The response of solids to irradiation by massive particles

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Metallic materials in extremes

Hydrogen embrittlement, corrosion

High temperature microstructure evolution

Severe plastic deformation

Radiation response

High strain rate deformation
Nuclear fission: energetic particles generated inside the solid CANDU reactor fuel assembly

A single fuel bundle

Fuel (UO$_2$):
$^{235}\text{U} \rightarrow$ neutrons, fission products with ~200MeV of kinetic energy

Garter spring (Ni-base alloys X-750):

$^{58}\text{Ni} + n \rightarrow ^{59}\text{Ni} + \gamma$

$^{59}\text{Ni} + n \rightarrow ^{56}\text{Fe} + \alpha \rightarrow 4.8\text{MeV}$

$0.3\text{MeV}$
Magnetic confinement fusion: particles from the plasma

- Deuterium
- Tritium
- Helium (3.5 MeV)
- Neutron (14.1 MeV)
Space: particles accelerated by massive bodies, powerful fields

Jovian magnetosphere: electrons, protons, and ions at 10s of keV
Interaction of massive particles with solids

My talk will focus on the thermal spike phase, where point defects are created
Modeling radiation effects in solids

Collision cascades: SRIM

Thermal spikes: molecular dynamics

Defect diffusion:
- Rate theory
- Reaction-diffusion
- Phase field
- Kinetic Monte Carlo
- Accelerated MD
- ...

Modeling the fate of defects over long times is very challenging.

My talk will focus on this

5MeV Ni self-ion irradiation

1.5keV self-ion in Cu
A closer look at thermal spikes

Ballistic phase, $<1\text{ps}$

Impinging particle

Liquid core: diffusion, mixing, demixing

\[ E_K > E_T \approx 20 - 80eV \]

Thermal phase, $2-3\text{ps}$
A closer look at thermal spikes

Resolidification, ~5ps
Thermal spikes in $\text{Ni}_3\text{Al}$

- Lowest energy state is an $\text{L}_1^2$-ordered compound
- Rapid quenching yields a metastable FCC solid solution
- Antisite defect
- Al Atom
- Ni Atom

Disordered zones (dark) in TEM of ion irradiated $\text{Ni}_3\text{Al}$

S. Muller et al., Phil. Mag. A 75, 1625 (1997)
Thermal spikes shapes in Ni$_3$Al

S. A. Skirlo and M. J. Demkowicz, Scripta Mater. 67, 724 (2012)
Compact thermal spikes cool slower

\[ \tau_{C\text{-high}} \]

\[ \tau_{C\text{-low}} \]

\[ T_A: \text{ambient temperature} \]

Decay Time (ps)

Compactness

- 100 K
- 300 K
- 600 K
- 900 K
Defect formation depends on compactness

Long decay times favor Frenkel pair recombination

Long decay times favor liquid phase disordering
Metal multilayer nanocomposites: radiation resistant, but metastable

Nearly void free after irradiation

200keV He\(^+\), 450°C, 3 dpa

Spherodization after annealing

Anneal at 700°C, 1hr

Can thermal spikes initiate layer pinchoff?

W. Han et al., Adv. Mater. 25, 6975 (2013)
S. Zheng et al., APL 105, 111901 (2014)
Molecular dynamics of metal multilayers

Thermal Spike (~0.5 ps)
PKA energy: 100 keV
~300Å
~2 million atoms
Layer thickness: 10Å

Cu: Yellow
Nb: Blue

After thermal spike (~200 ps)
Liquid phase mixing within thermal spikes

L. Zhang and M. J. Demkowicz, APL 103, 061604 (2013)
Phase field modeling of final microstructure

\[ f(\ ) = 4D f^2 (1-f)^2 \]

- Cahn-Hilliard equation
- Local free energy density function:

Interfacial thickness is set to be \( \sim 2\text{Å} \)

We are interested in the final state of multilayers (pinched off or not?)

Extensive de-mixing in solid state
No pinch off above a layer thickness of 2nm

10 independent simulations were performed for each layer thickness.

- Over a typical thermal spike lifetime, the liquid-phase interdiffusion distance is ~1nm.

- Composites with layer thickness above 2nm remain metastable under irradiation.
Metallic glasses

Volume change upon melting

Thermodynamic melting temperature

Glass transition temperature, typically $T_g \approx 0.6T_m$

Free volume

Supercooled liquid range

Fast quenching

Liquid

Slow quenching

Crystal

Glass

$\alpha_L > \alpha_C$

$\alpha_g \approx \alpha_c$

Image credit: K. Flores (WUSTL)

