MATERIAIS DE
APLICAÇÃO NA
ENGENHARIA
NUCLEAR

Materiais dos
Reatores
Nucleares

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e Raquel Lobo

Ipen

What Really IS a DPA?

$$\frac{DPA}{sec} = \int_0^{E_{max}} \Phi(E_i) * \sigma_D(E_i) dE_i$$

$\Phi(E_i)$ – fluxo de partículas com energia E_i

$\sigma_D(E_i)$ - Seção transversal de deslocamento

$$\sigma_D(E_i) = \int_{T_{min}}^{T_{max}} \sigma(E_i, T) \nu(T) dT$$

What Really IS a DPA?

$$\frac{DPA}{sec} = \int_{0}^{E_{max}} \Phi(E_i) * \sigma_D(E_i) dE_i$$

σ $(E_i\ ,T)$ – probabilidade de que um átomo deslocado por uma partícula com energia E_i saia com energia de recuo T (seção transversal de transferência de energia diferencial)

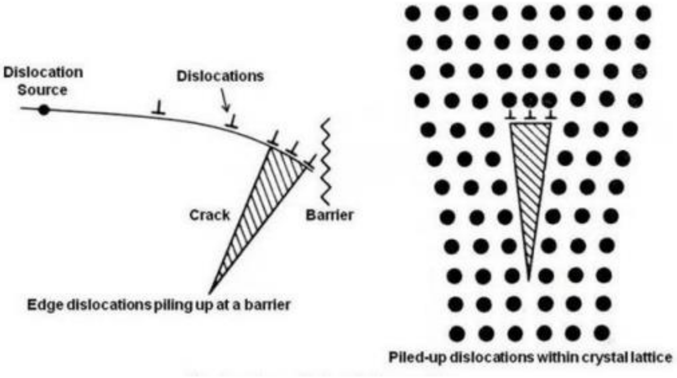
ν(T) – número de deslocamentos atômicos de um átomo com energia T

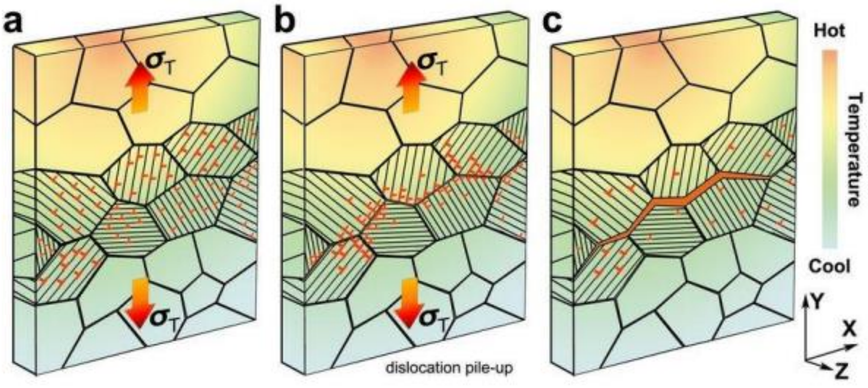
$$\sigma_D(E_i) = \int_{T_{min}}^{T_{max}} \sigma(E_i, T) \nu(T) dT$$

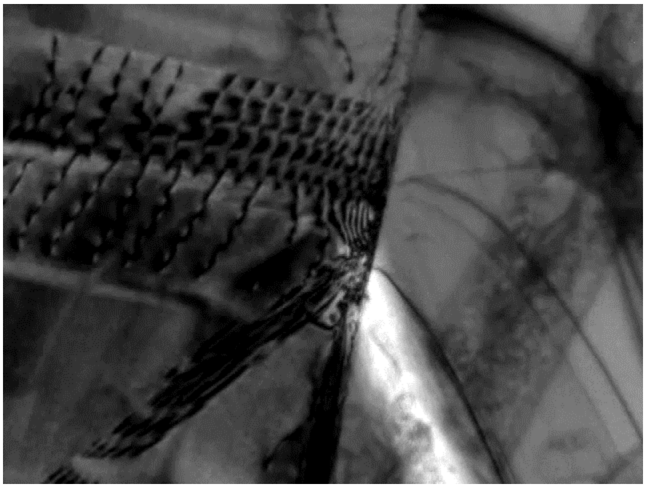
Noções Básicas de Danos da Radiação nos Materiais

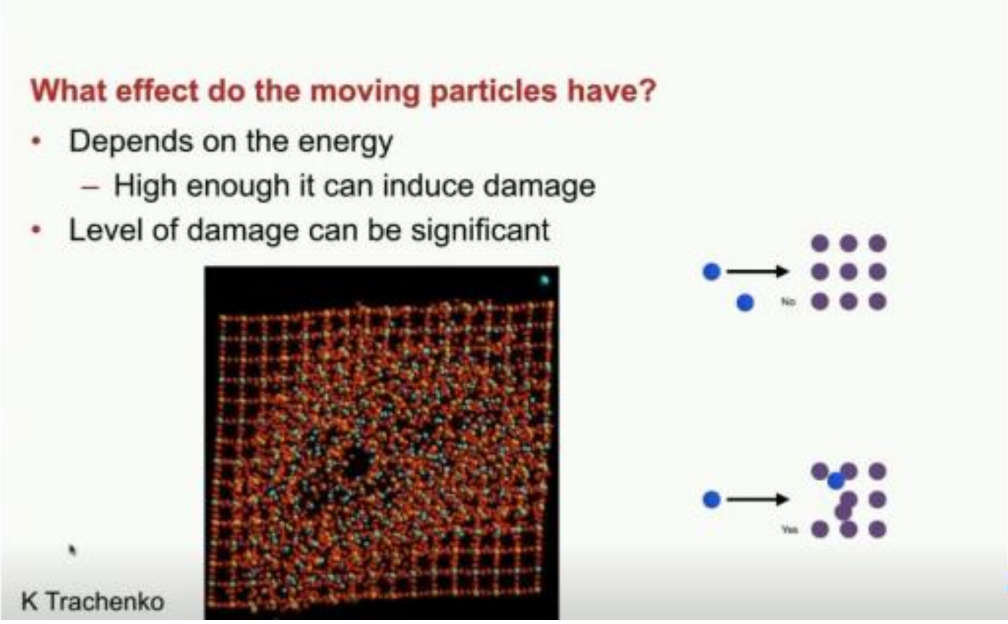
- Efeitos Básicos da Radiação Neutrônica
- Cascata de Deslocamento
- Causas e Consequências desses Danos
- Fabricação do Combustível Nuclear
- Interação Combustível – Revestimento
- Fatores Limitantes da Vida do Combustível

Mecanismo de Formação de Trinca









What effect do the moving particles have?

- Depends on the energy
  - High enough it can induce damage
- Level of damage can be significant

K Trachenko

Escalas de Tempo e de Energia para os Danos da Radiação devido aos Deslocamentos de Átomos

Tempo	Energia
Cascade Creation 10^{-13} s	**Neutron or Proton** 10^{5} - 10^{9} eV
Unstable Matrix 10^{-11} s	**Primary Knock-on Atom** 10^{4} - 10^{5} eV
Interstitial Diffusion 10^{-6} s	**Displaced Secondary** 10^{2} - 10^{3} eV
Vacancy Diffusion 10^{0} s	**Unstable Matrix** 10^{0} eV
Microstructural Evolution 10^{6} s	**Thermal Diffusion** kT

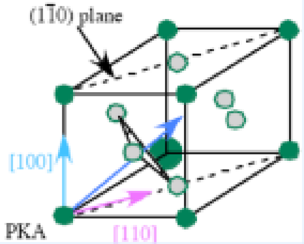
Displacement Energy

Atoms arranged in crystal structures,
thus FP displacement energy depends on PKA direction:
In UO_2:

Threshold displacement energies in UO_2 calculated using the sudden approximation method within the Mott-Littleton approach

[uvw]	[100]	[110]	[130]	[150]	[214]	[313]	[141]	[232]
E_d[O] (eV)	–	16	28	–	21	–	22	18
E_d[U] (eV)	52 (35)	85 (51)	46	44	79	43	55	74

Meis and Chartier, 2005



On average:

- E_d[O] ~ 20 eV
- E_d[U] ~ 50 eV
- For most metals E_d ~ 25-50 eV

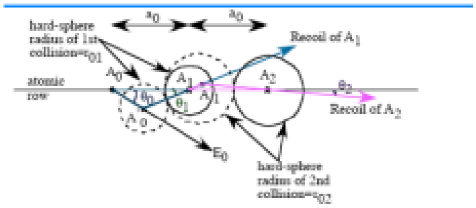
Efeitos na Rede

Átomos na maioria dos sólidos estão localizados em uma estrutura de rede regular.
Isso afeta a capacidade do PKA em causar deslocamentos atômicos por
Focalização e tunelamento

Focalização: Transferência de energia e átomos ao longo de uma fileira atômica em colisões quase frontais

Tunelamento: movimento de PKA ao longo de longas distâncias em direção aberta na rede

- ⇨ Eles afetam o número e a configuração de cascatas de colisão nos sólidos
- ⇨ Energia transferida por deslocamentos pode estar bem abaixo de E_d
- ⇨ Átomos podem viajar longas distâncias



Focalização (Focusing)

