

Challenges for Reactor Materials

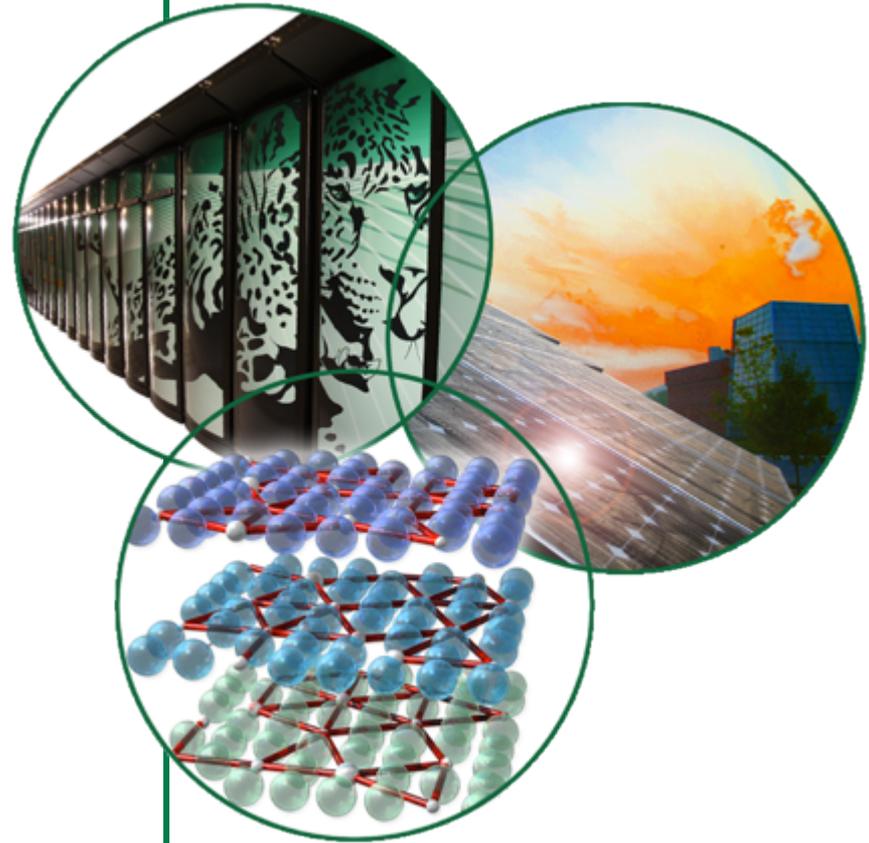
J.T. Busby (with slides from many)

**Fuel Cycle and Isotopes Division
Oak Ridge National Laboratory**

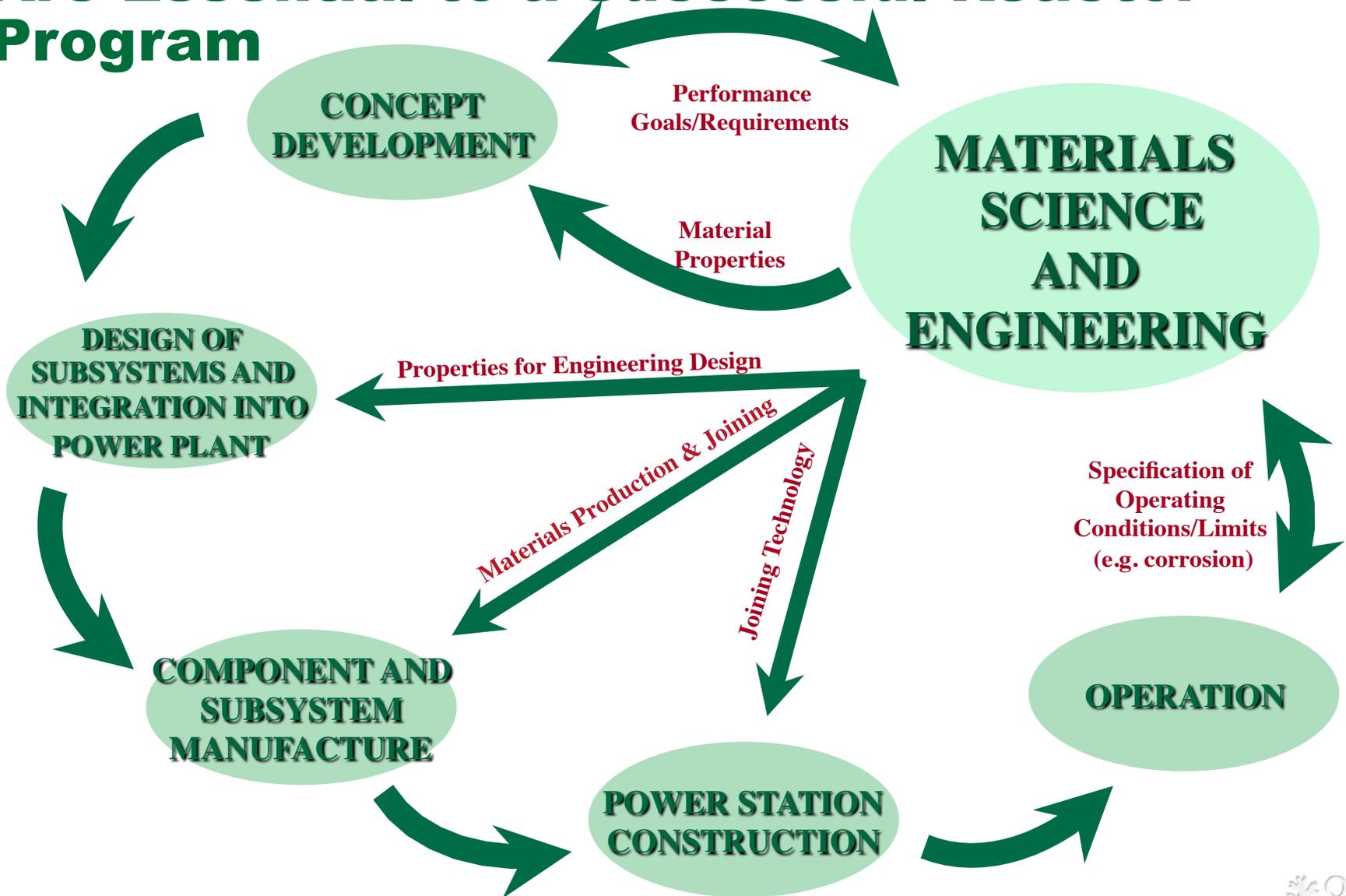
2012 Nanonuclear Workshop

February 28, 2012

Rice University

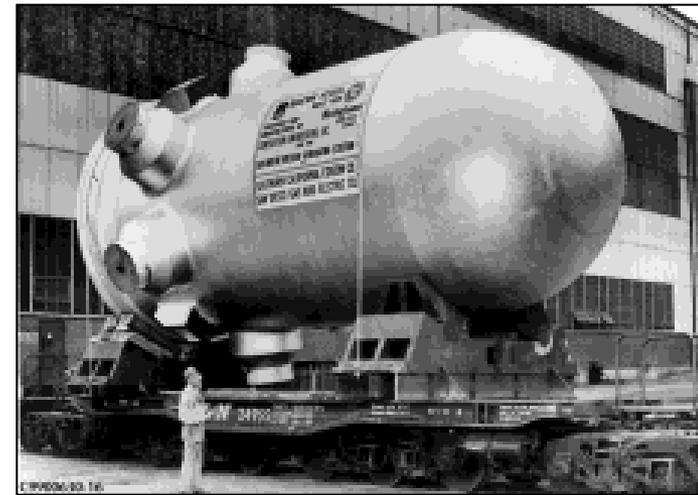
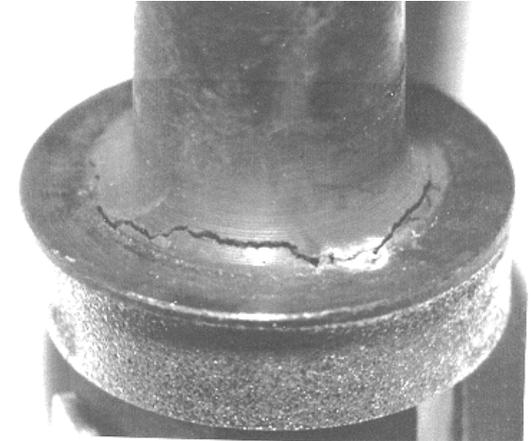


Sound Materials Science and Engineering Are Essential to a Successful Reactor Program



Materials performance: Why do we care?

- Materials degradation and performance is a common problem/concern in existing reactors/nuclear facilities
- Understanding the long-term behavior of materials in the reactor core, vessel, and many other subsystems is critical for safe, reliable, reactor operation.
- Understanding materials performance is a key need in designing any new reactor facility.



• *Understanding the limitations of materials in nuclear reactor applications will be key in moving forward in the “nuclear renaissance.”*

Materials issues are a key concern for the existing nuclear reactor fleet

- Materials research is already a key need for the existing nuclear reactor fleet
- Materials degradation can lead to increased maintenance, increased downtime, and increased risk.
- Materials issues must be resolved for:
 - Reactor Pressure Vessels and Primary Piping
 - Core Internals
 - Secondary System
 - Weldments
 - Concrete
 - Cabling
 - Buried Piping

