26. Irradiation Induced Mechanical Property Changes: Hardening and Embrittlement

- Effect on stress-strain curve
- Relationship to microstructure (Russell-Brown theory of obstacle hardening)
- Embrittlement; hardening, fatigue effects
- Reduction of creep rupture time
- Deformation localization
- Pressure vessel embrittlement
- Mechanical Properties (of UO2)
 - Elastic properties
 - Fracture and Flow properties
- Creep

Irradiation Hardening and Embrittlement Application to the Pressure Vessel

In light water reactors, (LWR), the principal pressure boundary is the reactor vessel. The vessel contains the reactor core and maintains the pressure necessary to keep the coolant liquid at high temperatures. In the case of accidents the pressure vessel helps contain the fission products and holds the water from the emergency core cooling system. As such, it is a critical safety and licensing issue that the pressure vessel maintain its integrity in face of the most severe combinations of *stressors* in the design basis.

Nuclear reactor pressure vessels are made of ferritic steel, with a stainless steel cladding. A PWR pressure vessel has a diameter of 4.25 m and is 23 cm thick. The most serious design accident for the pressure vessel involves a loss of coolant accident (LOCA) followed by a sudden recooling upon reinjection of coolant. This causes a Pressurized Thermal Shock (PTS). At the beginning of the reactor's life the pressure vessel material is ductile and able to withstand PTS.

The concern is that, as the pressure vessel is exposed to neutron irradiation, it becomes embrittled and less able to resist PTS. The failure mechanism is the unstable propagation of an existing crack or flaw, under the application of the loads attendant upon PTS. The effect of neutron irradiation is to reduce the size of the critical flaw size required for unstable propagation, or conversely, to reduce the stress required for unstable propagation of a crack of a given size. Utilities applying for plant-life extension have to show that the radiation-induced embrittlement has not been enough to allow the maximum existing flaw to propagate in a brittle manner.

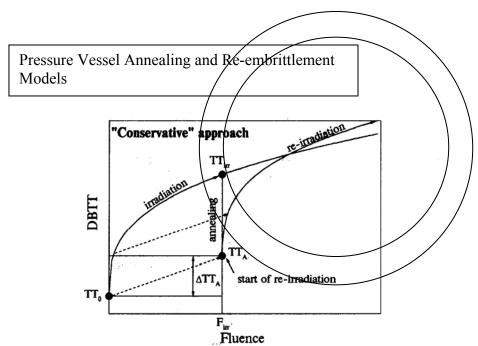


Figure 4 - The "Conservative" Model of Embrittlement under Re-Irradiation

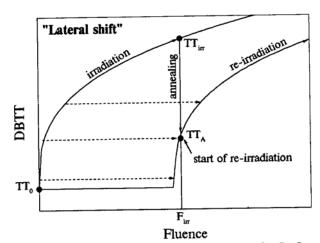


Figure 6 - The Lateral Shift Model of Embrittlement under Re-Irradiation

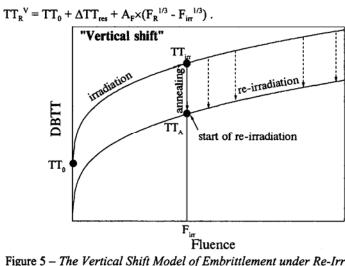
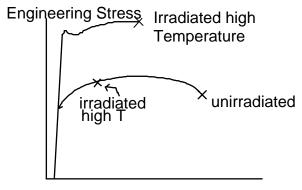


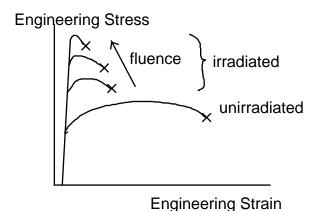
Figure 5 - The Vertical Shift Model of Embrittlement under Re-Irradiation

Y.Nikolaev, A.Nikolaeva, Effects of Radiation on Materials 19th Symposium, ASTM STP 1366, 2000, p.460