O primeiro átomo A_0 é permanentemente deslocado de sua posição original da rede e cai na posição ocupada por A_1
Cada colisão ao longo da linha atômica torna-se mias frontal que a colisão anterior, por isso o nome de focalização
O esquema acima mostra uma linha de átomos {100} em uma rede cúbica simples, com posições iniciais A_0 , A_1 , A_2 , ...
O primeiro átomo A_0 recebe energia E_0 , em uma direção Θ_0; move de A_0 para A_0, onde atinge o átomo A_1
O átomo A_1 se desloca de sua posição original com energia menor E_1 e, se ocorrer a focalização, em um ângulo menor Θ_1
O processo continua com $\Theta_i > \Theta_{i+1}$ até que a energia E_{i+1} seja muito pequena para deslocar (recolocar) os átomos atingidos por ele

Focalização

Ângulo crítico para focalização

Para cada energia de recuo E_0, há um ângulo crítico correspondente Θ_{0f}
Se $\Theta_0 < \Theta_{0f}$ - focalização ocorre
Se $\Theta_0 > \Theta_{0f}$ - focalização não ocorre
Note que Θ_{of} ↑ quando E_0 ↓

Energia crítica para focalização

Quando a energia de recuo se torna grande o suficiente para que $\Theta_{0f} = 0$, a energia máxima para a focalização ocorrer, denotada por E_f, é obtida

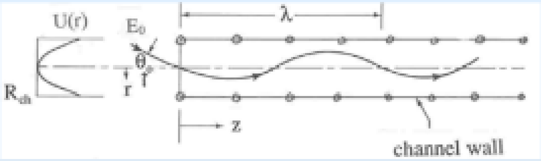
Canalização

Base experimental: quando íons são disparados em uma folha de monocristal que é mais espessa que a faixa de íons, a penetração dos íons é observada para direções cristalográficas a linha de átomos em empacotamento fechado da rede. O mecanismo é o movimento do íon ao longo de canais limitados pelas linhas atômicas de empacotamento fechado.

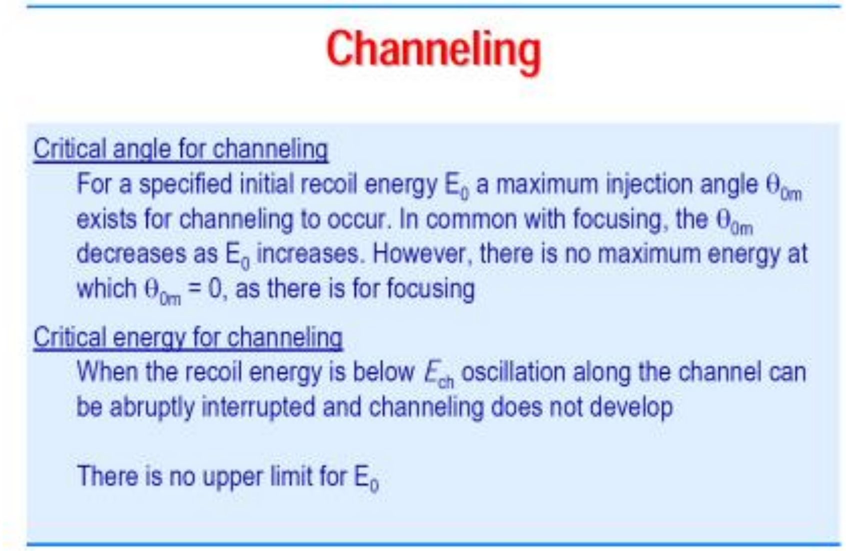
O efeito é importante em danos da radiação porque a canalização do íon pode ser um átomo de recuo gerado em uma cascata de colisão.

Canalização

O movimento dos átomos se movendo é similar ao de uma bola rolando para cima em um barranco inclinado: ele oscila na direção lateral e desacelera vagarosamente na direção axial.



Lateralmente a trajetória pode ser aproximada ao movimento harmônico simples com potencial canal parabólico.



Channeling

Critical angle for channeling

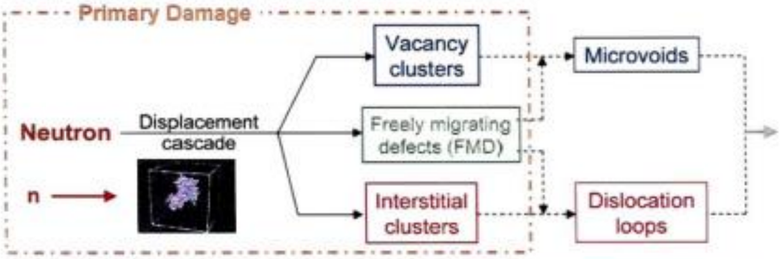
For a specified initial recoil energy E_0 a maximum injection angle θ_{0m} exists for channeling to occur. In common with focusing, the θ_{0m} decreases as E_0 increases. However, there is no maximum energy at which $\theta_{0m} = 0$, as there is for focusing

Critical energy for channeling

When the recoil energy is below E_{ch} oscillation along the channel can be abruptly interrupted and channeling does not develop

There is no upper limit for E_0

Efeitos da irradiação nos materiais



Primary Damage

Neutron

n

Displacement cascade

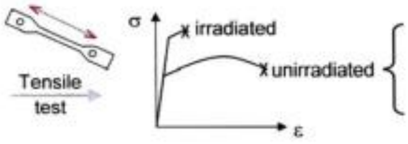
Vacancy clusters

Freely migrating defects (FMD)

Interstitial clusters

Microvoids

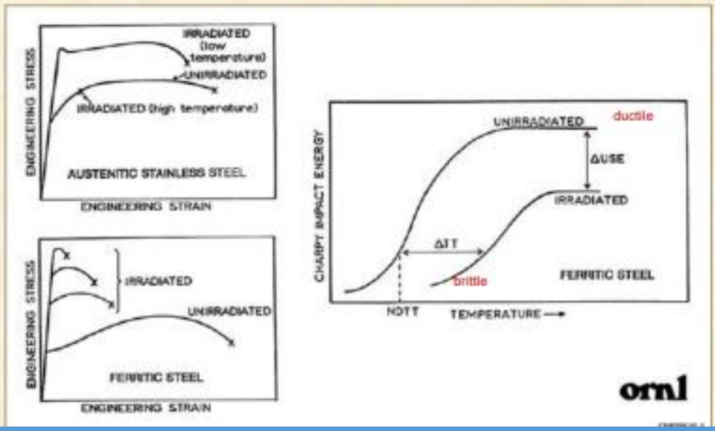
Dislocation loops



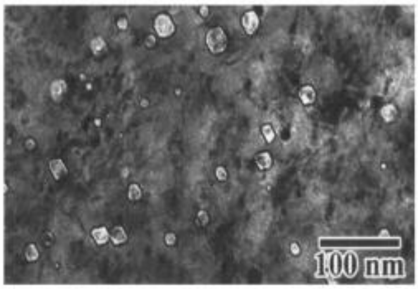
Mudanças nas propriedades mecânicas

"Endurecimento e Fragilização devido a Radiação"

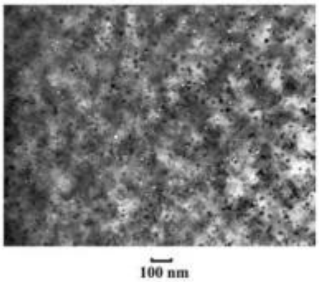
A irradiação neutrônica dos aços aumenta a *resistência mecânica* e diminui a *ductilidade* e a *tenacidade* – os aços ferríticos são mais susceptíveis que os aços inoxidáveis austeníticos



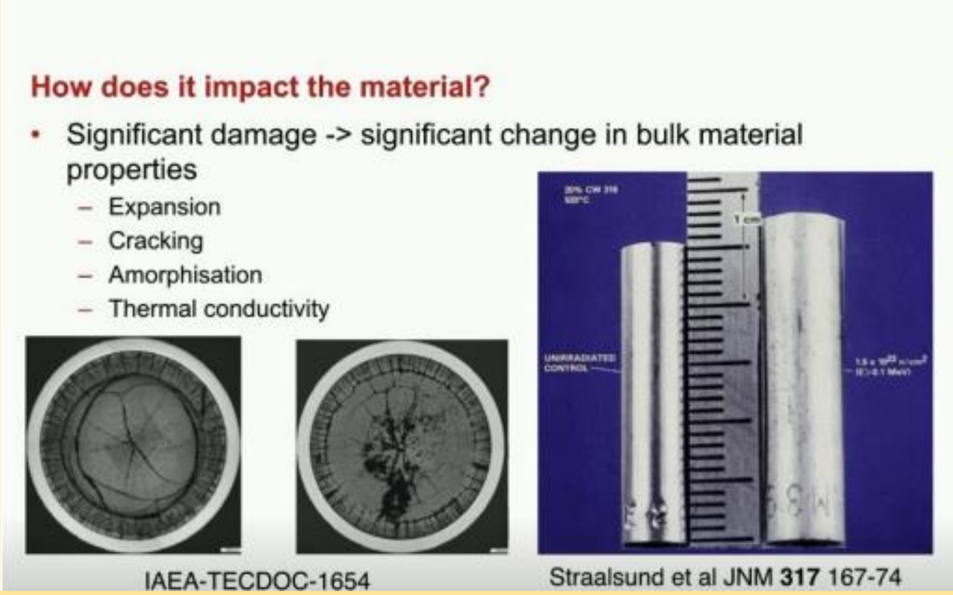
Dano da Radiação:
Acumulação de Defeitos



Void creation



Carbide precipitation



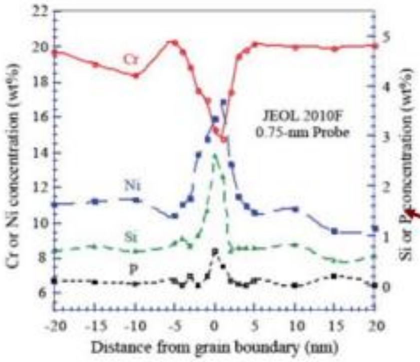
How does it impact the material?