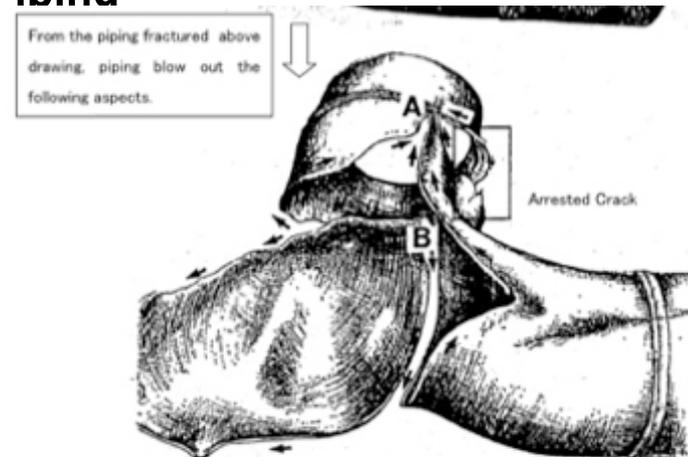
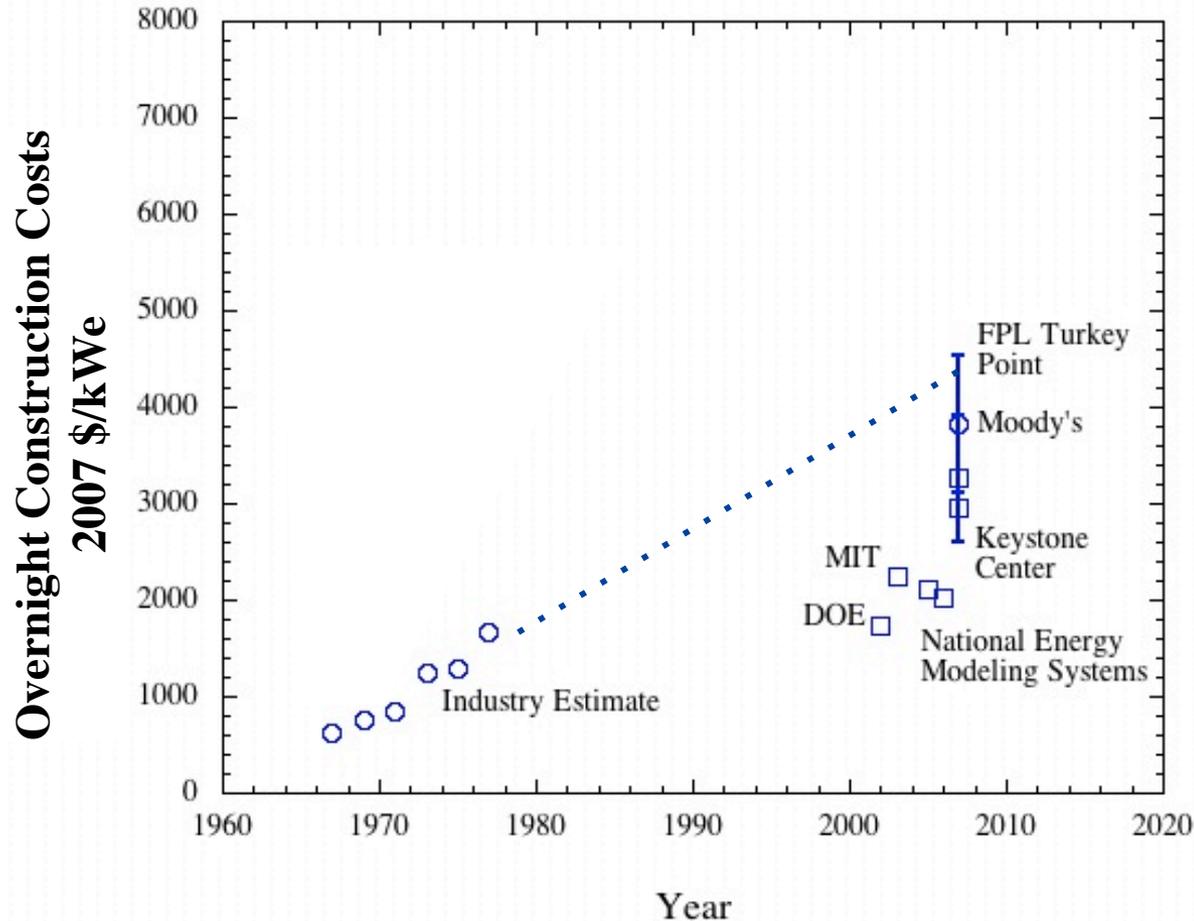


Fig.3 Detail Drawing of the Breaking Portion

Escalation of reactor construction costs provides a strong motivation for innovation in design and construction methods.



- *The investment required in a new nuclear plant is significant.*

Materials performance may have a significant impact on the economic case for nuclear power

Drivers of Nuclear Generation Cost Reduction*

Driver	Unit Cost Factor Improved
Series effect (from FOAK to NOAK)	Capital Investment
Design Standardization	Capital Investment and O&M
Design Simplification	Capital Investment and O&M
Multi-unit sites	Capital Investment and O&M
Decreasing Construction Time	Capital Investment
Increasing Power Level	Capital Investment
Increasing Availability Factor	Capital Investment
Increasing Plant Life	Capital Investment
Increasing Fuel Burnup	Fuel Costs

• **Materials performance can positively and directly influence these factors.**

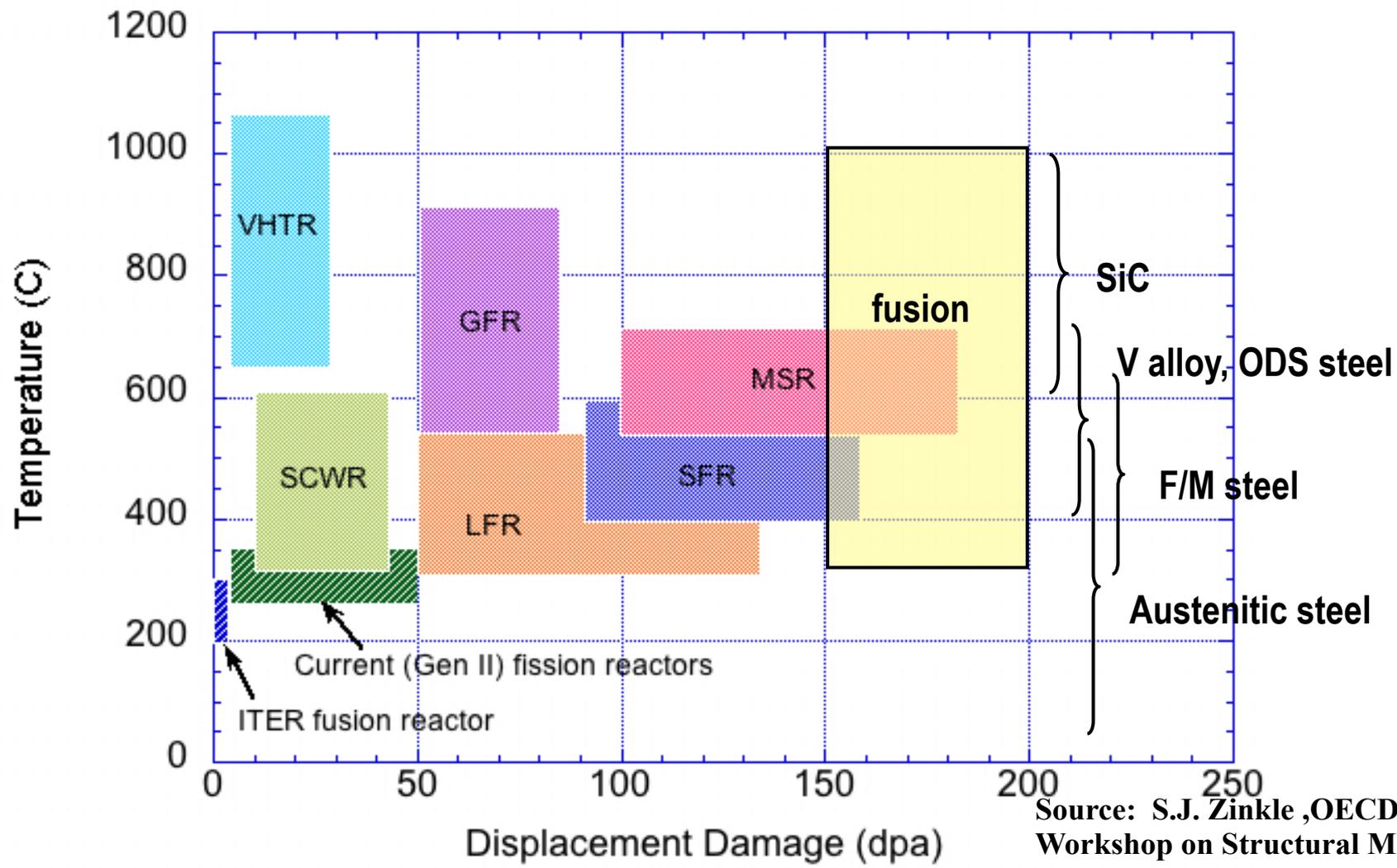
This presentation will describe a science-based approach to help overcome past materials limitations

Outline:

- **Motivation for understanding materials performance in reactor systems**
- **Common materials of construction**
- **Complexity of nuclear systems and requirements for materials use**
- **Challenges in different reactor systems**
 - **Irradiation damage**
 - **Corrosion processes**
 - **Other select processes**
 - **Concrete**

Materials of construction for reactor systems

Service environments and material choices vary widely between reactor concepts



Source: S.J. Zinkle, OECD NEA Workshop on Structural Materials for Innovative Nuclear Energy Systems, Karlsruhe, Germany, June 2007

Construction materials for current reactor designs are diverse

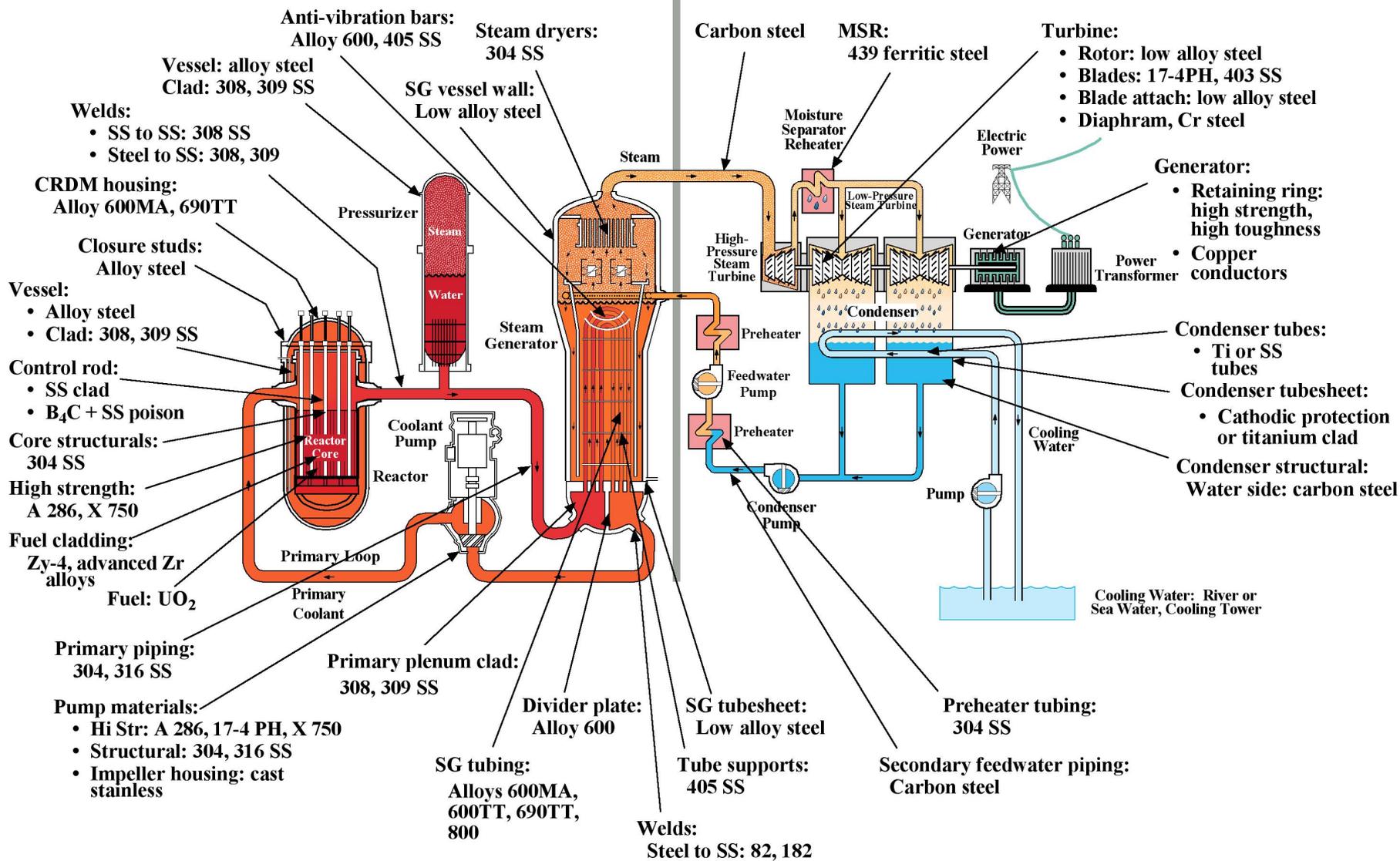
	LWR	SFR	GFR/VHTR
Coolant	Water	Sodium	Helium
Temperature	288-360°C	500-550°C	550-1100°C
Cladding	Zirconium-based	9 or 12Cr steels	SiC/SiC
Core Internals	304/316 SS	316 SS	SiC/Alloy 800H
Vessel	Steel/316 SS	316 SS	Steel/316 SS
Heat Exchanger	Alloy 600/690	9-12Cr/316 SS	Alloy 617
Piping	SS/LA Steel	9-12Cr/316 SS	Alloy 617