> As a result of irradiation, metals exhibit an increase in the both the yield stress σ_{ν} and the ultimate tensile strength σ_{UTS} . While the two values increase, they also become closer, which translates to a decrease in ductility. This manifests itself in the different responses that materials have to stress rupture tests before and after irradiation



Engineering Strain



Irradiation Hardening Mechanism

The principal irradiation hardening mechanism is the creation of a population of irradiation induced obstacles to dislocation motion. The yield stress is defined by the stress at which dislocations can bend around obstacles in their path. The change in yield stress in the presence of obstacles is inversely proportional to the inter-obstacle spacing ℓ :

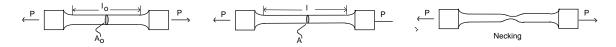
$$\Delta \sigma_{y} \alpha \frac{1}{\ell}$$

This behavior is assessed by different tensile tests representing different loading conditions and is the result of various features off microstructural evolution under irradiation

Uniaxial (Tensile) Test

When done rapidly, thre stress-strain curve is obtained. When done for a long time at high temperature, => Creep test

$$\sigma = \frac{P}{A}$$



Elastic Region

Plastic Region

 $\sigma = E \varepsilon$

Stress σ Engineering $\frac{P}{A_o}$ True $\frac{P}{A}$ strain ε $\frac{l-l_o}{l_o}$ $\int_{l_o}^{l} \frac{dl}{l} = \ln\left(\frac{l}{l_o}\right)$

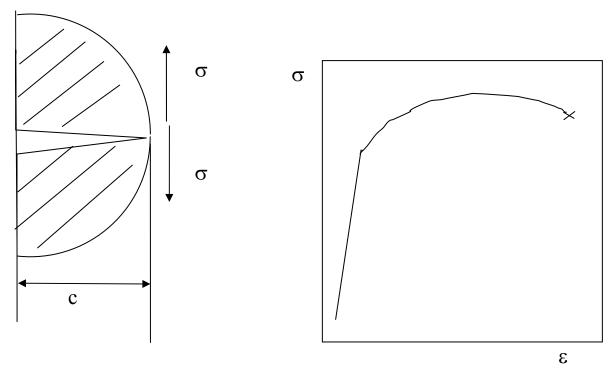
Departure from elastic behavior at $\boldsymbol{\sigma}_{_{\boldsymbol{y}}}$ Work hardening Region

 $\sigma = C \varepsilon^n$

n - work hardening coefficient (multiplication/tangling of dislocations)

Theory of Brittle Fracture - Linear Elastic Fracture Mechanics

Consider a unit length of material containing a crack depth c



The stress at the surface of the crack is zero and increases with distance to the plane of the crack. This means there is elastic energy stored in the solid, which is released when the crack propagates. There is an energy cost to creating that surface, given by

$$E_{surf} = (2\gamma_{surf} + \gamma_{plastic})c = \gamma * c$$

where γ [J/cm²] is an energy per unit area. γ_{surf} is the energy to create a new surface and $\gamma_{plastic}$ is the energy necessary to create a plastic zone at the tip of the crack.

The elastic strain energy is given by

$$E_{el} = \frac{E\varepsilon^2}{2} = \frac{\sigma^2}{2E}$$
 [J/m³]

The elastic strain energy decreases but there is a cost to creating a new surface. The balance between these two will determine if the crack will propagate or not. We assume all elastic energy is contained in a semi-circle radius c around the crack. The total elastic energy released by the crack is then:

$$\frac{E_{el}}{vol.} = \left(\frac{\pi c^2}{2}\right) \frac{\sigma^2}{2E}$$
 [J]

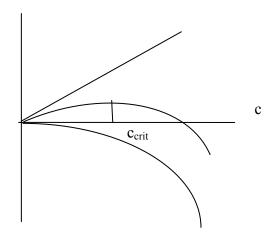
Light Water Reactor Materials, Draft 2006 © Donald Olander and Arthur Motta 10/5/2006 6