- Significant damage -> significant change in bulk material properties
  - Expansion
  - Cracking
  - Amorphisation
  - Thermal conductivity

IAEA-TECDOC-1654

Straalsund et al JNM **317** 167-74

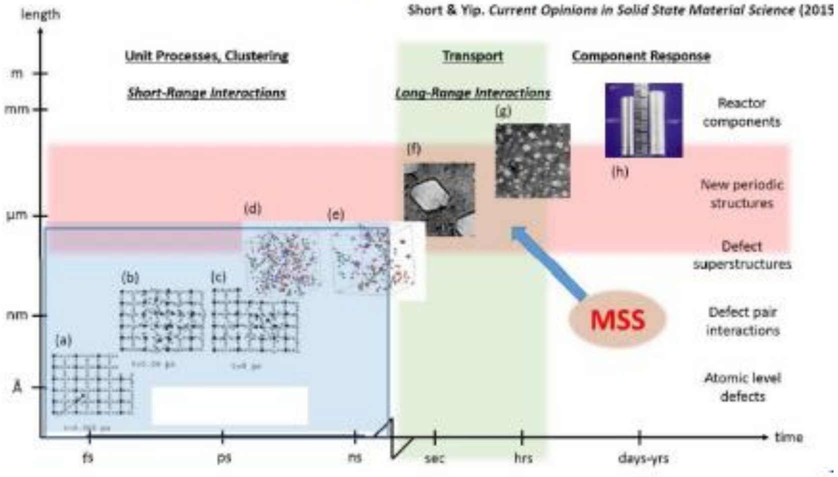
Dano da Radiação



- Radiation accelerates damage mechanisms
  - Stabilizes voids
  - Induces creep
  - Radiation induced segregation (RIS)
    - Grain sensitization
- Damage types change with temperature

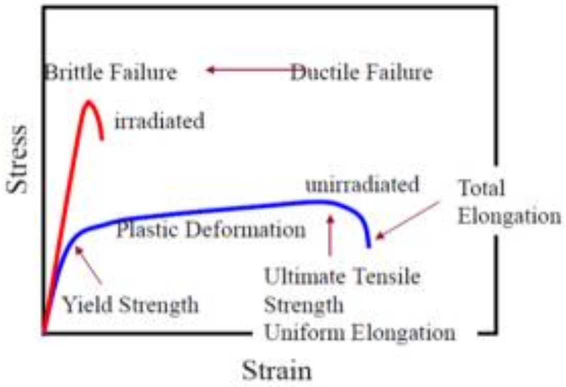
Radiation-induced segregation in 300-series stainless steel
Source: G. S. Was et al., J. Nucl. Mater., 411(1–3), pp. 41-50, 2011.

Pavimentando o caminho até os Efeitos da Radiação



Short & Yip. *Current Opinions in Solid State Material Science* (2015

Dano da Radiação:
DPA e Propriedades



Adapted, in part, from T. Allen in: "Materials Reliability and Degradation Management Issues In Nuclear Power Plants, May 4-7, Materials Aging Institute, France

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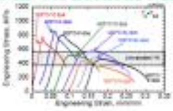
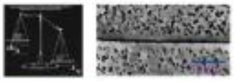
Radiation Damage can Produce Large Changes in Structural Materials

- **Radiation hardening and embrittlement ($<0.4\ T_M$, >0.1 dpa)**

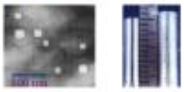


- **Phase instabilities from radiation-induced precipitation ($0.3\text{-}0.6\ T_M$, >10 dpa)**



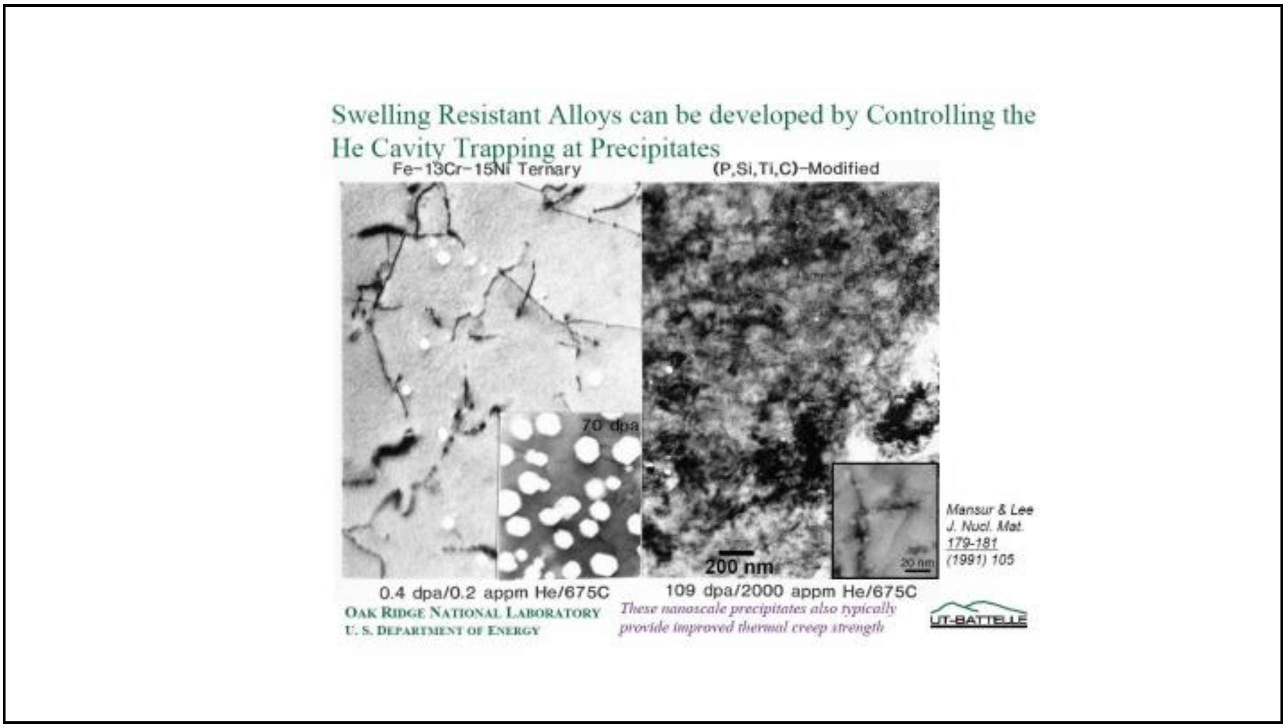
- **Irradiation creep ($<0.45\ T_M$, >10 dpa)**

- **Volumetric swelling from void formation ($0.3\text{-}0.6\ T_M$, >10 dpa)**



- **High temperature He embrittlement ($>0.5\ T_M$, >10 dpa)**

after S.J. Zinkle, Phys. Plasmas 12 *(2005) 058101*



Swelling Resistant Alloys can be developed by Controlling the He Cavity Trapping at Precipitates

0.4 dpa/0.2 appm He/675C

109 dpa/2000 appm He/675C

Mansur & Lee
J. Nucl. Mat.
179-181
(1991) 105

These nanoscale precipitates also typically provide improved thermal creep strength