~cm-scale BMG samples
474-million atom model of a-$\text{Cu}_{50}\text{Nb}_{50}$

R. E. Baumer and M. J. Demkowicz,
475 keV Nb PKA

150 nm
475 keV Nb PKA

Kinetic Energy

10 eV

0.5 eV

150 nm

$\text{t=0.065 ps}$

$\text{ts=125k}$
475 keV Nb PKA

$150 \text{ nm}$

$t=0.1 \text{ ps}$
$ts=180k$

Kinetic Energy

$10 \text{ eV}$
$0.5 \text{ eV}$

$475 \text{ keV Nb PKA}$
475 keV Nb PKA

10 eV
0.5 eV

Kinetic Energy

150 nm

$t=0.26 \text{ ps}
\text{ts}=345k$
475 keV Nb PKA

t=0.5 ps
t_s=419k

Kinetic Energy

10 eV

150 nm

0.5 eV
475 keV Nb PKA

150 nm
475 keV Nb PKA

t=5 ps
t_s=451k

10 eV

Kinetic Energy

0.5 eV

150 nm
475 keV Nb PKA

$t = 10$ ps
$ts = 479k$

150 nm

Kinetic Energy

10 eV
0.5 eV
475 keV Nb PKA

- **475 keV Nb PKA**
- **KE\textsubscript{max} > 1 keV**
- **0.5 < |\Delta \vec{r} \textsubscript{max}| < 1 nm**

150 nm
90% of PKA energy dissipated in binary collisions >1keV
Numerous thermal spikes
Thermal spikes are ~10nm wide
Voxel field analysis

- **Averages**: Potential energy, density, and stress fields

\[
p_{e} = \frac{1}{N} \sum_{i=1}^{N} p_{e_i} = \frac{1}{V} \sum_{i=1}^{N} m_{i} \quad k_{l} = \frac{1}{V} \left( V^{*} \right)_{i}
\]

- **Fitting**: Temperature field

\[
p(k_{e}) = 2 \left( \frac{k_{e}}{k_B T} \right)^{1/2} \left( \frac{1}{k_B T} \right)^{3/2} \exp \left( \frac{k_{e}}{k_B T} \right)
\]

- **Derived**: Strain field

\[
\sigma_{ij}^{\text{tot}} = \frac{1}{2} (F_{ki} F_{kj})_{ij}
\]

\[
\rho_{ij} = \sigma_{ij}^{\text{tot}} \left( S_{ijkl} + T_{ij} \right)
\]

- **Derived**: Diffusivity field

\[
D(t) = \frac{1}{6} \frac{<r^2>}{t}
\]
Zones where $T_g$ is exceeded

$T_{max} = 350$ K
$T_{max} > 1500$ K

150 nm
Thermal spike properties

\[ D(t) = \frac{1}{6} \frac{\partial r^2(t)}{\partial t} \]
Density and diffusivity within thermal spikes

Properties of voxels with $T_{\text{max}} > 1500$ K at a single time, $t = 5$ ps
Rapid quenching locks in free volume, excess energy

Properties of liquid voxels are determined by quench rate

![Graph showing the change in density and potential energy with quench rate](image)
Radiation swelling of metallic glasses

Irradiated \( \alpha \)-Fe\(_{40}\)Ni\(_{40}\)B\(_{20}\)


<table>
<thead>
<tr>
<th></th>
<th>Density [g cm(^{-3})]</th>
<th>Yield stress [GPa]</th>
<th>Yield strain [%]</th>
<th>Young’s Modulus [GPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Matrix QR=1×10(^{13}) K/s</td>
<td>8.199</td>
<td>2.38</td>
<td>0.0253</td>
<td>101.8</td>
</tr>
<tr>
<td>Quenched thermal spikes &lt;QR&gt;=6.5×10(^{13}) K/s</td>
<td>8.154 (~0.5%↓)</td>
<td>3.02</td>
<td>0.0361</td>
<td>88.6</td>
</tr>
</tbody>
</table>

*Similar reasoning may explain radiation-induced ductilization*
The high pressure liquid in a thermal spike acts as an inclusion with a transient misfit => emits an elastic pulse.
Pressure excursions in thermal spikes
Plasticity adjacent to thermal spikes

Stress pulse exceeds material yield stress adjacent to thermal spike, leading to plastic flow
Plasticity adjacent to all thermal spikes
Thermal spike plasticity is polarized

Consistent with a pressurized ellipsoidal inclusion
Prediction: a directed particle beam causes metallic glasses to deform plastically

Amorphous alloy sample

No applied stress!

Beam of neutrons or heavy ions with energy <1MeV

Predicted strain per fluence:

\[ A = 1.5 \times 10^{-15} \text{ cm}^2/\text{Nb} \]

This prediction still awaits experimental validation
Summary

• Thermal spikes play a major role in radiation response of solids
  • Their shape (compactness) affects defect formation
  • Liquid phase interdiffusion in thermal spikes limits the thermal stability of nanocomposite metals
  • High-rate quenching of thermal spikes reduces the density of amorphous metals
  • In amorphous metals, stress pulses emitted from thermal spikes cause anisotropic plastic deformation in the surrounding solid material

• Much is known about thermal spikes, yet much remains to be discovered

• We are beginning to use our understanding of thermal spikes to engineer materials for radiation resistance
Lead: Texas A&M University, director: M. J. Demkowicz (MSEN)
Collaborating institutions: UCSB, U. Michigan, U. Connecticut
Goals:
- Discover, understand, and predict the influence of microstructural heterogeneities—such as interfaces, inclusions, and porosity—on the high strain rate (>10⁴/s) mechanical response of additively manufactured, multiphase materials
- Train the next generation of leaders in stockpile stewardship through close collaboration with partners at NNSA labs
- Advisory committee with members from NNSA labs and academia
  - From LLNL: Mukul Kumar

Visionary leadership team
Developing new capabilities
Training the next generation of leaders