 \cdot 10^6$ atoms

Cohesive energy model (bcc)

$$Ea = Ea(X_i) + \frac{Ef - Ei}{2}$$

Vacancy:

$$E = \sum_j \mathcal{E}_{(Fe-Fe)}^{(i)} + \sum_k \mathcal{E}_{(V-V)}^{(i)} + \sum_l \mathcal{E}_{(Fe-V)}^{(i)} + \sum_m \mathcal{E}_{(Fe-X)}^{(i)} + \sum_n \mathcal{E}_{(V-X)}^{(i)} + \sum_p \mathcal{E}_{(X-Y)}^{(i)}$$

- RPV: 1nn and 2nn pair interactions
- FeCr: 2BM potential

SIA:

- solute - dumbbell

- dumbbell - dumbbell

● Solute atoms
● Fe atom

$$E_{dumb} = \sum_i \left(E_f + \sum_j E_l^{1nnComp}(dumb_i - X_j) + \sum_j E_l^{1nnTens}(X_j) + \sum_{i,j} E_l^{mixte}(X_j - X_k) + \sum E_l(dumb - dumb) \right)$$

FIA (C):

~ 100 ab initio data considered in the model

Cohesive model: ε_{X-Y} and ε_{V-X} determination

Binary alloys

- $E_{mélange} = -4\varepsilon_{(Fe-Fe)}^{(1)} - 3\varepsilon_{(Fe-Fe)}^{(2)} + 8\varepsilon_{(Fe-X)}^{(1)} + 6\varepsilon_{(Fe-X)}^{(2)} - 4\varepsilon_{(X-X)}^{(1)} - 3\varepsilon_{(X-X)}^{(2)}$
- $E_{interface(100)} = -2\varepsilon_{(Fe-Fe)}^{(1)} - \varepsilon_{(Fe-Fe)}^{(2)} + 4\varepsilon_{(Fe-X)}^{(1)} + 2\varepsilon_{(Fe-X)}^{(2)} - 2\varepsilon_{(X-X)}^{(1)} - \varepsilon_{(X-X)}^{(2)}$
- $E_{cohésion}(Z) = 4\varepsilon_{(Z-Z)}^{(1)} + 3\varepsilon_{(Z-Z)}^{(2)}$
- $E_{formation}(lac^Z) = 8\varepsilon_{(lac-Z)}^{(1)} + 6\varepsilon_{(lac-Z)}^{(2)} - 4\varepsilon_{(Z-Z)}^{(1)} - 3\varepsilon_{(Z-Z)}^{(2)}$
- $E_{liaison}^{(i)}(lac-lac) = 2\varepsilon_{(Fe-lac)}^{(i)} - \varepsilon_{(Fe-Fe)}^{(i)} - \varepsilon_{(lac-lac)}^{(i)}$
- $E_{liaison}^{(1)}(lac-X) = \varepsilon_{(Fe-lac)}^{(1)} + \varepsilon_{(Fe-X)}^{(1)} - \varepsilon_{(Fe-Fe)}^{(1)} - \varepsilon_{(lac-X)}^{(1)}$

$i = 1 \text{ or } 2$

$X, Y = \text{solute atoms}$

$Z = \text{Fe or solute atom}$

Ternary alloys...

$$E_{liaison}^{(i)}(X-Y) = \varepsilon_{(Fe-X)}^{(i)} + \varepsilon_{(Fe-Y)}^{(i)} - \varepsilon_{(Fe-Fe)}^{(i)} - \varepsilon_{(X-Y)}^{(i)}$$

Ab initio data

Parameters

Adjustment on thermal annealing experiment

Figure 1: Crystal structure of Fe₂O₃. (a) 3D view of the rhombohedral structure with Fe atoms (red) and O atoms (green). (b) 2D projection of the structure along the c-axis, showing the arrangement of Fe atoms (black circles) and O atoms (gray circles) in the ab plane. The lattice parameters are given as $a = b = 2.41$ Å and $c = 1.16$ Å. The angle between the c-axis and the a-axis is 111° . The Fe-Fe distance is 0.28 Å.