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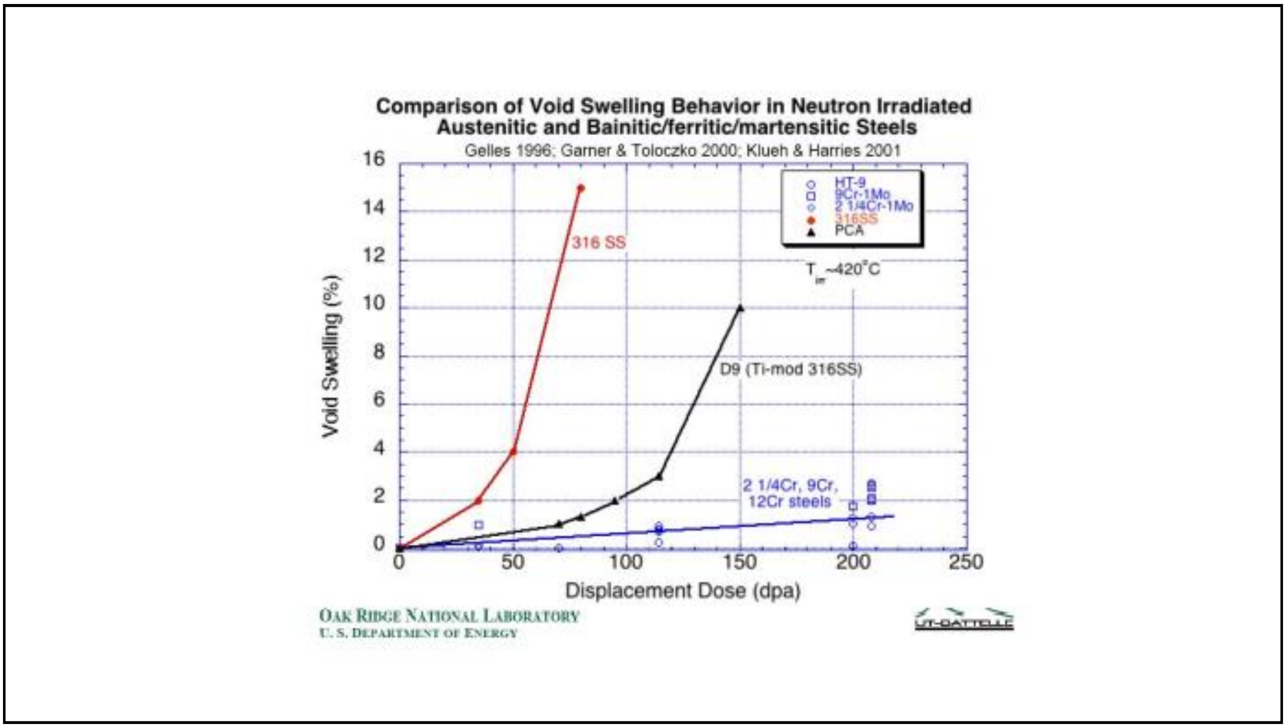
UT-BATTELLE



Comparison of Void Swelling Behavior in Neutron Irradiated Austenitic and Bainitic/ferritic/martensitic Steels

Gelles 1996; Garner & Toloczko 2000; Klueh & Harries 2001

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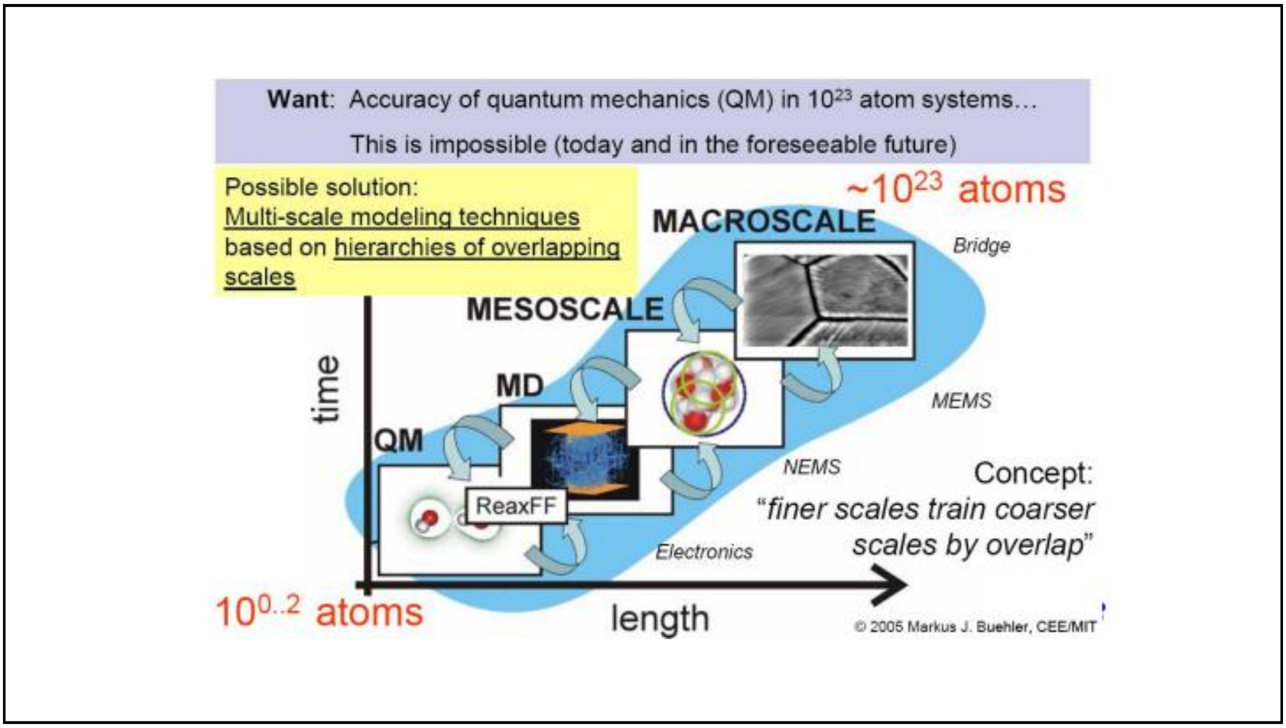


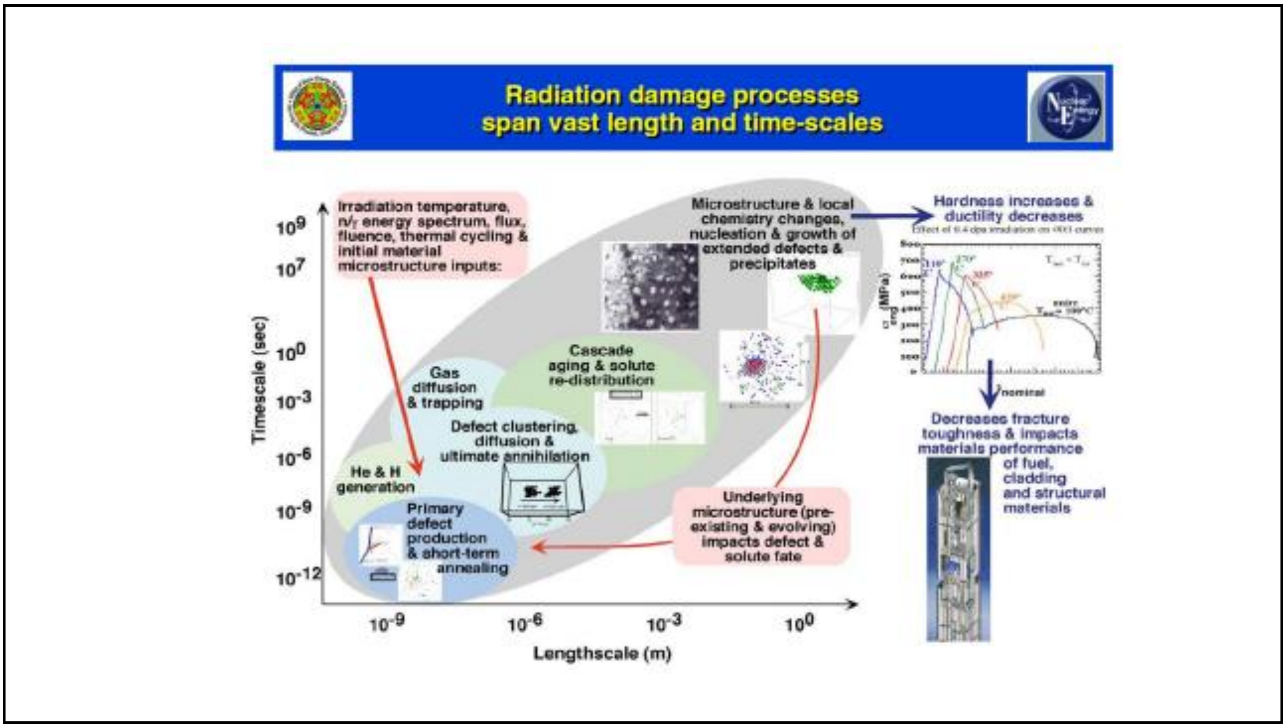
Want: Accuracy of quantum mechanics (QM) in 10^{23} atom systems…

This is impossible (today and in the foreseeable future)

Possible solution:
Multi-scale modeling techniques based on hierarchies of overlapping scales

Concept:
"finer scales train coarser scales by overlap"

© 2005 Markus J. Buehler, CEE/MIT



Radiation damage processes span vast length and time-scales

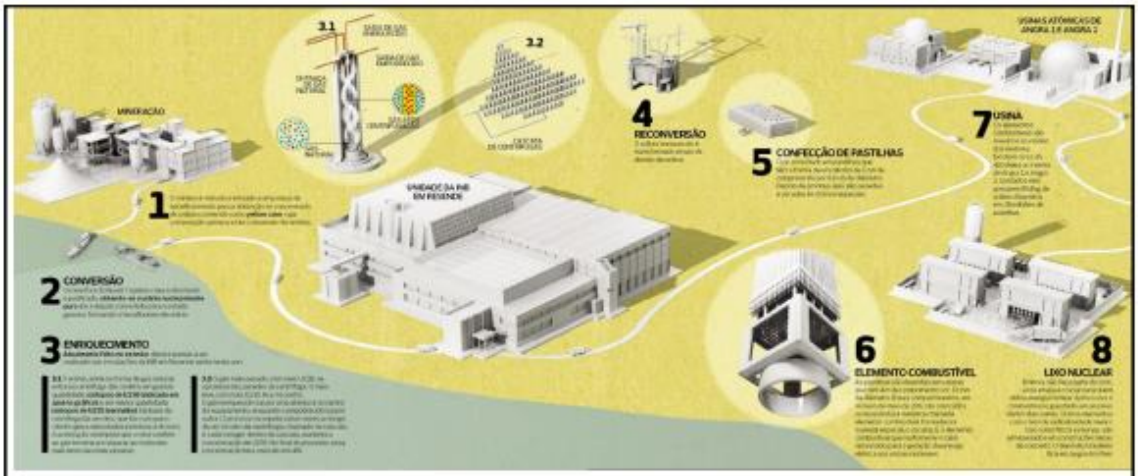
Irradiation temperature, n/γ energy spectrum, flux, fluence, thermal cycling & initial material microstructure inputs:

Microstructure & local chemistry changes, nucleation & growth of extended defects & precipitates

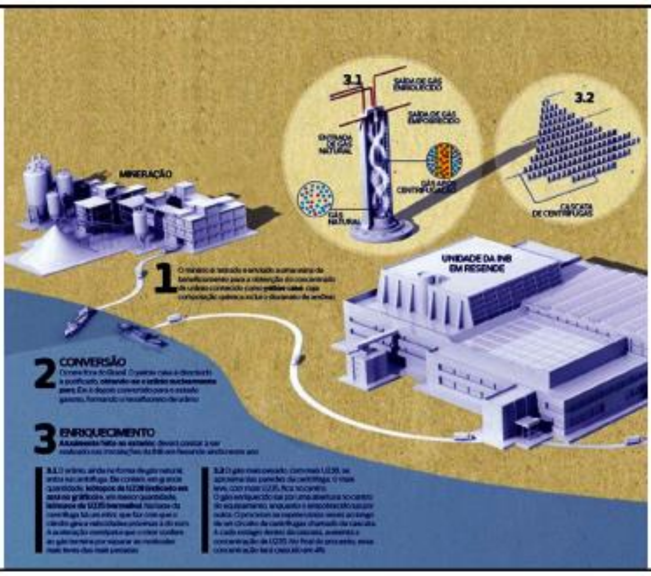
Hardness increases & ductility decreases

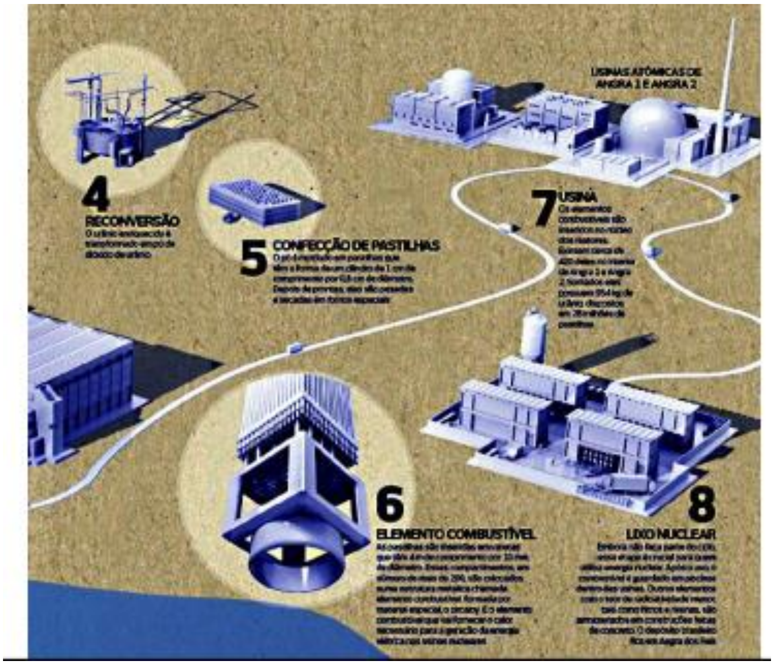
Decreases fracture toughness & impacts materials performance of fuel, cladding and structural materials

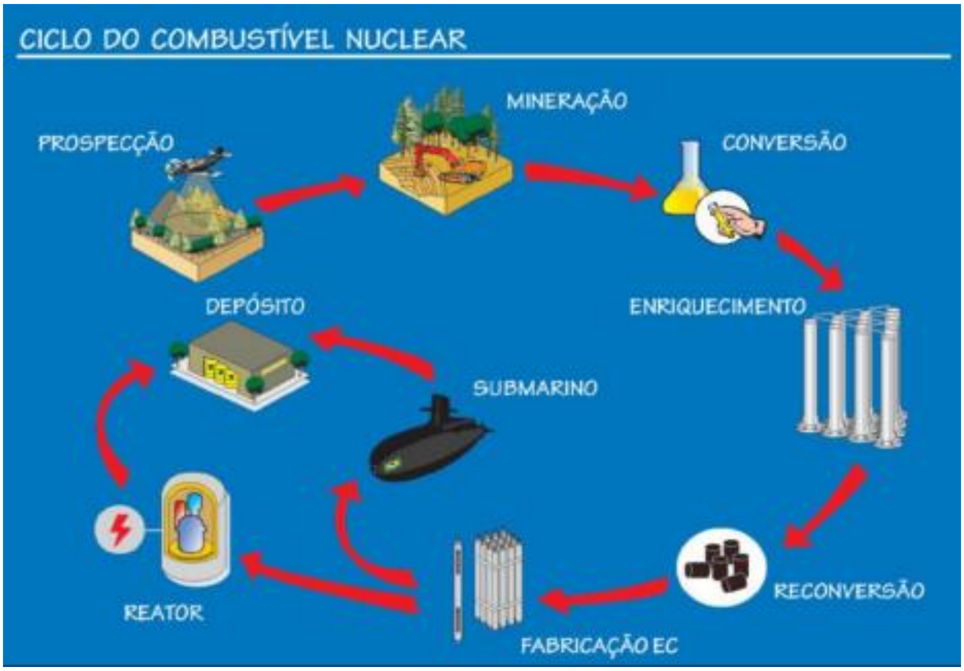
Underlying microstructure (pre-existing & evolving) impacts defect & solute fate



Ciclo do Combustível - Urânio




USINAS ATÔMICAS DE ANGRA 1 E ANGRA 2
4
RECONVERSÃO
5
CONFECÇÃO DE PASTILHAS
7
USINA
6
ELEMENTO COMBUSTÍVEL
8
LIXO NUCLEAR



CICLO DO COMBUSTÍVEL NUCLEAR

PROSPECÇÃO

MINERAÇÃO

CONVERSÃO

ENRIQUECIMENTO

RECONVERSÃO

FABRICAÇÃO EC

REATOR

SUBMARINO

DEPÓSITO

Ciclo do Combustível - Urânio

Usina nuclear de 1000 MW de eletricidade em um ano

80.000 t de minério

250 t urânio natural

35 t urânio enriquecido

215 t urânio empobrecido

Queima do urânio-235

35 t combustível queimado
33.4 t urânio-238
0.3 t urânio-235
0.3 t plutônio
1.0 t produtos de fissão

COMBUSTÍVEL NUCLEAR

- Atender às necessidades de **ENERGIA**
  - Auto-suficiência na produção de combustível para as *usinas nucleares*
- Atender às necessidades de **DEFESA**
  - Auto-suficiência na produção de combustível para o *protótipo e submarinos*
- Atender às necessidades de **APLICAÇÕES**
  - Auto-suficiência na produção de combustível para os *reatores de pesquisa e produção de radioisótopos*

