Breakthrough in Understanding Radiation Growth of Zirconium

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17th International Symposium on Zirconium
in the Nuclear Industry
3-7 February, 2013, Hyderabad, India
The phenomenon of radiation growth

- Expansion in $a$-directions
- Contraction in $c$-direction

Radiation growth of Zr-7%Pu. Horak, Rhude, J. Nucl. Mater. 3 (1961). Growth rate $\sim 10^{-2}$ dpa$^{-1}$ (1%/dpa)

MICROSTRUCTURE EVOLUTION

- Nucleation and growth of prismatic interstitial and vacancy loops at low doses
- Nucleation and growth of basal vacancy loops at higher doses
Dose dependence of radiation growth

Annealed Zr

- Initial high strain rate
- Strain saturation
- Breakaway

Carpenter et al., JNM 159 (1988).

Cold-worked Zr

- High strain rate
- No saturation and breakaway
- Occasional negative a-strain
- Some times coexistence of interstitial and vacancy prismatic loops

Most observations have never been explained
Status of the Problem

The first model was suggested in 1962 by Buckley, but

... reliable mechanistic models to predict the deformation of even a pure Zr single crystal are not known... We therefore still rely on a phenomenological approach.


... understanding of the basic creep mechanisms in anisotropic materials like zirconium alloys is still not strong enough to be truly predictive... Today, most models are empirical in nature...


The radiation growth model presented here explains and describes quantitatively all the observations.
Basic reasons for the lack of understanding

Main assumptions of earlier models

- Single vacancies and SIAs
- Preferential absorption of SIAs by edge dislocations is the main driving force

The models might work for irradiations with 1 MeV electrons

Experiment, molecular dynamics simulations and theory revealed that:

- The model assumptions are wrong in the case of neutron and heavy-ion irradiations
Primary damage created in cascades

- ~ 90% of defects are recombined in cascades (compared to NRT)
- ~ 50% of interstitials are in the form of clusters
- SIA clusters migrate one-dimensionally in close-packed directions

Fraction of defects produced by cascades in Cu irradiated with neutrons vs temperature

Fraction of clustered interstitials vs PKA energy

The cascade damage depends only weakly on crystal lattice and composition since $E_{\text{PKA}} \gg E_{\text{cohesive}}$
Production Bias Model (PBM, Singh et al.)


Further development

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Recent review

Radiation Damage Theory, In: Konings R.J.M. (ed.)
Comprehensive Nuclear Materials
V. 1, pp. 357-391, Amsterdam: Elsevier.

Framework of PBM

- Production of point defects and interstitial clusters
- 3-D migration of vacancies
- 3-D, 2-D or 1-D migration of single interstitials
- 1-D migration of the interstitial clusters
Striking observations explained in the framework of PBM

- Void lattice formation
- Grain-boundary / grain-size effects in swelling
- PKA-energy effect in swelling
- Maximum swelling rate of 1%/dpa
- Formation of rafts of interstitial loops
- Non-homogeneous spatial distribution of defects, etc.


50 times higher swelling at 10 times smaller defect production rate
Alignment of vacancy-type defects in HCP metals

Voids in Mg
Risbet, Levy, JNM 50 (1974).

Voids in Zr
Carlan et al., ASTM STP 1295 (1996).

Vacancy loops in Zr
Griffiths et al., JNM 225 (1995).

The phenomenon is similar to void lattice formation in cubic crystals and has the same origin.
Framework of the radiation growth model

- Microstructure consists of prismatic and basal dislocations with Burgers vectors \(<1\text{-}210\rangle\) and \(<0001\rangle\)
- Single vacancies and interstitials migrate 3-D, whereas interstitial clusters migrate 1-D in \(<1\text{-}210\rangle\) close-packed directions
- Interstitial clusters interact only with \(a\)-dislocations of the same Burgers vector
- Interaction of interstitial clusters with \(c\)-dislocations is neglected
- Dislocation bias for point defects is neglected

**Generation rates**

\[
G_v = G_{NRT} (1 - \varepsilon_r), \\
G_i = G_{NRT} (1 - \varepsilon_r)(1 - \varepsilon_i^g), \\
G_{cl} = G_{NRT} (1 - \varepsilon_r) \varepsilon_i^g / x,
\]

- \(\varepsilon_r\) is recombined fraction
- \(\varepsilon_i^g\) is fraction of clustered interstitials
- \(x\) is mean cluster size

**Balance equations**

\[
\frac{dC_v}{dt} = G_v - D_v C_v \sum_{k=1}^{4} \rho_k, \quad (i = a_1, a_2, a_3, c),
\]

\[
\frac{dC_i}{dt} = G_i - D_i C_i \sum_{k=1}^{4} \rho_k, \quad (G_i < G_v)
\]

\[
\frac{dC_{cl}^m}{dt} = \frac{1}{3} G_{cl} - D_{cl} C_{cl}^m k_m^2, \quad (m = a_1, a_2, a_3).
\]
Strain rate calculations

due to climb of prismatic dislocations

\[
\frac{d\varepsilon_v}{d\phi} = \sum_{k=1}^{3} \left\{ (D_i C_i - D_v C_v) \rho_k + \sum_{m} D_{cl} xC_{cl}^m k_{km}^2 \right\} \cos^2 (\bar{v}\bar{a}_k),
\]

\( k_{km}^2 \) partial sink strength of \( k \) dislocations for \( m \) SIA clusters

due to climb of basal dislocations

\[
\frac{d\varepsilon_c}{d\phi} = (D_i C_i - D_v C_v) \rho_c,
\]