The presence of the crack relieves the strain energy in the shaded region. Thus a solid with initial elastic energy E_{el}^o would have

$$E_{el} = E_{el}^o - \frac{\pi \sigma^2 c^2}{4E}$$

and

$$E_{tot} = E_{surf} + E_{el}$$



To find the condition for crack propagation:

$$\frac{dE}{dc} = 0$$

$$E_{tot} = \gamma^* c - \frac{\pi \sigma^2 c^2}{4E}$$

$$\frac{dE_{tot}}{dc} = \gamma^* - 2\frac{\pi\sigma^2c}{4E} = 0$$

$$\sigma = \sqrt{\frac{2E\gamma^*}{\pi c}} \quad \Longrightarrow \quad (\sigma\sqrt{c})_{crit} = \sqrt{\frac{2E\gamma^*}{\pi}}$$

The RHS of this equation contains only material properties=> fracture toughness

The combination ($\sigma\sqrt{c}$) as occurs in the stress intensity factor

$$K_I = Y \sigma \sqrt{c}$$

where Y is a geometrical factor depending on the type of crack (between 1 and 2). Thus there is a critical stress intensity factor K_{IC} at which fast fracture occurs. The condition is

Condition for Crack Instability: $K_I = K_{IC}$

The design is based on this principle (i.e. Linear Elastic Fracture Mechanics LEFM). K_{IC} is a material property, called the *fracture toughness*. In an inert environment with $K_{I} < K_{IC}$, cracks are indefinitely stable. In an aggressive environment failure can occur by slow crack growth, i.e. if

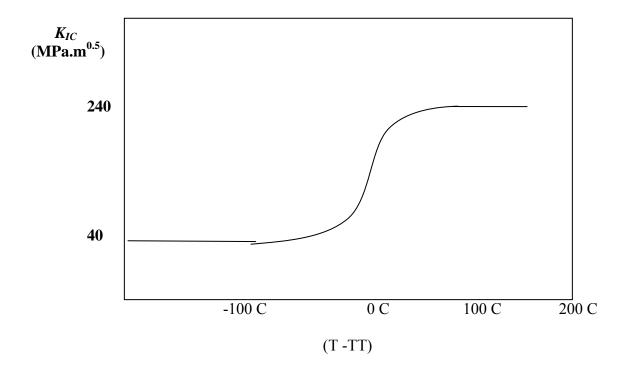
Condition for Slow Crack Growth:

$$K_{IC} > K_I > K_{ISCC}$$

There is an analogous process in ductile failure: when the applied stress is higher than the yield stress the material fails, but when the yield stress is higher than the applied stress, the material can still fail by slow deformation, or *creep*.

 K_{IC} (critical stress intensity or fracture toughness): measured in laboratory with carefully prepared cracks

Temperature Effect on Fracture Toughness

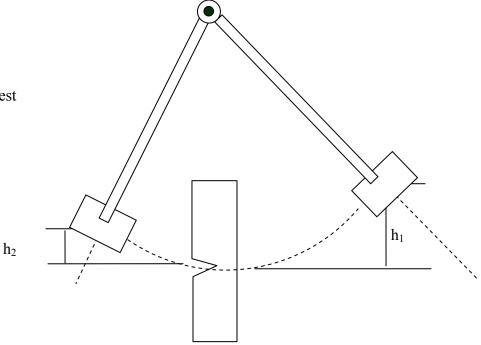


As the temperature decreases below the ductile to brittle transition the fracture toughness decreases. This behavior can be empirically described by an equation of the form:

$$K_{IC} = A \exp(-B[T - TT]) + C$$

Notch Toughness

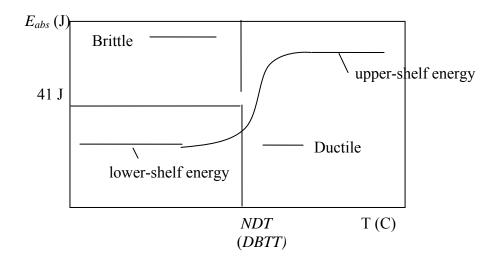
Charpy V-notch impact test



The energy absorbed in fracturing is given by

$$E_{abs} = g(h_1 - h_2)$$

The energy absorbed is a measure of fracture toughness, but <u>has no mechanistic relationship</u> to K_{IC} or other material properties.