- **Despite considerable differences in operating parameters, there are common material uses between LWR and SFR applications**

Materials in PWRs

Primary Circuit

Secondary Circuit

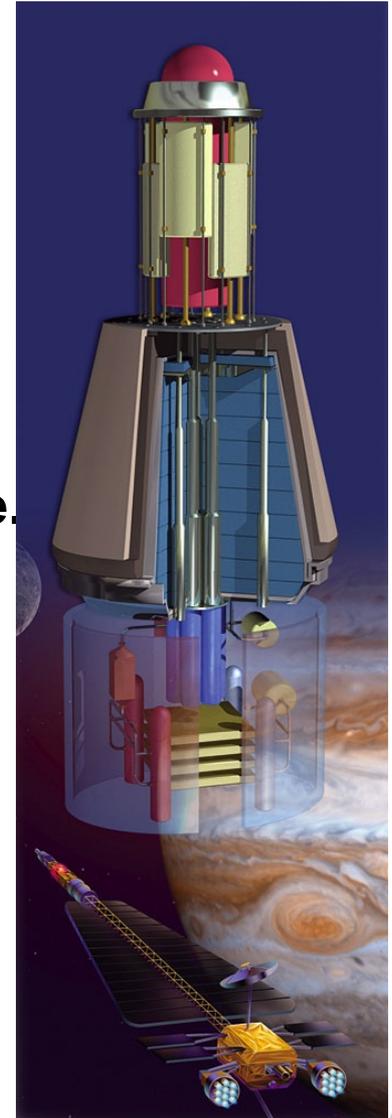


Material selection in a reactor environment is complex

- **Many factors are important for reactor service.**
 - Availability
 - Cost
 - Fabrication/Processing
 - Reproducibility/Uniformity
 - Irradiation resistance
 - Mechanical performance
 - Creep performance
 - Corrosion performance
 - Thermal properties
 - Joining
 - Fatigue
 - Fracture toughness
- **All materials must also meet all regulatory (or code qualification) standards.**

Structural materials are a critical component for space fission reactor performance

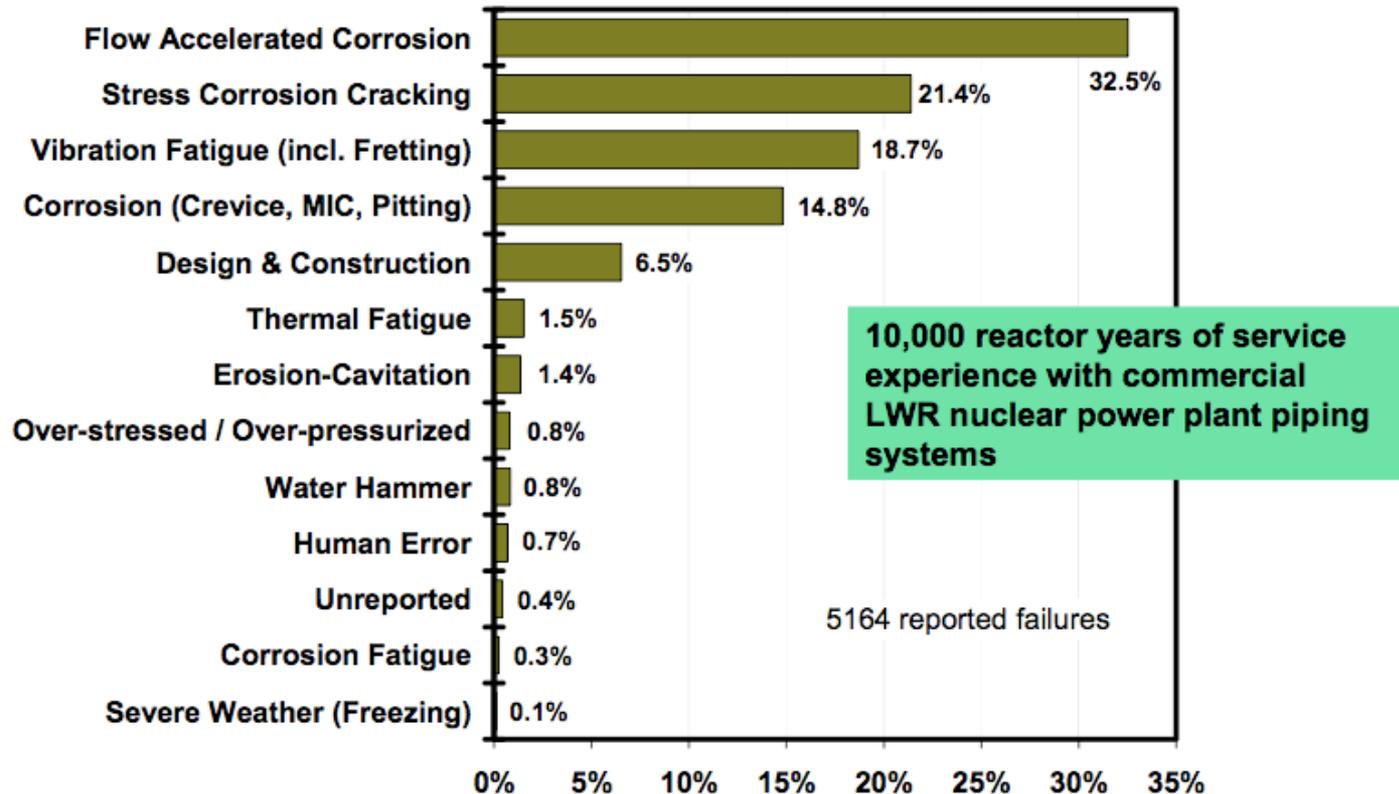
- Space reactors are complex systems and create a harsh environment for structural materials
 - High irradiation fields
 - Often liquid metal coolants
 - Long lifetimes under stress.
 - No opportunity for maintenance or surveillance.
- Structural material performance will determine
 - Reactor temperature
 - Reactor lifetime
 - Reactor configuration
 - Many other design features (mass!)
- The selection of structural materials is a key consideration that impacts all phases of reactor design, construction, and operation.



Complexity of material performance in reactor systems

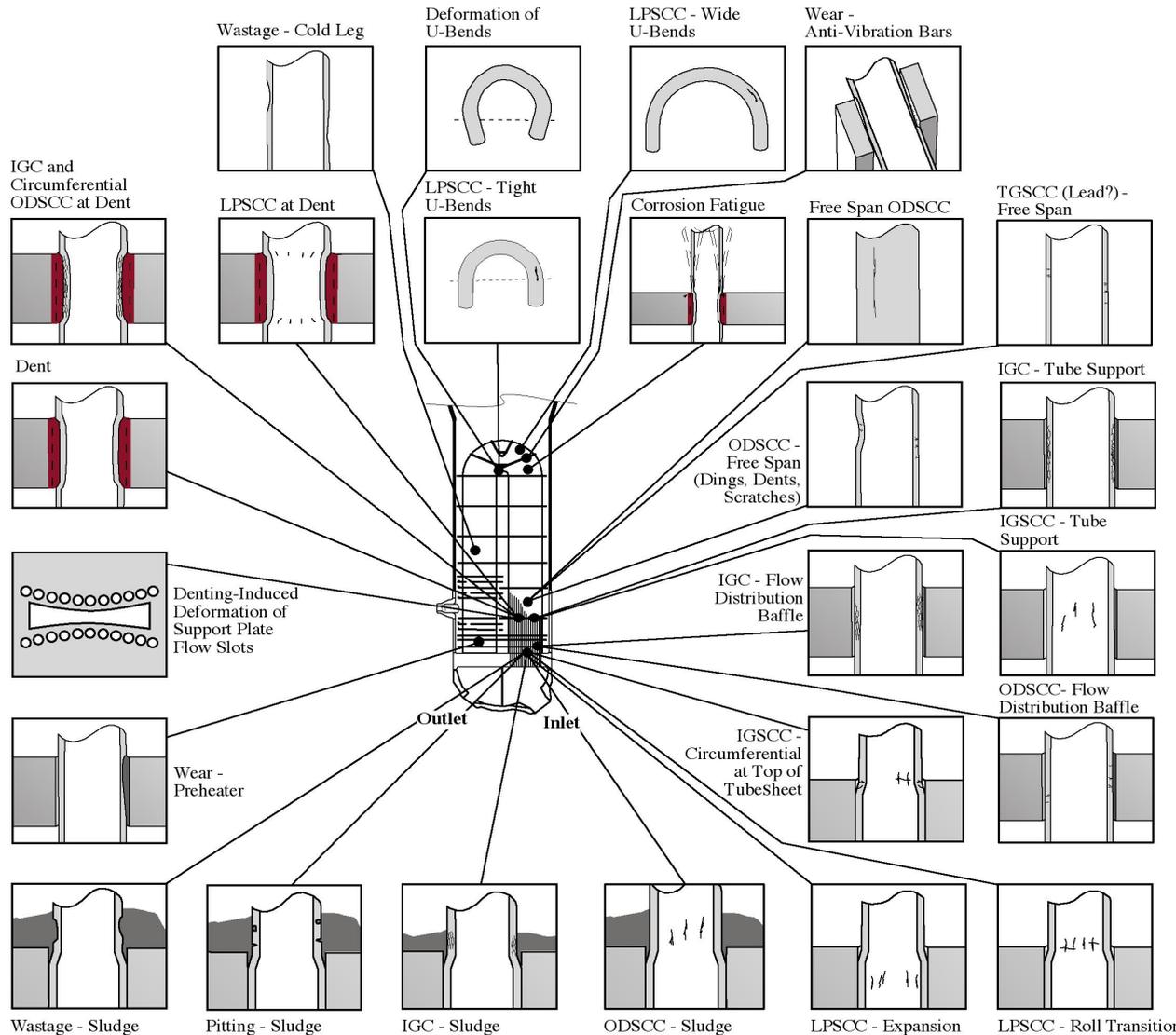
Even within a single component, many modes of degradation may exist