Resende
RJ

Unidade de produção de pastilhas da Fábrica de Combustível Nuclear da INB

FCN - COMPONENTES E MONTAGEM

TÜV CERT DIN EN ISO 9001

Certificado NBR ISO 9001 BRTÜV

Capacidade instalada:
240 t/ano de urânio enriquecido

FCN COMPONENTES E MONTAGEM

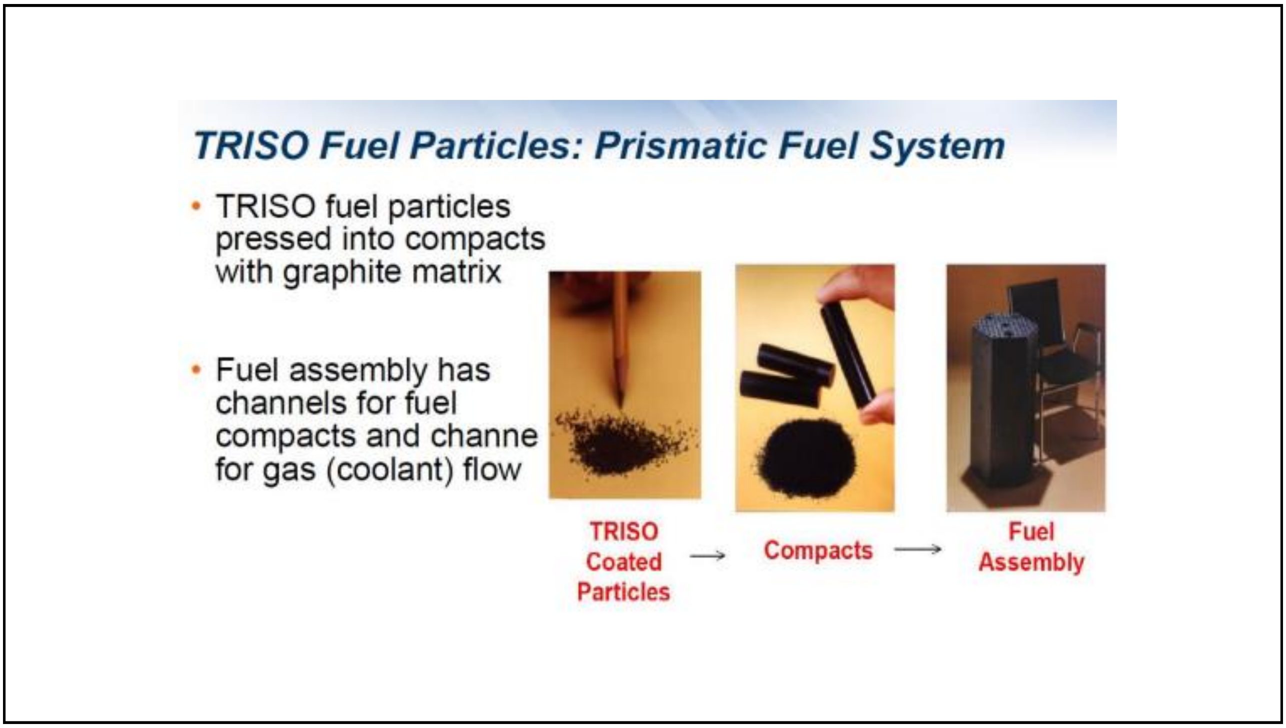
VISTA INTERNA

Fuel Types

- A nuclear fuel is a removable component that is introduced into a reactor core and contains the U (or Pu) to be fissioned in a reactor
- Nuclear fuels differ widely from reactor to reactor
  - Geometrical configuration of fuel and cladding
    - Fuel rods
    - Fuel plates
    - TRISO pellets
  - Materials used for U-bearing (or Pu) fuel
    - Ceramic compounds
    - Metallic alloys
  - Materials used for cladding
- Fuel-clad system is designed to
  - Produce and transfer heat to the coolant while
  - Prevent fission products from reaching the coolant

Uranium solid ceramic pellets

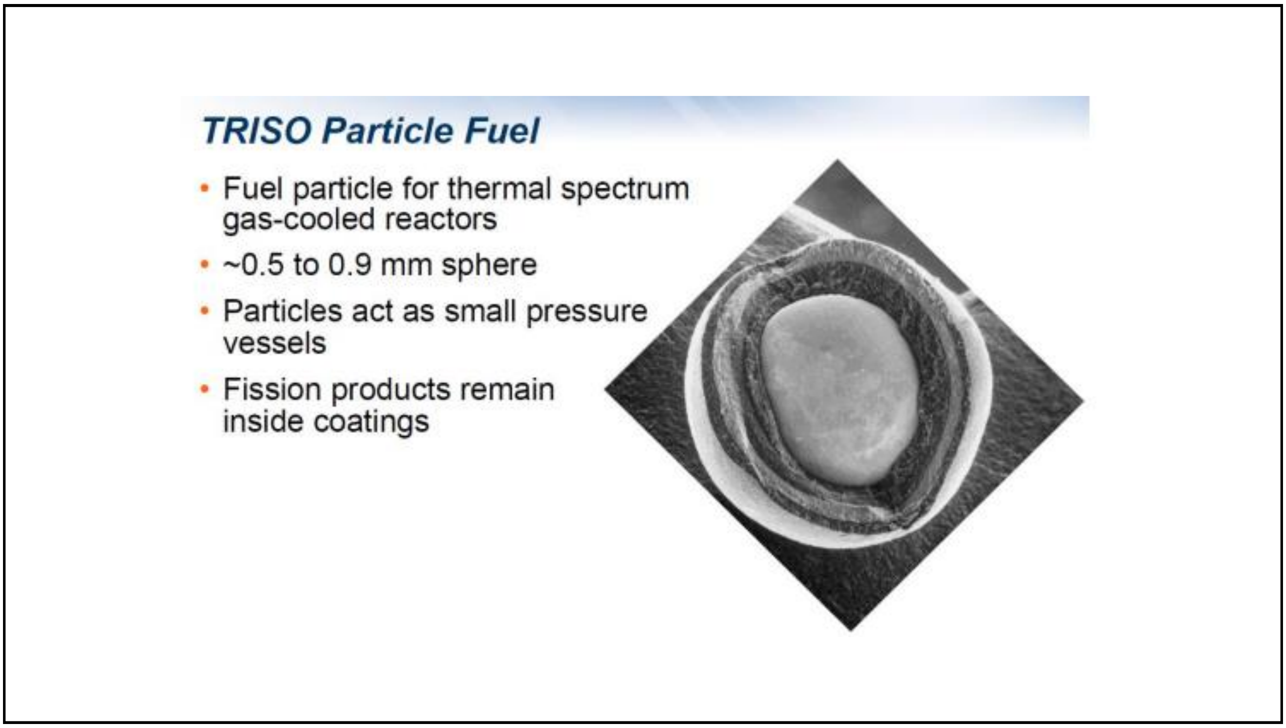
- Typical pellets:
  - 0.25" (6.3 mm) diameter
  - 0.50" (12 mm) height
- Pellet fabricated geometry
  - dish and chamfer
  - accommodate swelling
- U.S. Commercial Reactors
  - fuel enriched to 5% ^{235}U
  - ^{238}U breeds ^{239}Pu
  - ^{239}Pu fissions occur



TRISO Fuel Particles: Prismatic Fuel System

- TRISO fuel particles pressed into compacts with graphite matrix
- Fuel assembly has channels for fuel compacts and channe for gas (coolant) flow

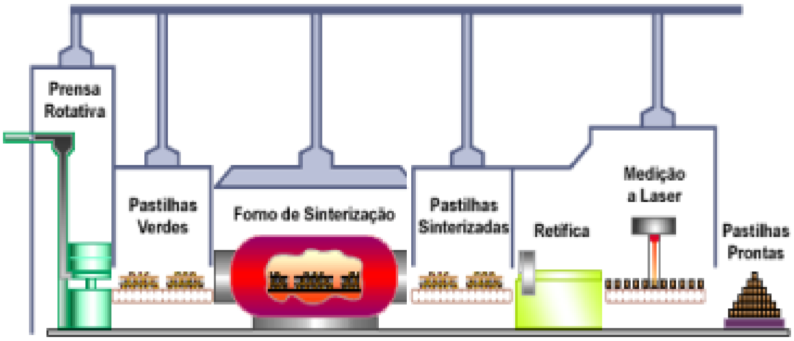
TRISO Coated Particles → **Compacts** → **Fuel Assembly**



TRISO Particle Fuel

- Fuel particle for thermal spectrum gas-cooled reactors
- ~0.5 to 0.9 mm sphere
- Particles act as small pressure vessels
- Fission products remain inside coatings

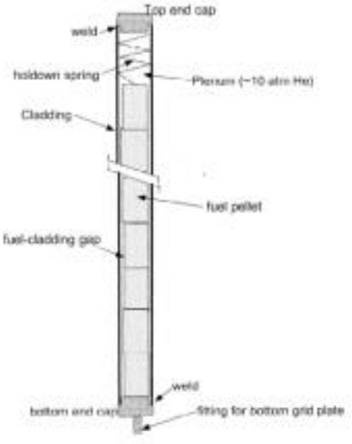
Processo de Fabricação de Pastilhas de UO_2



Fuel pellets grouped into fuel pins and assemblies

- Fuel height ~ 93 in. (3.7 m)
- 186 pellets/pin
- 264 pins/assembly
- 49,000 pellets/assembly
- 193 assemblies/core
- 9,473,000 pellets/core
- 1/3 of core replaced per refuel
- Refuel every 18 months
- 3,158,000 pellets/reload/reactor
- 104 commercial reactors in U.S.