Fe-CuNiMnSi

686+19 Fe atoms
PAW GGA
300 eV - 1 kpoints

Neutron irradiation of FeCuNiMnSi alloys

Medium term evolution by atomic Kinetic Monte Carlo

Fe-0.2Cu-0.53Ni-1.26Mn-0.63Si (at.%) at 300°C

Flux: $6.5 \cdot 10^{-5} \text{ dpa.s}^{-1}$

Dose: $1.3 \cdot 10^{-3} \text{ dpa}$

- Cu ● Ni
- Si ● V
- Mn ○ SIA

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V-solute complex

SIA-solute complexes

Small solute clusters

Point defect clusters = germs for precipitation

[> 1 month on 1 CPU]

Fe – CuMnNiSiP (at.%) alloys

- The biggest solute clusters are associated with PD clusters
  - In agreement with induced segregation mechanism to account for solute clusters formation
- Clusters associated with interstitial clusters are enriched in Mn, and P/Ni
- Clusters associated with vacancy clusters are enriched in Si/Cu/Mn (mostly) and Ni
- I-Solute complexes > V-Solute complexes

Average composition (nb solute & vac & int / cluster)

Vacancy - solute

Interstitial - solute

Solute clusters

All Solute clusters
AKMC

Solute clusters
Atom Probe [Meslin et al.]

Simulation

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Experimental results
[R. Ngayam happy PhD]

Cohesive energy model (fcc)

$$E = \sum_j \mathcal{E}_{(Fe-Fe)}^{(i)} + \sum_k \mathcal{E}_{(V-V)}^{(i)} + \sum_l \mathcal{E}_{(Fe-V)}^{(i)} + \sum_m \mathcal{E}_{(Fe-X)}^{(i)} + \sum \mathcal{E}_{(V-X)}^{(i)} + \sum_p \mathcal{E}_{(X-Y)}^{(i)}$$

1nn (and 2nn) pair interactions
(no reliable FeNiCr EAM
potentials for thermodynamical
and defect properties)

$$E_{dumb} = \sum_i \left(E_f + \sum_j E_l^{1nnComp} (dumb_i - X_j) + \sum_j E_l^{1nnTens} (X_j) + \sum_{i,j} E_l^{mixte} (X_j - X_k) + \sum E_l (dumb - dumb) \right)$$

● Tensile atoms   ● SIA
● Compressive atoms

FeNiCr interaction parameters adjustment on DFT data ($\text{Fe}_{70}\text{Ni}_{10}\text{Cr}_{20}$)

Chemical interactions in the Bulk

Cohesive energies $\rightarrow$ X-X terms

$$E_c^{Fe} = 6\epsilon_{FeFe}$$

$$E_c^{Ni} = 6\epsilon_{NiNi}$$

$$E_c^{Cr} = 6\epsilon_{CrCr}$$

Binding energies in dilute γ -Fe $\rightarrow$ X-Y terms

$$E_b^{NiNi/Fe} = -E_{mix}^{FeNi}/12 = 2\epsilon_{FeNi} - \epsilon_{FeFe} - \epsilon_{NiNi}$$

$$E_b^{CrCr/Fe} = 2\epsilon_{FeCr} - \epsilon_{FeFe} - \epsilon_{CrCr}$$

$$E_b^{NiCr/Fe} = \epsilon_{FeCr} + \epsilon_{FeNi} - \epsilon_{NiCr} - \epsilon_{FeFe}$$

Vacancy-solute interactions

E_f in pure fcc Fe $\rightarrow \epsilon_{FeV}$

$$E_f^V = 12\epsilon_{FeV} - 6\epsilon_{FeFe}$$

$$\Delta E_f^V = -0.04 \times 1nn\_Ni + 0.05 \times 1nn\_Cr$$

Evolution of E_f in $\text{Fe}_{70}\text{Ni}_{10}\text{Cr}_{20} \rightarrow \epsilon_{VCr}$ and ϵ_{VNi}

$$\Delta E^{Fe \leftrightarrow Cr} = \epsilon_{VFe} + \epsilon_{FeCr} - (\epsilon_{FeFe} + \epsilon_{VCr})$$

$$\Delta E^{Fe \leftrightarrow Ni} = \epsilon_{VFe} + \epsilon_{FeNi} - (\epsilon_{FeFe} + \epsilon_{VNi})$$