U.S. Failures by Degradation Mechanisms



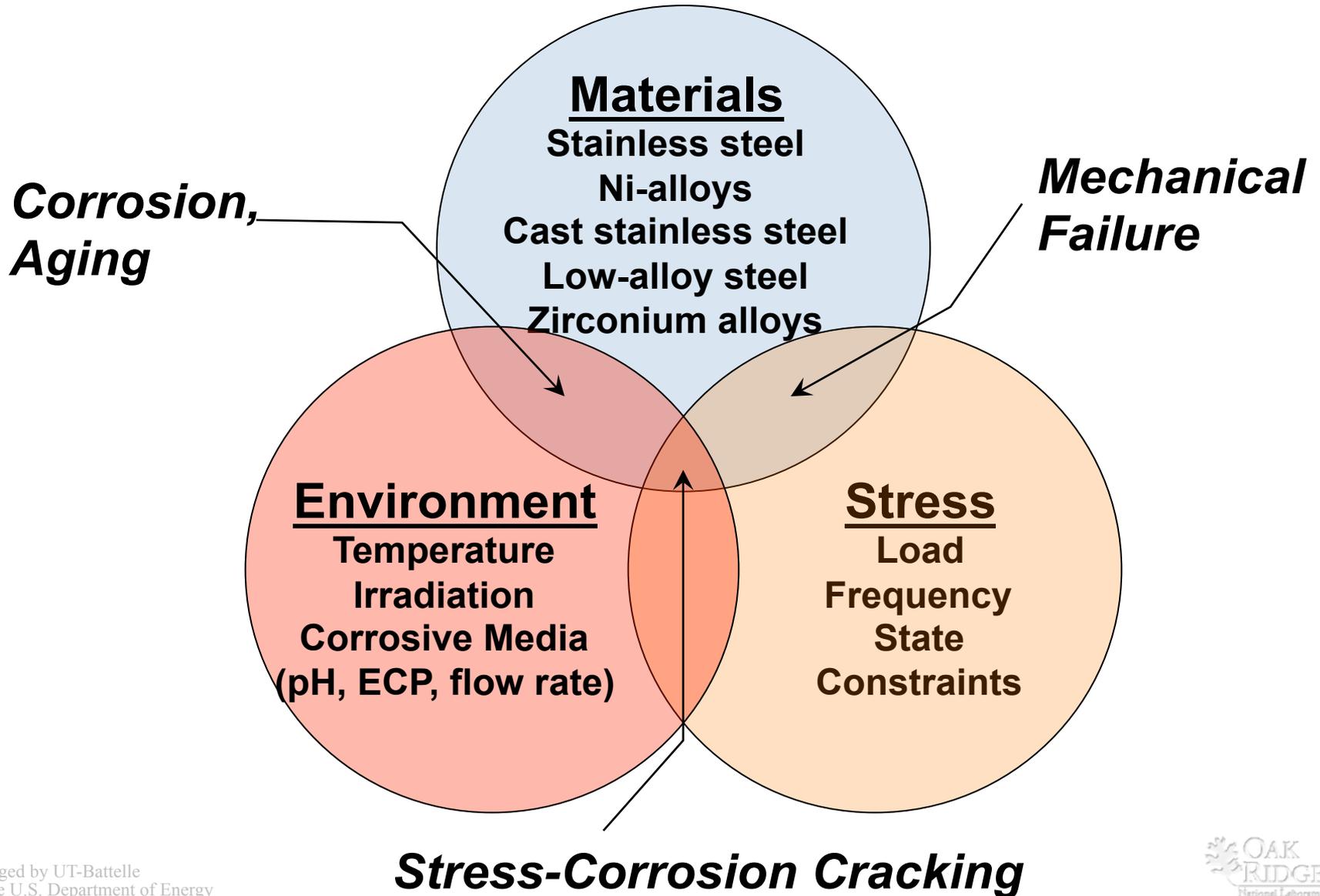
Source: PIPExp Database Data from 1970-2007

Materials degradation within subsystems is complex



25 different mode-location cases of corrosion have been identified in a SG with Alloy 600 tubes and drilled hole tube supports

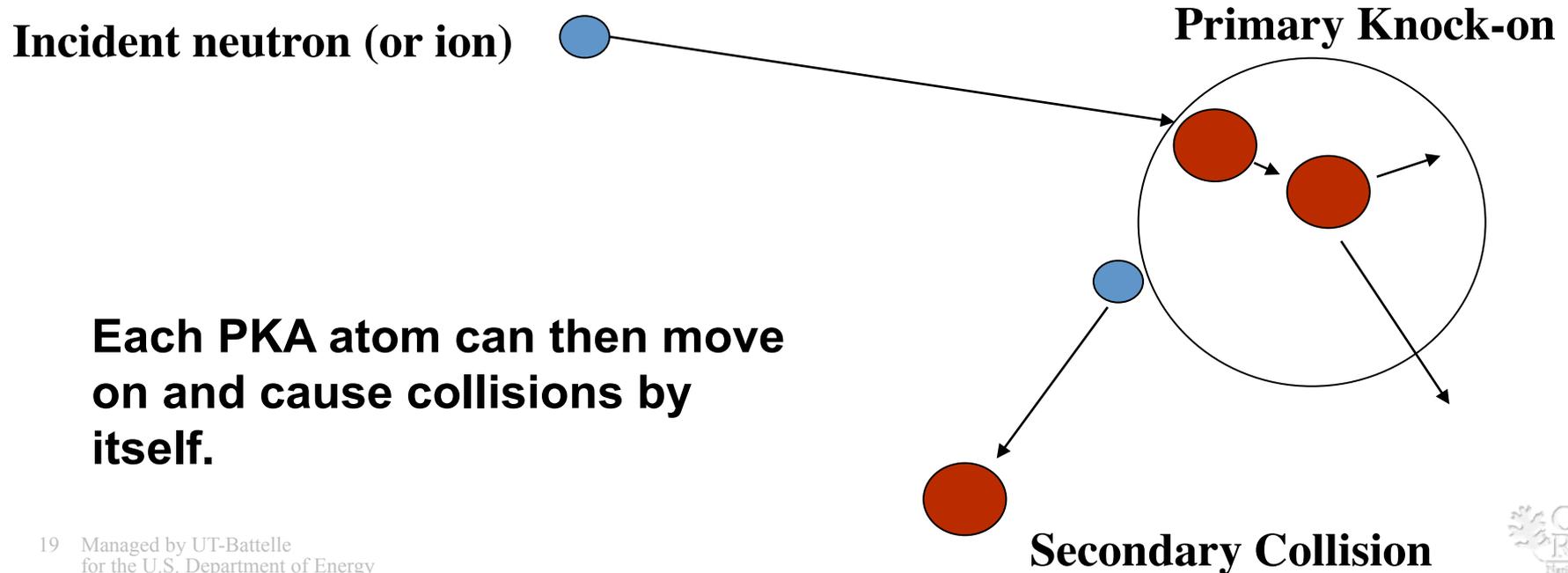
Materials aging and degradation in nuclear reactor systems is complex



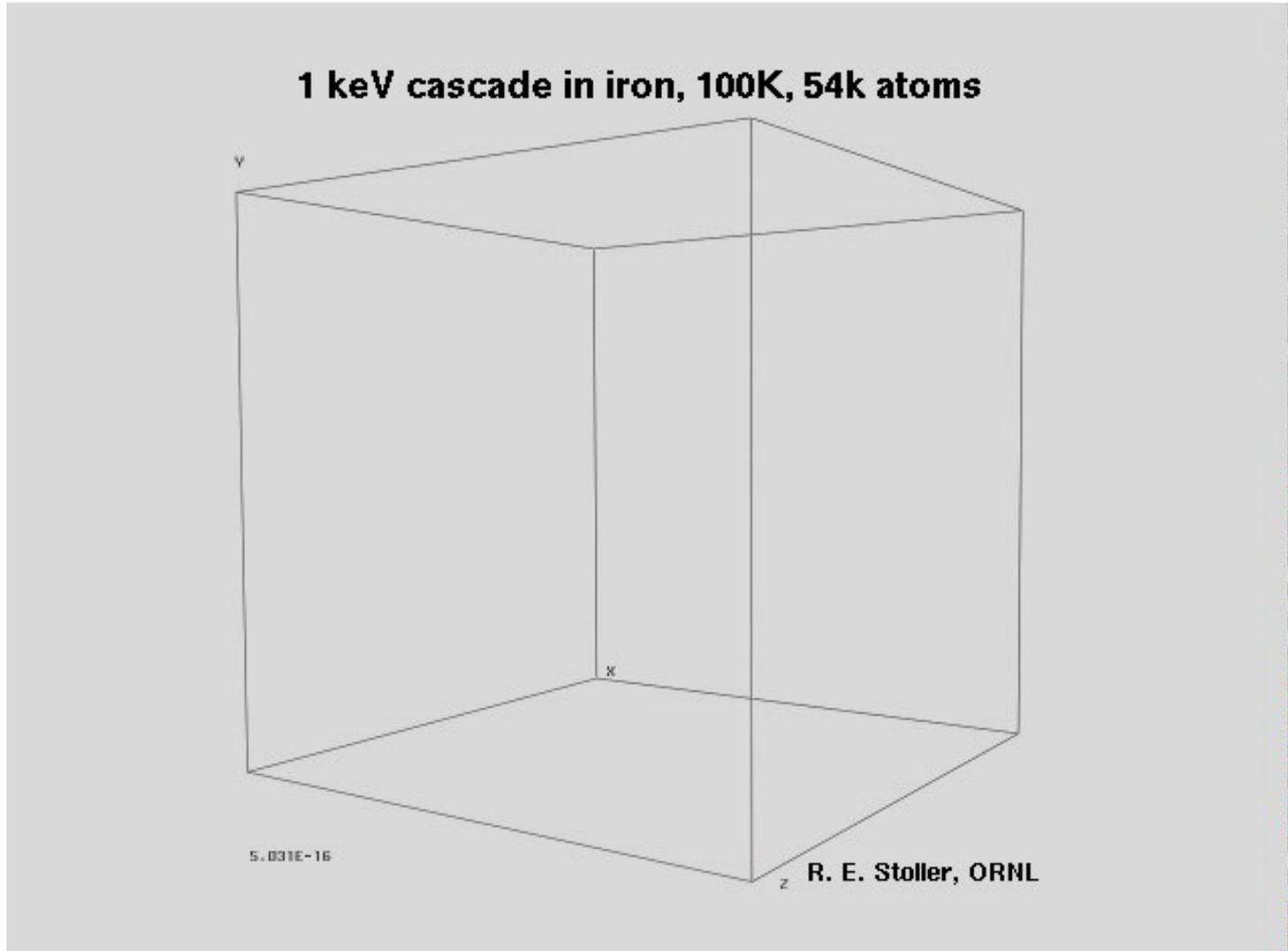
Challenges for material performance in reactor systems