LWR fuel elements



1000 MWe light water reactor

Quantity of fuel (UO_2), tons	100
Quantity of Zry, tons	25
Quantity of SS, tons	6
Control Materials*, tons	2
Neutron Fluxes: n/cm²-s Thermal Fast (>0.2 MeV)	 3×10^{13} 5×10^{13}
Irradiation time, years	3 - 6

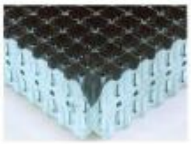
PWR fuel element characteristics

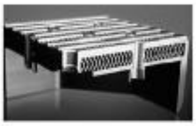
Fuel	UO_2 up to 5% enriched in ^{235}U
Fuel-pellet density	96% of theoretical density
Cladding	Zircaloy
Maximum cladding temperature	340°C
Maximum fuel temperature	1500°C
Coolant temperature	280 → 320°C
Maximum rod linear power	~400 W/cm
Fuel assembly	square, 20x20 cm (16x16 rods)
Fuel-rod outside diameter	10.7 mm
Cladding thickness	0.6 mm
Initial fuel-cladding radial gap	80 μm
Length of fuel portion	365 cm
Length of plenum	20 cm

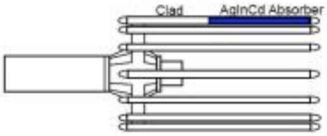
** Ag-Cd-In alloy (PWR); B_4C (BWR); Gd_2O_3 (mixed with UO_2)*

Fuel Assembly – PWR

- Fuel Assembly
  - Fuel rods
    - Cladding
    - Pellets
  - Grids (Spacers)
  - Guide tubes
  - Nozzles (tie plates)
- Control Rods

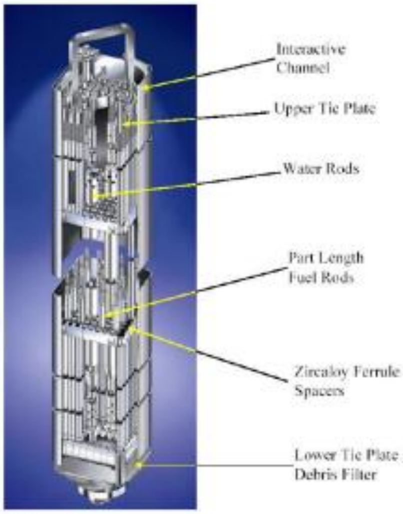


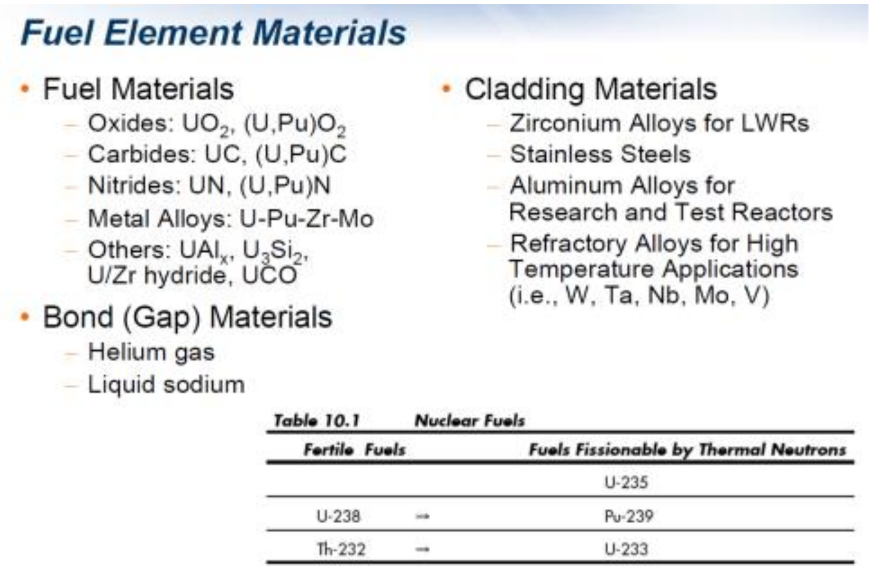




BWR Fuel

- Fuel Assembly
  - Fuel rods
    - Cladding
    - Pellets
  - Spacers
  - Water Rods / Box
  - Tie Plates
  - Channel
- Control Blades



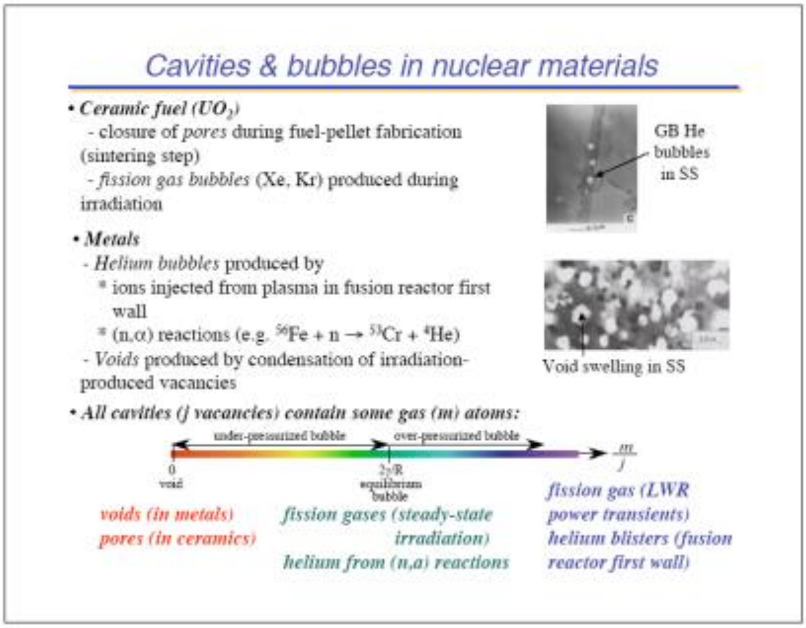


Fuel Element Materials

- Fuel Materials
  - Oxides: UO_2, $(U,Pu)O_2$
  - Carbides: UC, (U,Pu)C
  - Nitrides: UN, (U,Pu)N
  - Metal Alloys: U-Pu-Zr-Mo
  - Others: UAl_x, U_3Si_2, U/Zr hydride, UCO
- Bond (Gap) Materials
  - Helium gas
  - Liquid sodium
- Cladding Materials
  - Zirconium Alloys for LWRs
  - Stainless Steels
  - Aluminum Alloys for Research and Test Reactors
  - Refractory Alloys for High Temperature Applications (i.e., W, Ta, Nb, Mo, V)

Table 10.1 **Nuclear Fuels**

Fertile Fuels		Fuels Fissionable by Thermal Neutrons
		U-235
U-238	→	Pu-239
Th-232	→	U-233



Cavities & bubbles in nuclear materials

- ***Ceramic fuel (UO_2)***
  - closure of *pores* during fuel-pellet fabrication (sintering step)
  - *fission gas bubbles* (Xe, Kr) produced during irradiation
- ***Metals***
  - *Helium bubbles* produced by
    - ions injected from plasma in fusion reactor first wall
    - (n,α) reactions (e.g. $^{56}Fe + n \rightarrow {}^{53}Cr + {}^{4}He$)
  - *Voids* produced by condensation of irradiation-produced vacancies
- ***All cavities (j vacancies) contain some gas (m) atoms:***

voids (in metals)
pores (in ceramics)

fission gases (steady-state irradiation)
helium from (n,a) reactions

fission gas (LWR power transients)
helium blisters (fusion reactor first wall)

Inchamento/Liberação de Gases de Fissão

- Produtos de Fissão

  -Dois átomos substituem cada átomo de U (ou Pu) que se fissiona

  -25% dos produtos de fissão são átomos de gases (Kr, Xe)

- Inchaço do combustível

  -O combustível incha devido à geração de produtos de fissão

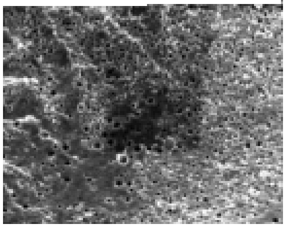
  -Átomos de gases coalescem em bolhas, acelerando o inchamento

  -O inchaço do combustível tende à reduzir ou fechar o “gap”

- Liberação de Gases de Fissão

  -Parte dos gases de fissão escapa do combustível (menos que 10%)

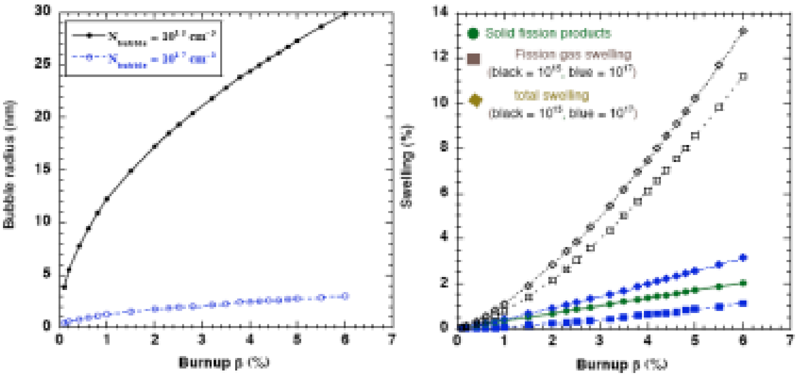
  -Pressionam o plenum



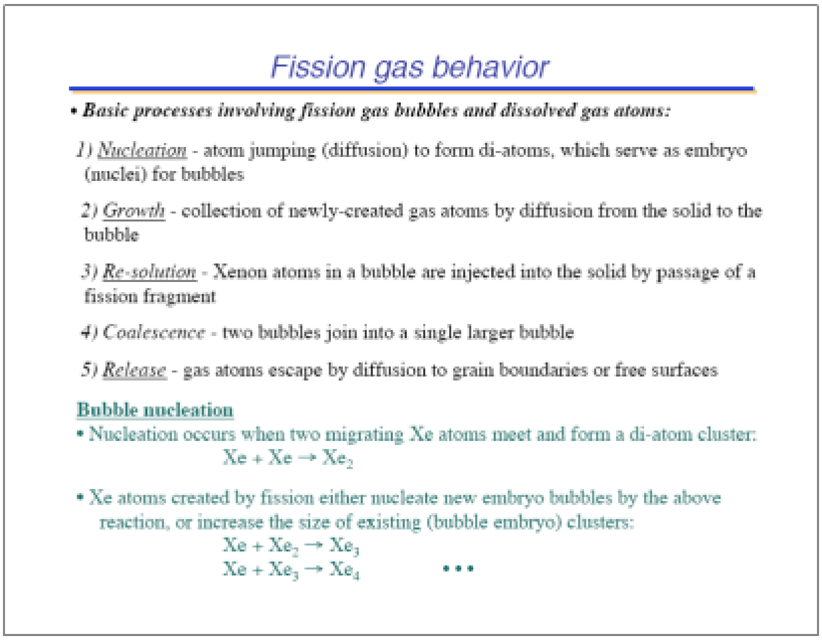
bolhas no combustível

Fuel swelling due to fission gases

Example of fission gas radius and swelling for no re-solution.