Using balance equations, the strain rates are given by

\[
\frac{d\varepsilon_v}{d\phi} = \chi \sum_{k=1}^{3} \left\{ \frac{1}{3} - \frac{\rho_k}{\rho} \right\} \cos^2 (\bar{v}\bar{a}_k),
\]

\[
\frac{d\varepsilon_c}{d\phi} = -\chi \frac{\rho_c}{\rho},
\]

\( \chi = (1 - \varepsilon_r) \varepsilon_i^g \) is the fraction of clustered interstitials

\( \rho \) is the total dislocation density

\( \chi \approx 2 \times 10^{-2} \) (derived for FCC Cu)

Strain rates are determined by

- properties of cascades
- fractions of dislocations with different Burgers vectors
Strain rates in Cartesian coordinates

\[
\frac{d\varepsilon_x}{d\phi} = \chi \left( \frac{1}{2} - \frac{\rho_x}{\rho} \right), \\
\frac{d\varepsilon_y}{d\phi} = \chi \left( \frac{1}{2} - \frac{\rho_y}{\rho} \right), \\
\frac{d\varepsilon_z}{d\phi} = -\chi \frac{\rho_z}{\rho}.
\]

Model predictions

- c-strain is always negative
- a-strain may be either positive \((\rho_{x,y}/\rho < 1/2)\) or negative \((\rho_{x,y}/\rho > 1/2)\) depending on the distribution of a-dislocations
- For uniform distribution, \(\rho_x = \rho_y\), strains in all prismatic directions are positive and fully determined by the density of c-dislocation

Saturation and breakaway

In annealed Zr, dislocation density is low, thus

- Nucleation and growth of a-loops leads to a decrease of \(\rho_z/\rho\), hence to strain saturation
- Nucleation and growth of c-loops leads to an increase of \(\rho_z/\rho\), hence to breakaway growth
Estimates of strain rates

Typical situation

\[ \rho_x = \rho_y \]

\[ \rho_z = (0.1 \div 0.2) \rho_{x,y} \]

\[ \chi \approx 2 \times 10^{-2} \]

Maximum strain rate

\[ \frac{d\varepsilon_x}{d\phi} = -\chi \frac{\rho_z}{\rho}, \]

\[ \frac{d\varepsilon_y}{d\phi} = -\chi \frac{\rho_z}{2 \rho}. \]

\[ \frac{d\varepsilon_z}{d\phi} \approx 0. \]

Maximum rate observed is \( \approx 10^{-3} \text{dpa}^{-1} \)

Dose dependence of strain

\[
\frac{d\varepsilon_x}{d\phi} = \chi \left( \frac{1}{2} - \frac{\rho_x(\phi)}{\rho(\phi)} \right), \\
\frac{d\varepsilon_y}{d\phi} = \chi \left( \frac{1}{2} - \frac{\rho_y(\phi)}{\rho(\phi)} \right), \\
\frac{d\varepsilon_z}{d\phi} = -\chi \frac{\rho_z(\phi)}{\rho(\phi)}. 
\]

\[
\rho_{x,y,z}(\phi) = \rho_{x,y,z}(0) + 2\pi R_{x,y,z}(\phi) N_{x,y,z}(\phi),
\]

\(R_{x,y,z}(\phi), N_{x,y,z}(\phi)\) are the loop radii and densities.

\[
\frac{dR_{a_i}}{d\phi} = \frac{1}{b} \left\{ (D_i C_i - D_v C_v) + D_{cl} C_{cl} a_i x \frac{k^2}{\rho_{a_i}} \right\},
\]

\[
\frac{dR_c}{d\phi} = \frac{1}{b} \left\{ (D_v C_v - D_i C_i) \right\}.
\]

Loop nucleation scenario used

Best-fit value \(\chi \approx 2 \times 10^{-2}\) is the same as for Cu

Fitting to experiment

\[\phi_a, \phi_2 = \rho_a = 10^{17} \text{ m}^{-2}; \quad \rho_c = 0.6 \times 10^{17} \text{ m}^{-2}\]
Comparison of calculations with experiments

Annealed Zr

Effect of cold work

\[ \rho_a = \rho_b = \rho_{a_3} = 10^{12} \text{ m}^{-2} \]
\[ \rho_c = 0.6 \times 10^{12} \text{ m}^{-2} \]
Growth strain at high doses

Growth strain rate at high doses is determined by the ratio of densities of $c$- and $a$-dislocation loops.
Negative $\alpha$-strain

Calculated growth strain for non-uniform distribution of $\alpha$-dislocations

Co-existence of vacancy and interstitial prismatic loops and negative strain are due to anisotropy of $\alpha$-dislocation distribution over $\alpha$-directions

Zee at al. JNM 150 (1987).
## Summary

- The model of radiation growth is developed which accounts for the true nature of the primary damage produced in displacement cascades and diffusional properties of self-interstitial defects.
- The model explains all observations including strain saturation, breakaway growth and effect of cold work.
- Anisotropy of prismatic dislocations over different $a$ directions is shown to be crucial for strain behavior.
- The maximum strain rate in Zr-based materials of $10^{-3}$ dpa$^{-1}$ is predicted, in agreement with experiment.
- Negative a-strain and co-existence of vacancy and interstitial prismatic loops are explained for the first time.
- The model predicts growth strain to increase linearly with irradiation dose with the rate which can be calculated from microstructure at intermediate doses.