Radiation Damage: the basics

- All of radiation damage boils down to a common step: **collisions between incoming neutrons and atoms in the crystal lattice!**

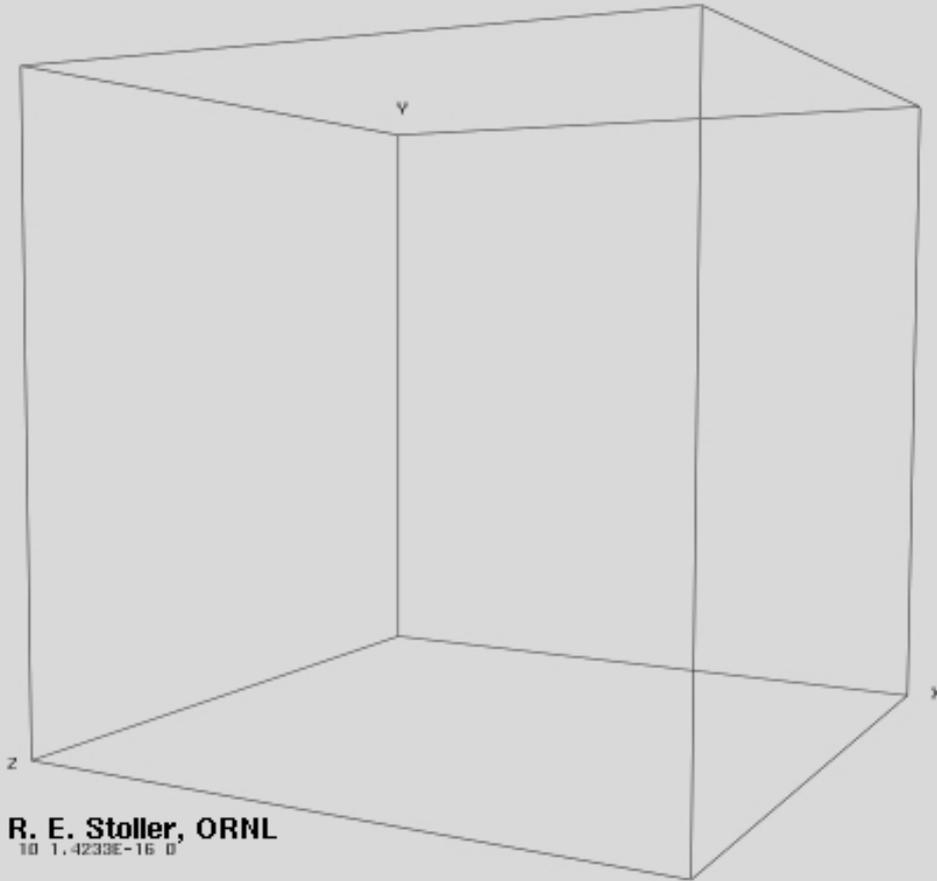


Molecular dynamic simulations provide a good picture of this process



Molecular dynamic simulations provide a good picture of this process

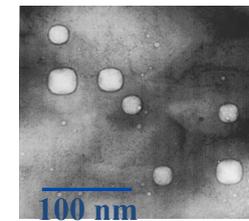
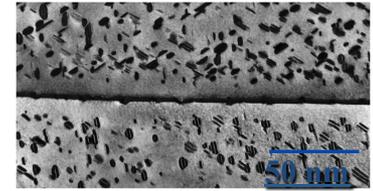
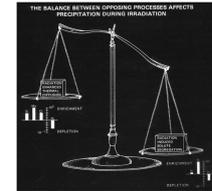
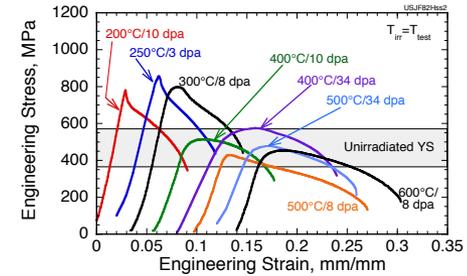
50 keV cascade in iron, 100K, ~2.25M atoms



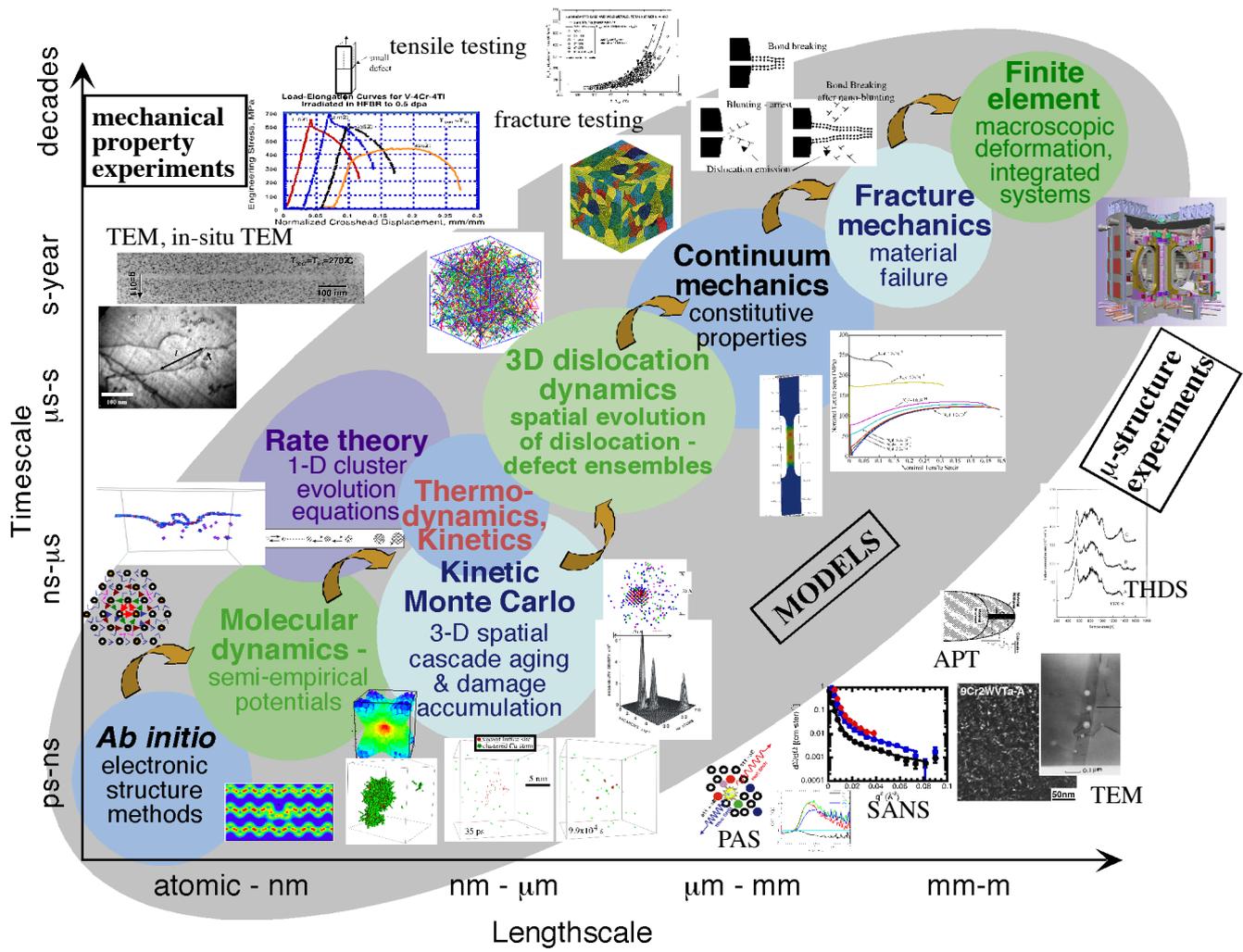
R. E. Stoller, ORNL
10 1.4233E-16 0

Radiation Damage can Produce Large Changes in Structural Materials

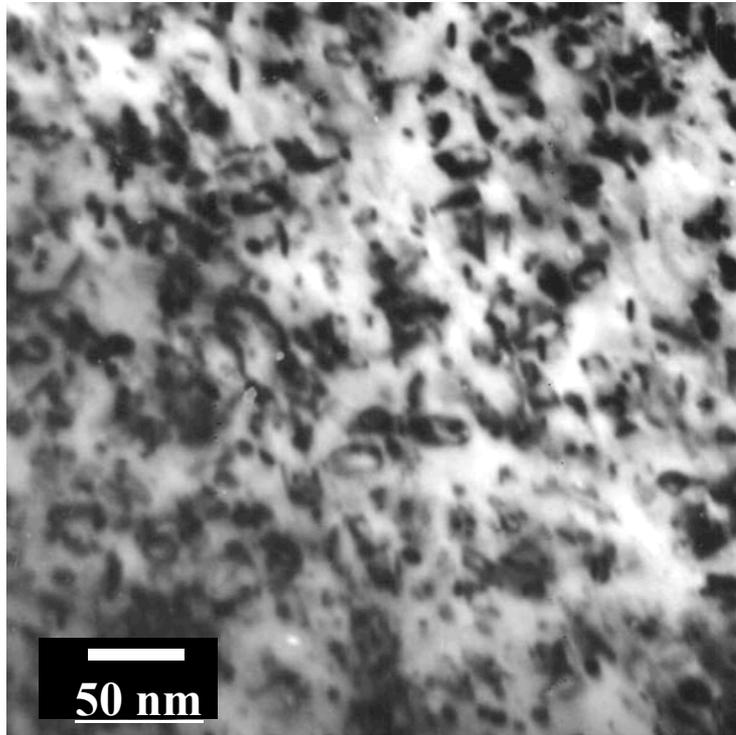
- **Radiation hardening and embrittlement ($<0.4 T_M$, >0.1 dpa)**
- **Phase instabilities from radiation-induced precipitation ($0.3-0.6 T_M$, >10 dpa)**
- **Irradiation creep ($<0.45 T_M$, >10 dpa)**
- **Volumetric swelling from void formation ($0.3-0.6 T_M$, >10 dpa)**
- **High temperature He embrittlement ($>0.5 T_M$, >10 dpa)**



