MECHANISMS OF RADIATION DAMAGE REDUCTION IN EQUIATOMIC SOLID SOLUTION ALLOYS

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High-Entropy and Equiatomic multicomponent Alloys

- High-entropy (HEA) and Equiatomic MultiComponent (EAMC) alloys are metal mixtures with multiple elements at equal or roughly equal concentrations, homogeneously distributed, in a single simple crystal

Definitions:

- HEA: 5 or more elements
- EAMC: 2 or more elements

- Rapidly rising interest to them due to promising mechanical, corrosion-resistant and radiation hardness properties
Experiments by Yanwen Zhang *et al* (ORNL) show that damage in some FCC high-entropy alloys can be clearly lower than in the corresponding pure elements. Standard point of comparison: Ni, which is already quite radiation-hard.

Simulation details

- We simulated several different alloys, including Ni, NiFe, NiCo, and NiCoCr with 108000 and 500 000 atoms in a cell.

- Potentials:
  - Tests showed that the structures were stable, with averaged cohesive energies and reasonable lattice parameters.
  - Bonny et al. [Bonny, Castin & Terentyev, Model. Simul. Mater. Sci. Eng 21 (2013) 085004] was additionally used for Ni and NiFe.

- Thermostat at the borders.
- 3 different runs for each alloy.
Single cascades in HEA’s

- It is not a priori clear why damage should be lower in high-entropy alloys
  - some alloys, such as NiAl, are susceptible to amorphization under irradiation
- Single cascades in HEA’s do not really show a difference to pure elements
- Example: 5 keV cascade in model CoNiFeCr HEA:
  - Recombination as usual, very similar to pure Ni
  - Damage slightly higher than in Ni
- Conclusion:
  - Single cascade simulations cannot explain experiments
To try to understand the damage saturation effects in HEA’s, we ran > 1500 overlapping cascades in them.

Key observation: after about 0.05 dpa, almost all damage is in clusters.

After that the damage keeps evolving.

Example: FeNi

The clustered damage shows a similar damage reduction effect as the experiments.

The reduction in damage level correlates clearly with dislocation mobility.

- In the alloys, each atom has a local strain field, and this reduces dislocation mobility.
- Lower dislocation mobility keeps dislocations from growing, and the smaller dislocations can recombine easier during cascade overlap.

**Final damage level** vs. **Slope of dislocation mobility** => Clear correlation

[Levo, Granberg, Nordlund, Djurabekova, submitted for publication]
Analyses of dislocation structures

We have analyzed all the frames for dislocations with the ovito DXA analysis (constructing Burgers vectors to detect dislocations)
Ni has larger dislocation loops and much more SFT’s than NiCoCr.
Dislocation reactions affecting overall damage level

- The dislocations dominate the overall damage level
- Numerous dislocation reactions occur driven by the irradiation
- Example: Shockley partial stepwise becoming a Frank loop

[Levo, Granberg, Nordlund, Djurabekova, submitted for publication]
Stacking fault tetrahedra

Comparison of evolution of numbers of SFTs in the alloys with the dose

[E. Levo, F. Granberg, C. Fridlund, K. Nordlund, and F. Djurabekova, Radiation damage buildup and dislocation evolution in Ni and equiatomic multicomponent Ni-based alloys, Journal of Nuclear Materials 490, 323 (2017)]
Interpretation of experimental RBS spectra

- In the experiments, the RBS signal appears very high, about "1/2 randomly displaced atoms"
- 50% damage does not at all correspond to TEM, resistivity or MD results, which show <1% defective atom fraction
- Explanation just determined by us: dislocations give a very high RBS signal due to strain effects
- New code RBSADEC to simulate RBS/channeling from arbitrary atom coordinates shows
  - Although simulated RBS spectra were obtained by fitting the defect profiles to correspond to the experimental ones, the density of RDA is much higher than the measured disorder. While much smaller concentration of extended defects results in the same signal

Preparation of the sample for RBS simulations

- Simulation of 1 um cell to match the depth penetration of the cascades to match the experiment is rather infeasible for simulations.
- Instead the same sample irradiated to the different doses where merged together to obtain one large sample, which was used to simulate the RBS/C spectra. Each cell in the stack was selected at the dose to correspond the nuclear energy deposition profile for the given...
Direct comparison of damage structure with experiments

- Using the RBSADEC code we can compare our structures directly with experiments (with no fitting!)
- Agreement is very good considering defect migration is not included in MD simulations and we use a single ion energy

Rather high chemical potentials for Cu-Ni, Cu-Co and Cu-Fe (0.9, 0.85 and 0.7 eV, respectively) leads to miscibility of this alloy at rather high temperatures (800 K). Already at 400K, the Cu tends to segregate as the variance constrained semi-grand canonical MC-MD simulations show (carried out by L. Koch, TU Darmstadt)

Defect structure at different instances of high-dose irradiation

- Red spots show the unidentified defect mesh, which at closer look appeared to be a vacancy or small vacancy clusters.
We observed similar tendency of growing larger dislocation structures in Ni, compared to the alloy.
We analysed the Warren-Cowley SRO parameter for all pairs after the 0.57 dpa irradiation dose (left), and also the preferred sites of precipitation (right).

### Damage buildup in high-entropy alloys vs. pure metals

<table>
<thead>
<tr>
<th></th>
<th>Ni</th>
<th>HEA</th>
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<tbody>
<tr>
<td><strong>Low dose</strong></td>
<td>Point defects, small defect clusters</td>
<td>Point defects, small defect clusters</td>
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<tr>
<td></td>
<td>(dpa &lt; 0.01, \ rpa &lt; 0.3)</td>
<td>- A bit more than in Ni due to slower defect mobility =&gt; less I-V recombination</td>
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<tr>
<td><strong>Intermediate dose</strong></td>
<td>Defects grow to form small partial and perfect dislocation loops and SFT’s. Vacancies and their clusters remain</td>
<td>Defects grow to form small partial dislocations</td>
</tr>
<tr>
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<td>(dpa \sim 0.05, \ rpa \sim 1.5)</td>
<td>- Growth slower than in Ni due to lower dislocation mobility</td>
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<tr>
<td><strong>High dose</strong></td>
<td>Perfect dislocation loops absorb most other defects and grow large Vacancies, their clusters and SFT’s remain</td>
<td>Dislocations grow slowly, perfect loops rarely form</td>
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<td>(dpa &gt; 0.1, \ rpa &gt; 3)</td>
<td>- Since features are smaller, recombination by heat spikes more likely</td>
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<td>- More vacancy clusters than in Ni</td>
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Conclusions

- Dislocation mobility is reduced in FCC high-entropy alloys
- After about ~ 0.05 dpa, overall damage level is dominated by dislocation structures, and their reactions affect the development
- This reduces the radiation damage in high-entropy alloys compared to the corresponding pure elements
- Relaxation of the structure with respect to the chemical potentials by MC-MD method does not indicate strong phase separation except for Cu, whose atomic volume is the largest in the considered alloy
- All alloys reach the steady state of defect concentration and of chemical order under irradiation, irrespective of the initial structure