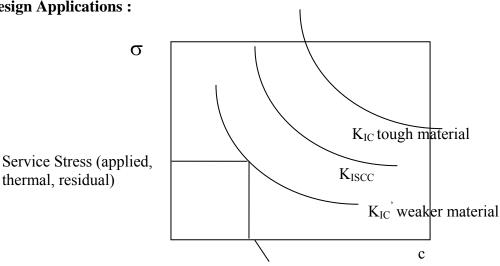


The advantage of the Charpy test is that it is simple and small and correlates with the fracture toughness. Empirical correlations have been developed to relate the yield stress with K_{IC} , one example is given below:

$$\left(\frac{K_{IC}}{\sigma_{y}}\right)^{2} = \frac{5}{\sigma_{y}} \left(E_{abs} - \frac{\sigma_{y}}{20}\right)$$

This is an empirical correlation so each material has its own.

Design Applications:



Maximum allowable flaw size for steel with K_{IC}

Dynamic Fracture Toughness

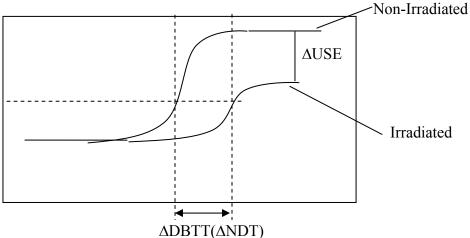
Under dynamical conditions (high strain rate) we use a different measure of resistance to fracture, K_{ID}

$$K_{ID} < K_{IC}$$

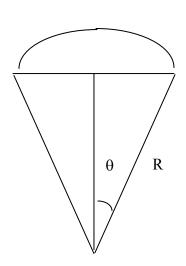
Once moving, the crack will arrest at K_{Ia}

Irradiation Hardening

Neutron Irradiation increases the σ_y (yield strength). Hard materials tend to be more brittle.



The increase in yield stress is related to the stress necessary to have a Frank-Read dislocation source operating, That stress is the stress needed to have the dislocation bow between pinning points, limit is



line tension
$$\frac{E}{L} \cong G b^2$$

Force per unit length due to stress $\frac{F}{L} = \tau b$

Restoring force due to line tension = $\frac{G b^2}{R}$

 τ is maximum for R minimum

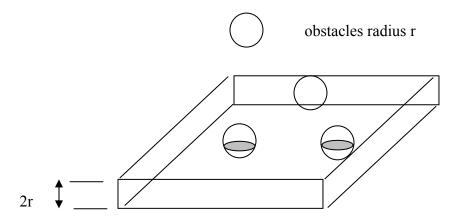
$$R_{\min} = \frac{L}{2}$$

$$\tau = \frac{Gb}{R}$$

$$au_{FR} = \frac{2Gb}{L}$$

This equation is valid also for the obstacles formed during irradiation, because the dislocation has to either bow around them or cut through them (normally less favorable

energetically). Each barrier will then have a given density, which can be calculated as follows:



Obstacle density is N (cm-3). Number of obstacles in volume above is 2rN, which is also the number of intersections per unit area on the slip plane. If we now equate the inverse square of the average obstacle spacing with the density of intersections on the plane we get

$$L = \frac{1}{\sqrt{2rN}}$$

and thus the increase in obstacle density causes a corresponding increase in yield stress given by

$$\left(\Delta\sigma_{y}\right)_{k} = \frac{2Gb}{L_{k}}$$

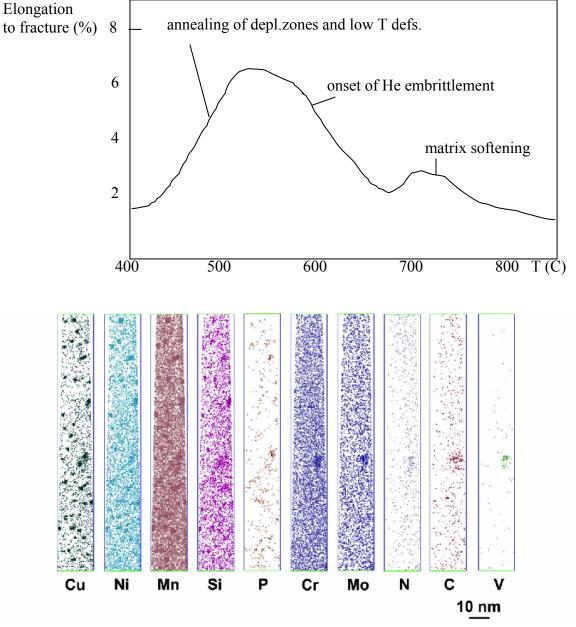
where k stands for all possible obstacles to dislocation motion formed under irradiation (dislocation loops, precipitates, depleted zones, etc)

Mechanisms of Hardening by Irradiation

$$\sigma_{y}(irrad) = \sigma_{y}(non - irrad) + \Delta\sigma_{y}$$

$$\Delta\sigma_{y} = (\Delta\sigma_{y})_{voids} + (\Delta\sigma_{y})_{def.clusters} + (\Delta\sigma_{y})_{ppts} + (\Delta\sigma_{y})_{loops}$$

In addition, irradiation creates helium by (n,α) reactions. Helium weakens the grain boundaries and reduces the elongation at fracture. This is embrittlement without hardening.



Atom maps showing the solute distribution in neutron irradiated KS-01 weld. A high number density of Cu-, Mn-Ni-, Si-and

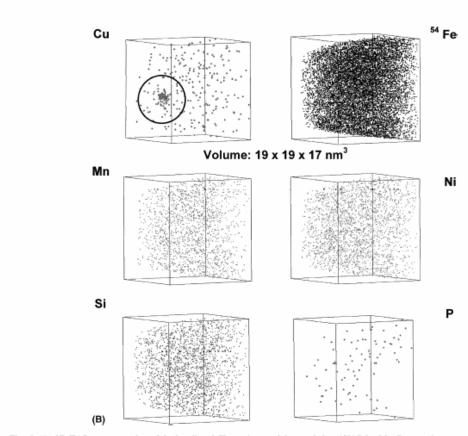


Fig. 8. (A) 3D TAP reconstruction of the irradiated Chooz A material annealed at 450°C for 2 h. Copper clusters are observed with no associated Ni, Mn, and Si. (B) 3D TAP reconstruction of the irradiated Chooz A material annealed at 450°C for 100 h. Copper clusters observed in the 2 h annealed materials are growing as pure copper particles.

Auger et al. Journal of Nuclear Materials 280 (2000) 331□}344

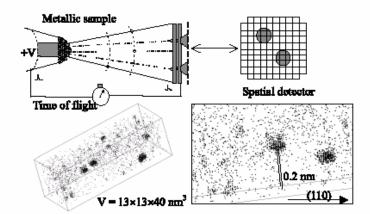


Fig. 1. Schematic representation of the tomographic atom-probe. On top left, the conventional system depicting field ion evaporation and the time of flight measurements. On top right a presentation of the multi-anode position sensitive detector. Bottom left, 3D reconstructed volume a Fe-1.5 wt% Cu alloy irradiated with swift krypton ions [7]. Only copper atoms are shown. Particles are observed. Bottom right, zoom in the (1 1 0) crystallographic direction of the left volume. Atomic planes in the copper particles are easily observed.

Pareige et al. NIM B, 178 2001 233