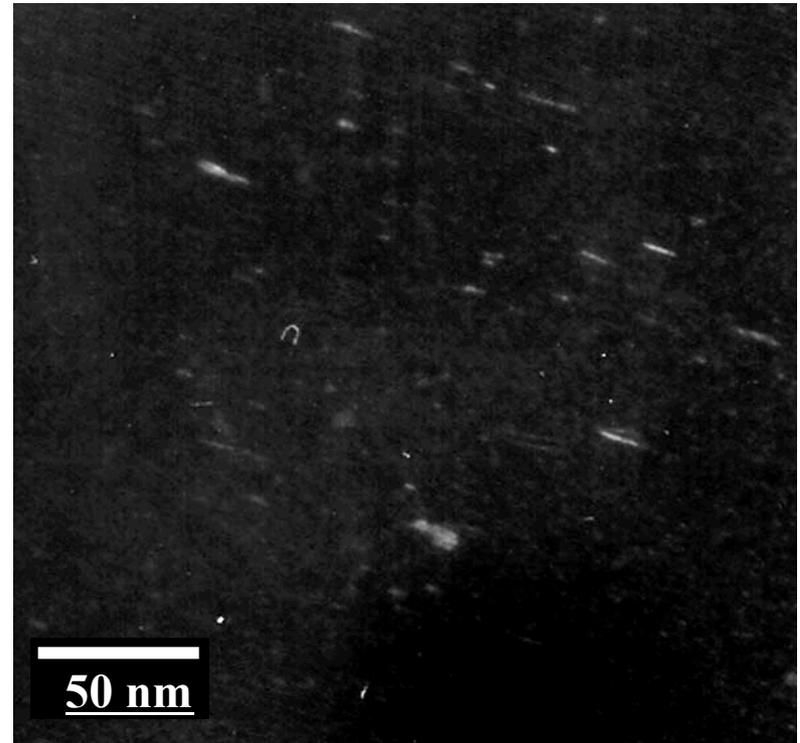
Radiation damage is inherently multiscale with interacting phenomena ranging from ps to decades and nm to m