V-V Binding energy in dilute γ -Fe $\rightarrow \epsilon_{VV}$

$$E_b^{VV \rightarrow Fe} = 2\epsilon_{FeV} - \epsilon_{FeFe} - \epsilon_{VV}$$

TNES & RIS profil

Electron irradiation
0.6 dpa
723 K

TNES

RIS

TNES results:
Cr enrichment
Ni depletion

RIS results:
Cr depletion
Ni enrichment

→ Coherent with experimental results

Long term microstructure modelling Object Kinetic Monte Carlo

Objects:

- vacancy
- self interstitial
- dilute solute (with vacancy interactions)
- sink (e.g. grain boundaries, ...)
- trap (e.g. impurities, ...)
- dislocation
- foreign interstitial atoms

He in austenitic alloys

C or N in ferritic or austenitic alloys

Long term simulation of the microstructure under irradiation of Fe by object kinetic Monte Carlo

Input required:

Mobility: diffusion coefficient

Local interaction rules: Interaction and binding energies

HFIR n irradiation

Long term simulation of the microstructure: Application example: flux effect study in bcc Fe

$7 \cdot 10^{-11}$ dpa/s

(param Set II)

DEFECT POPULATION at 0.1 dpa

$7 \cdot 10^{-5}$ dpa/s

343K

Multiscale modeling of plasticity: phenomenological scales

Physics modeling

Irrad. microstructure

10 nm

Mechanical properties

RVE mechanics

Dislocation-defect

10 μ m

Collective dislocation behavior

10 μ m

Atom-meso transition

Meso-continuum transition

Homogenization

Dislocation at the atomic scales

Screw core structure: compact
ab initio

EAM: Mendelev03

EAM: Ackland04

Critical stress for (110) screw dislocation
(temperature, strain rate)

Critical stress for (112) edge dislocation
(temperature, strain rate)

Screw motion by DK

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Irradiation strengthening in RPV

Shear stress (MPa)

[Queyreau et al.]

Integration: RPV & INTERN platforms

Dislocation dynamics cristalline law of Fe alloys (at low temperature)

DD velocity of scrow dislo

$$v_{screw}(\tau, T) = \frac{v_D b^2}{l_c^2} l_{sc}^s \exp\left(-\frac{\Delta G(\tau_{app})}{kT}\right)$$

[Naamane, Monnet, Devincré]

Orowan Law for plastic flow

$$\dot{\gamma}_s = b \rho_{sc}^s v_{sc}^s(\tau, T) + b \rho_{ed}^s v_{ed}^s(\tau, T)$$

$$\dot{\gamma}_s = 4 \rho_{sc}^s \frac{v_D b^3}{l_c^2} l_{sc}^s \exp\left(-\frac{\Delta G_o}{kT}\right) \sinh\left(\frac{\Delta G_o}{kT} \sqrt{\frac{\tau_{eff}^s}{\tau_o}}\right)$$

Dislocation density evolution law

$$\dot{\rho}_s = \frac{|\dot{\gamma}_s|}{b} \left(\frac{\sqrt{\sum a_{su} \rho_u}}{K} - g_c \rho_s \right)$$

[Monnet, Vincent, Mécanique & Industries 12, 193–198 (2011)]

Finite elements simulations: stress strain curves

[Kuramoto 1979]

[Monnet, Vincent, Mécanique & Industries 12, 193–198 (2011)]

Material Multiscale Modeling Challenge

Conclusions & perspectives

- ⊙ A multi-scale modelling approach is developed for more than 10 years (e.g. through internal & EURATOM European project **SIRENA**, **PERFECT** & **PERFORM60**, **GETMAT**): **RPV** & **INTERN** platform.
- ⊙ Improvement of the knowledge elementary properties allow to better predict material evolution.
- ⊙ Some important progress thanks to the use of HPC machines.
- ⊙ The **prediction** of the evolution of the **mechanical properties requires** to know the **plasticity** of the materials.
- ⊙ The **prediction** of the evolution of the **irradiated microstructure** requires as input physical parameters the **properties** of **each point defect clusters** (mobility and stability).
- ⊙ The properties of each point defect clusters (size, configurations, chemistry) need to be defined by ab initio calculations. Represent numerous configurations / mechanisms to be investigated.