Less swelling with more numerous, but smaller bubbles!



Fission gas behavior

- ***Basic processes involving fission gas bubbles and dissolved gas atoms:***

1) *Nucleation* - atom jumping (diffusion) to form di-atoms, which serve as embryo (nuclei) for bubbles

2) *Growth* - collection of newly-created gas atoms by diffusion from the solid to the bubble

3) *Re-solution* - Xenon atoms in a bubble are injected into the solid by passage of a fission fragment

4) *Coalescence* - two bubbles join into a single larger bubble

5) *Release* - gas atoms escape by diffusion to grain boundaries or free surfaces

Bubble nucleation

- Nucleation occurs when two migrating Xe atoms meet and form a di-atom cluster:

$$Xe + Xe \rightarrow Xe_2$$

- Xe atoms created by fission either nucleate new embryo bubbles by the above reaction, or increase the size of existing (bubble embryo) clusters:

$$Xe + Xe_2 \rightarrow Xe_3$$
$$Xe + Xe_3 \rightarrow Xe_4 \qquad \cdots$$

Research explores safer fuel for nuclear reactors

March 21, 2016 by Shea Bracken

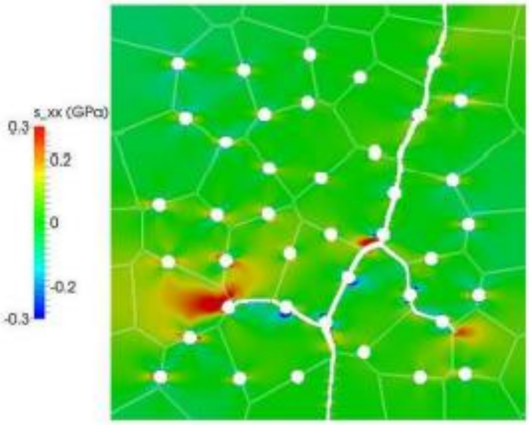
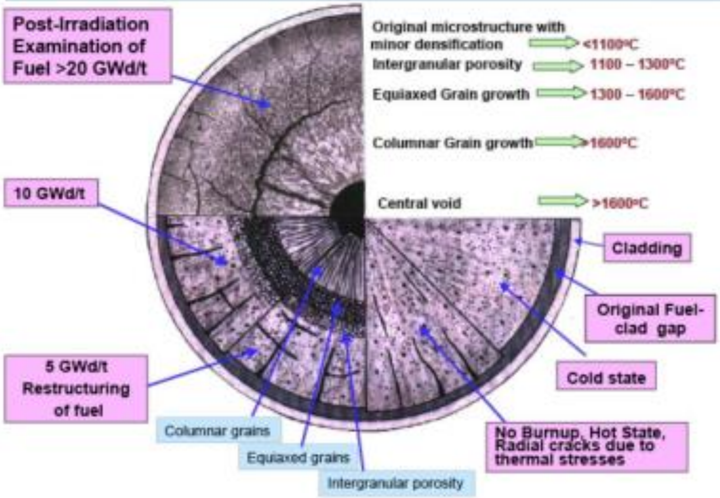


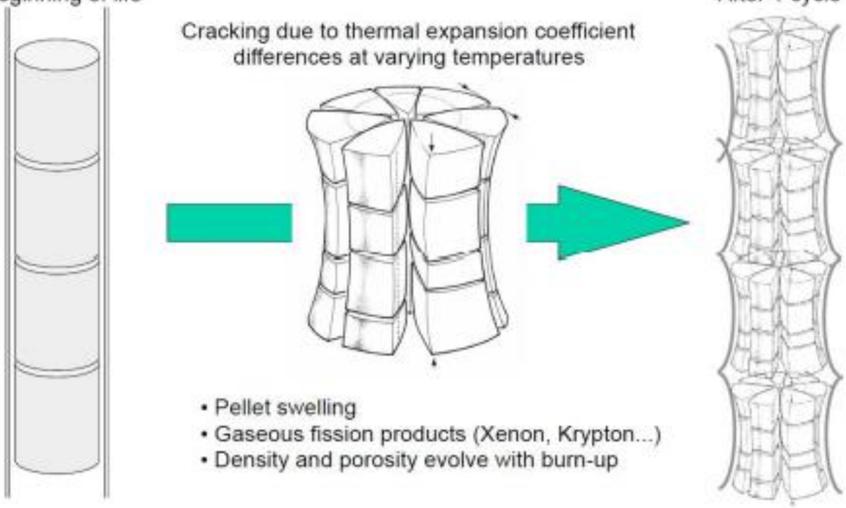
Image shows a mesoscale simulation used to predict the impact of fission gas bubbles on uranium dioxide fracture strength. Similar simulations will be used to investigate microcracking in silicon carbide composite cladding. Credit: Michael Tonks

Fuel Environment

Microstructural Evolution in Oxide Fuel

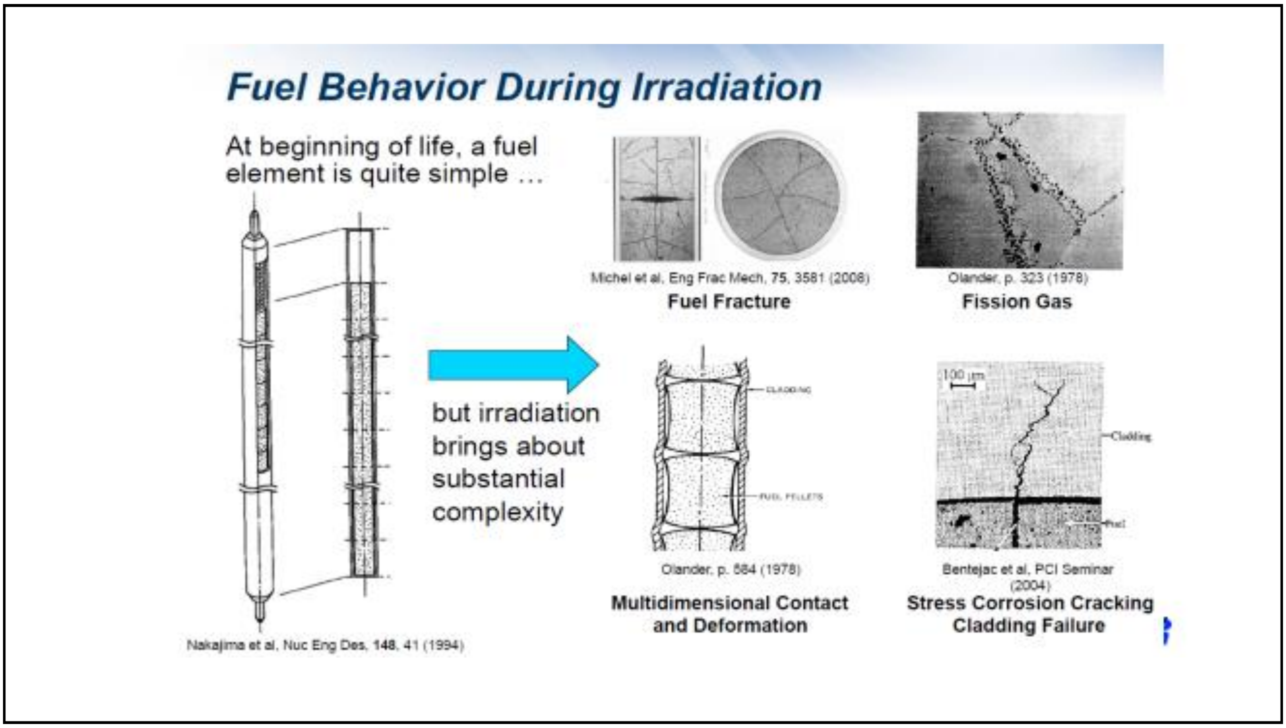


Fuel Response to Irradiation



Cracking due to thermal expansion coefficient differences at varying temperatures

- Pellet swelling
- Gaseous fission products (Xenon, Krypton...)
- Density and porosity evolve with burn-up



Fuel Behavior During Irradiation

At beginning of life, a fuel element is quite simple …

Nakajima et al, Nuc Eng Des, 148, 41 (1994)

but irradiation brings about substantial complexity

Michel et al, Eng Frac Mech, 75, 3581 (2008)

Fuel Fracture

Olander, p. 323 (1978)

Fission Gas

Olander, p. 584 (1978)

Multidimensional Contact and Deformation

Bentejac et al, PCI Seminar (2004)

Stress Corrosion Cracking Cladding Failure