Dislocation loop microstructure



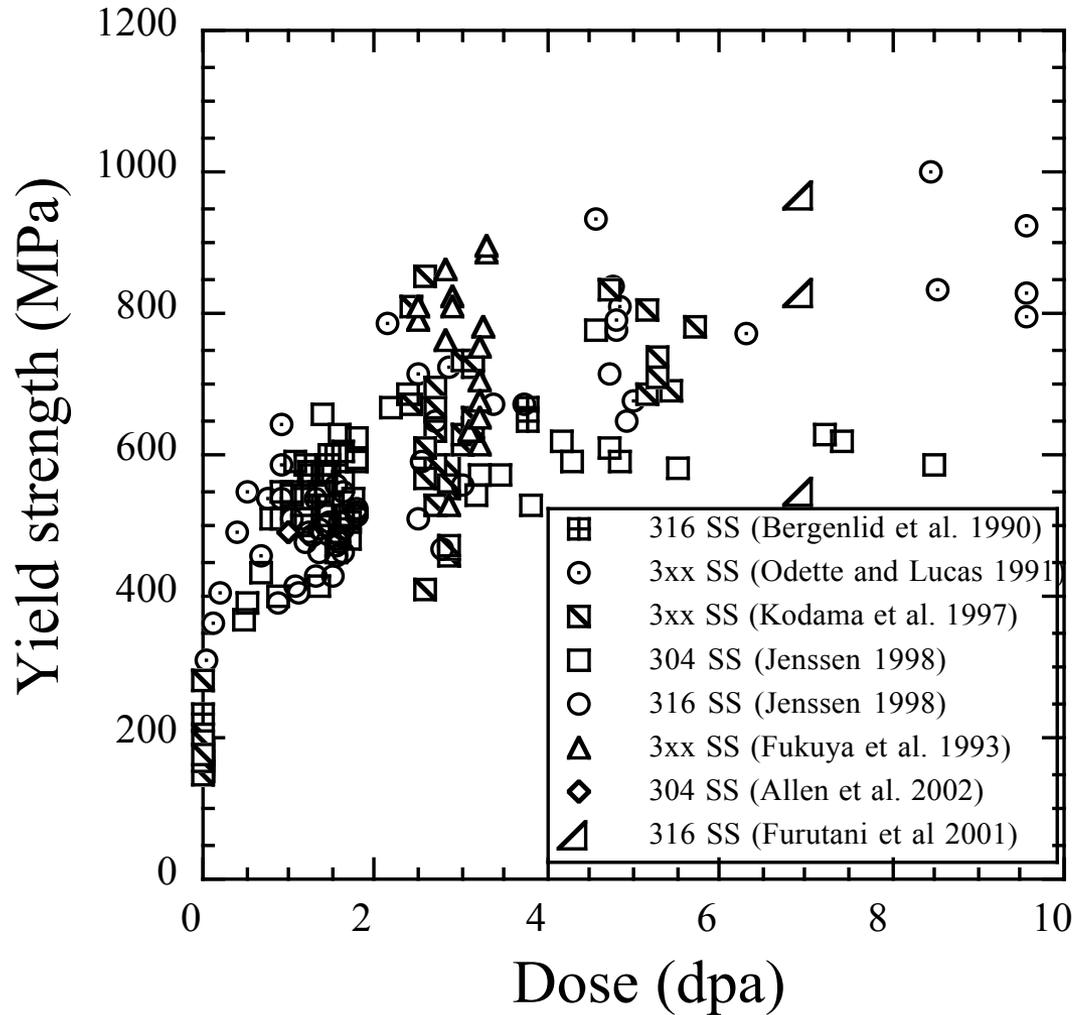
Bright Field



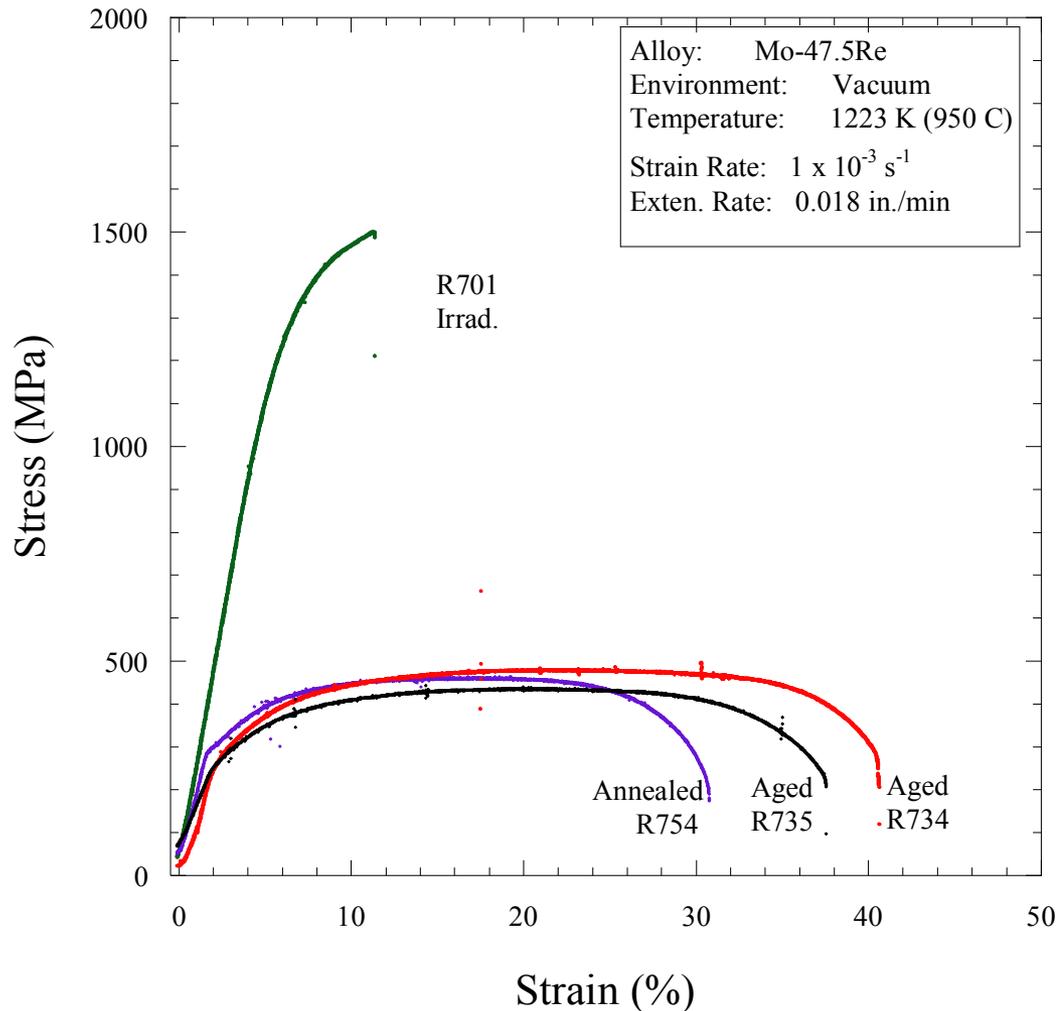
Dark Field

CP-304 SS irradiated to 0.55 dpa with protons at 360°C

Irradiation Hardening



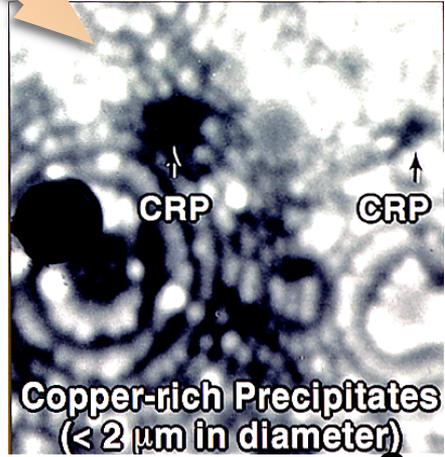
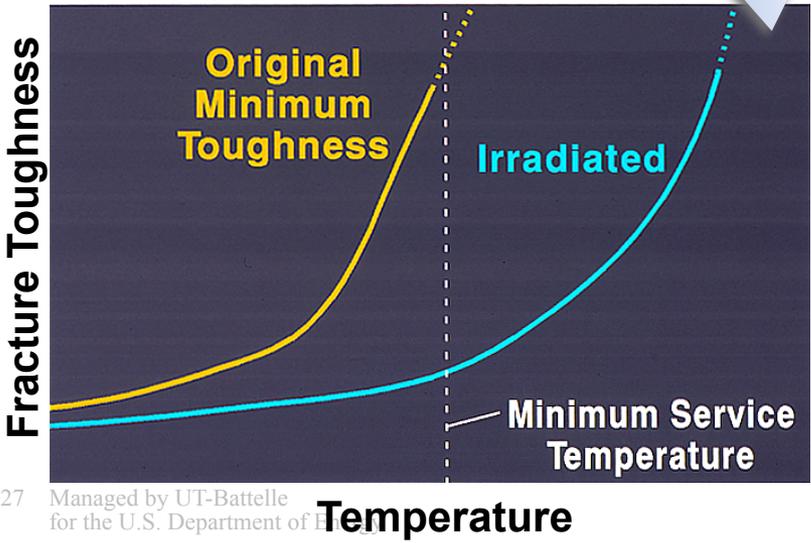
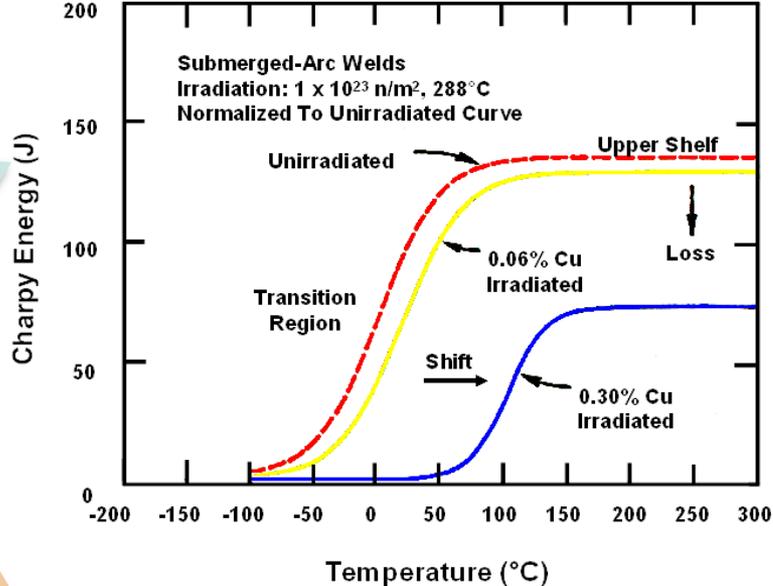
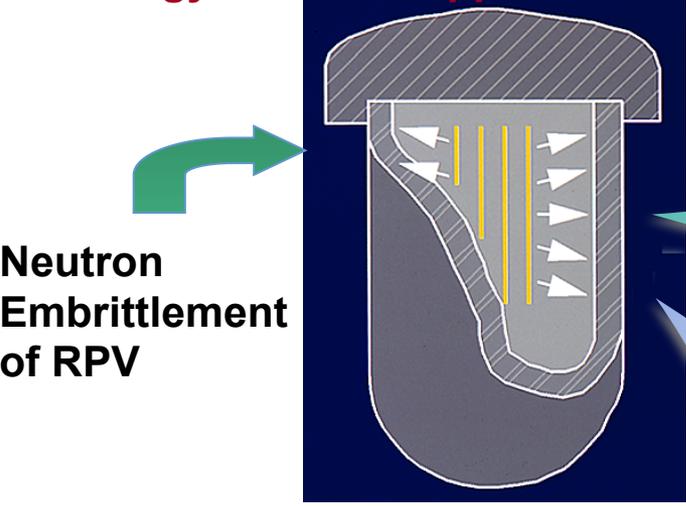
Radiation hardening



- On a macroscopic scale, this hardening is also observed.
- Increases in YS and UTS are commonly observed.
- Irradiation also results in a drop in ductility.

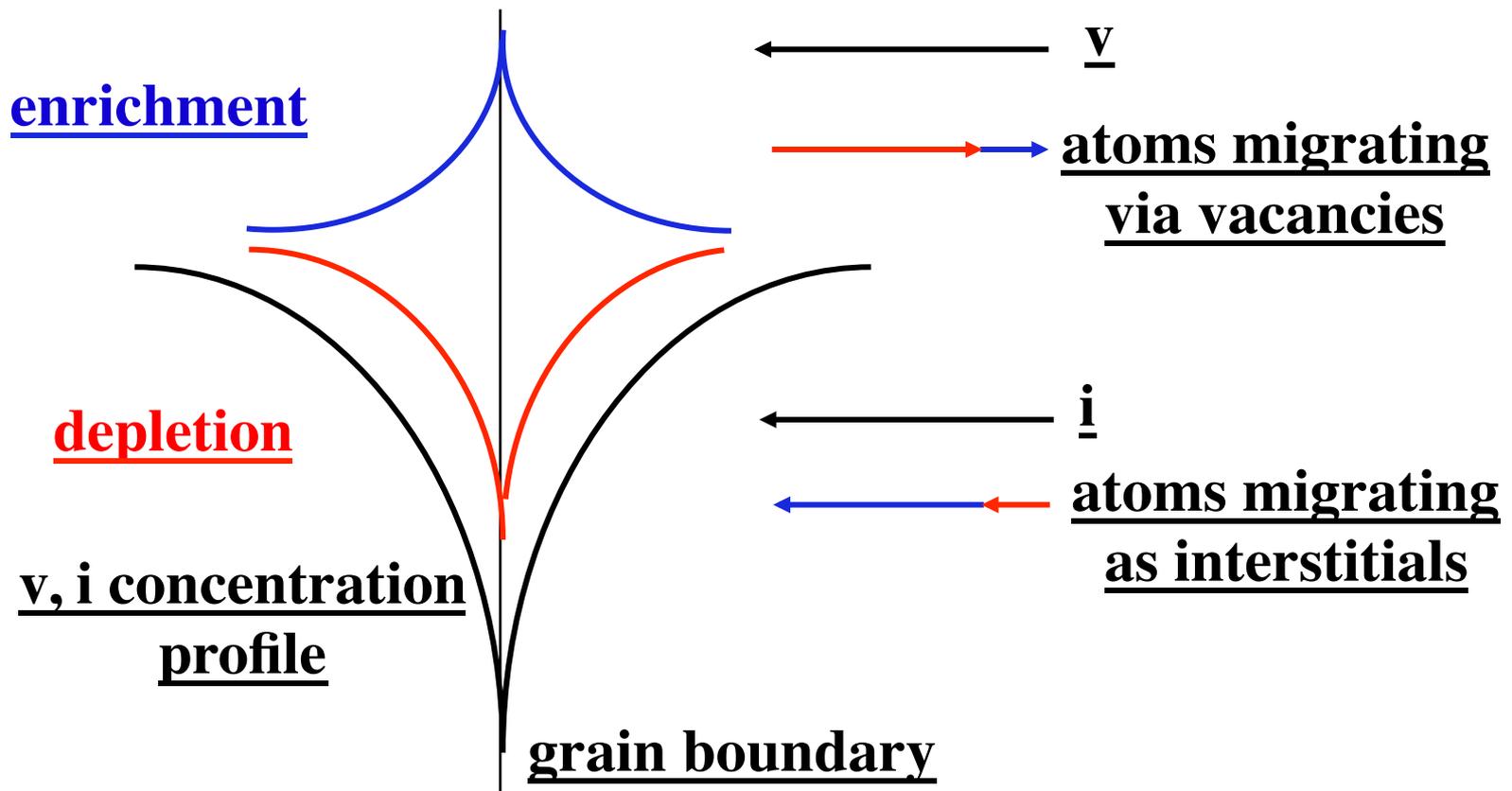
Reactor Vessel Integrity Assessments Must Account for Potential Degrading Effects of Neutron Irradiation

Irradiation Causes Ductile/Brittle Transition Temperature Shift and Upper Shelf Energy Loss — Copper Increases The Effect



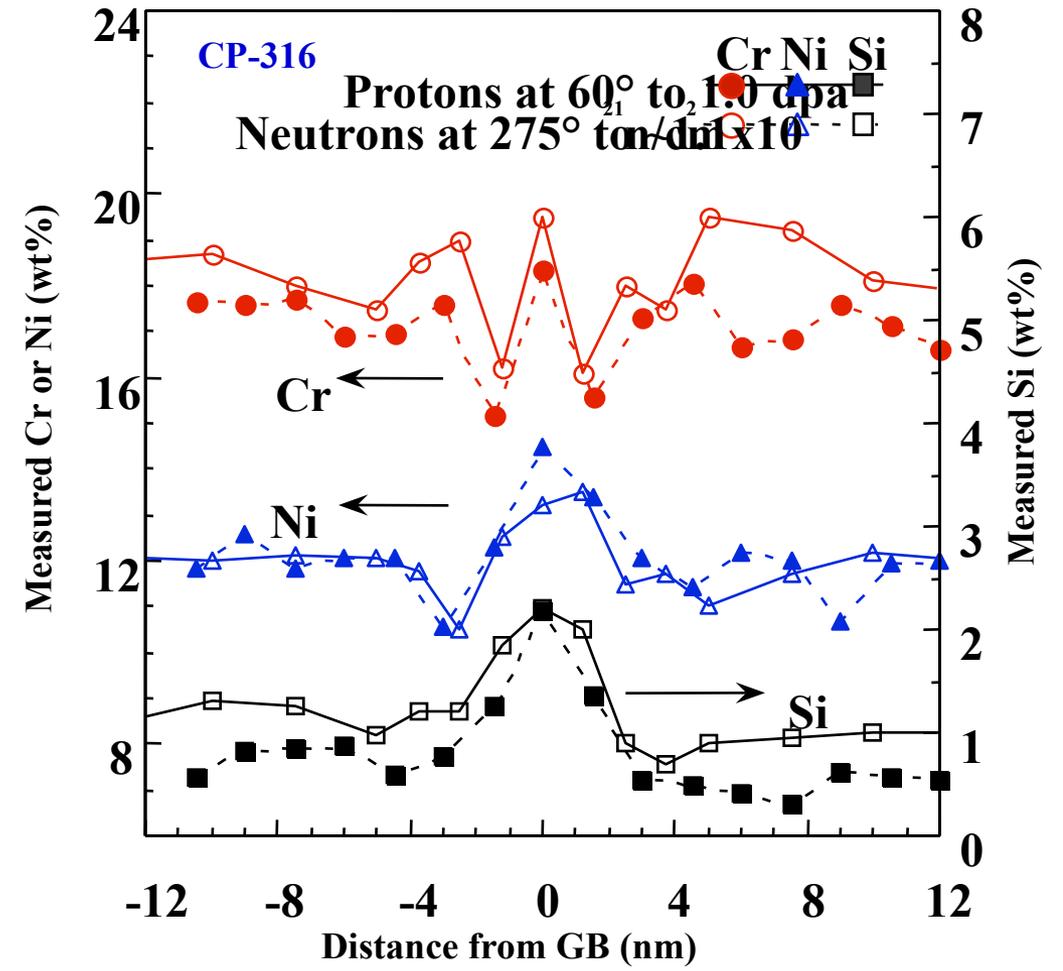
Irradiated Microstructures: Precipitates and Matrix Damage

Radiation-Induced Segregation

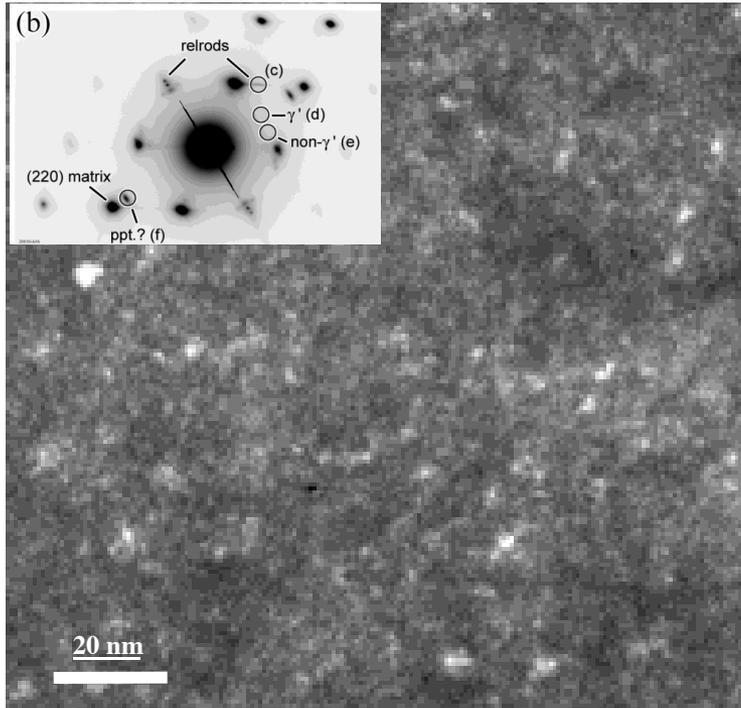


- High concentrations of radiation-induced defects will migrate to defect sinks.
- Any preferential association between an atom and one type of defect will result in segregation.

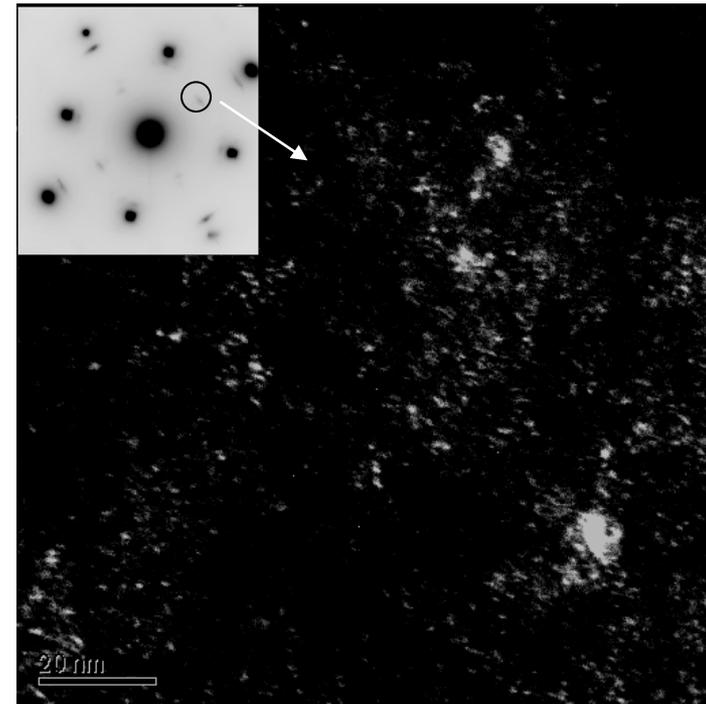
RIS comparison for proton- and neutron-irradiated 316 SS after 1.0 dpa



Comparison of γ' in proton- and neutron-irradiated SS



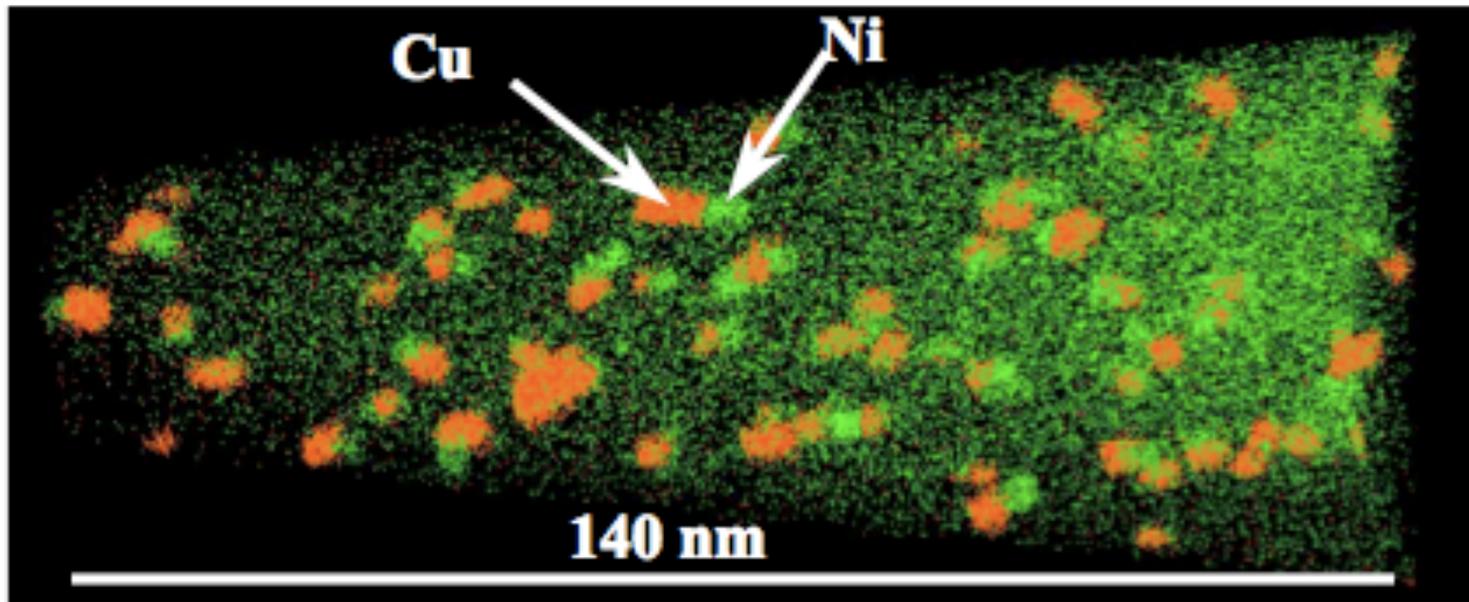
Tihange baffle bolt:
neutron-irradiated to ~7
dpa at 299°C*.



304+Si proton-irradiated
to 5.5 dpa at 360°C.

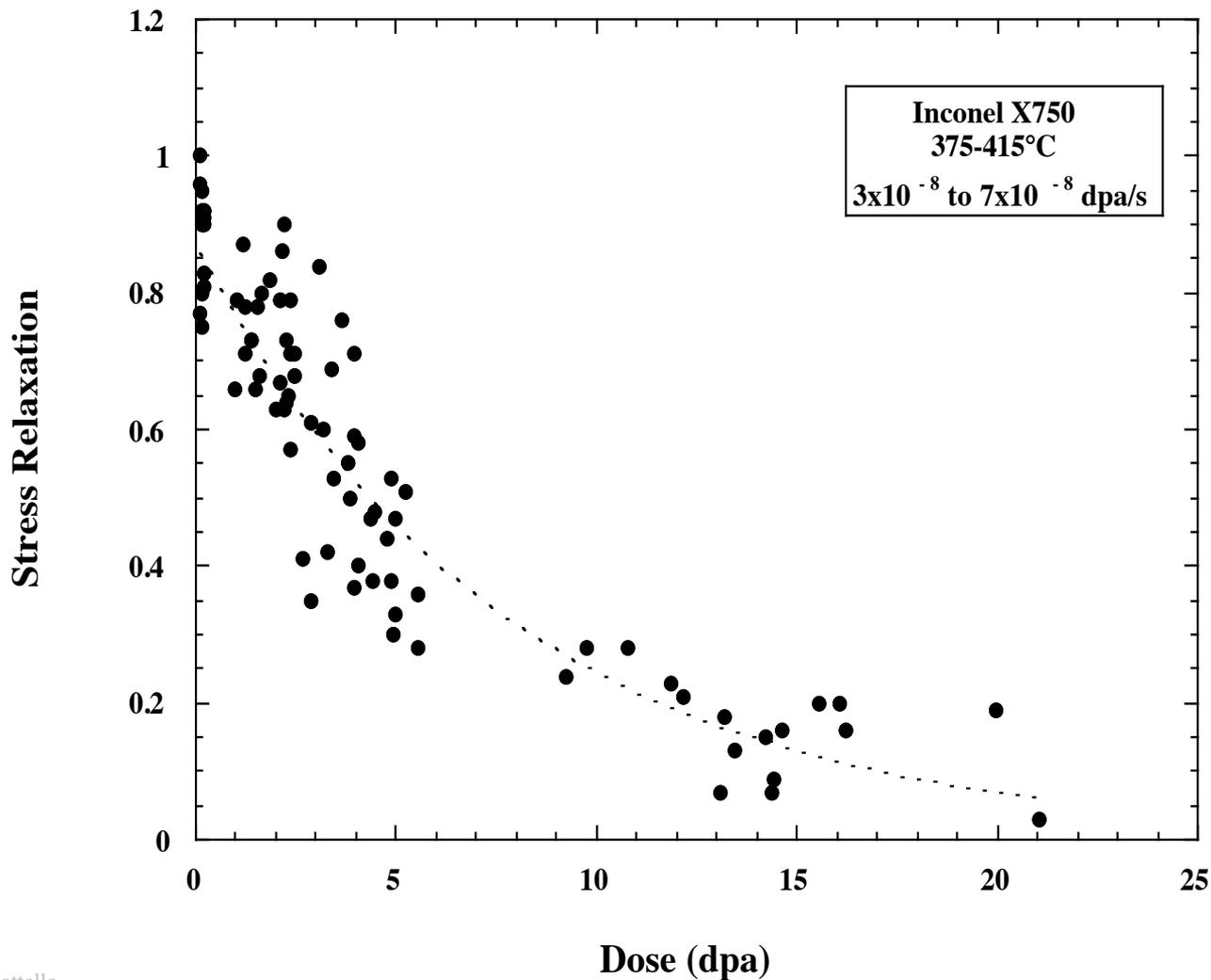
Advances in analytical techniques have allowed for more rapid and detailed analysis of materials

3-D APT of Ion-irradiated HCM12A (7.0 dpa at 400°C)



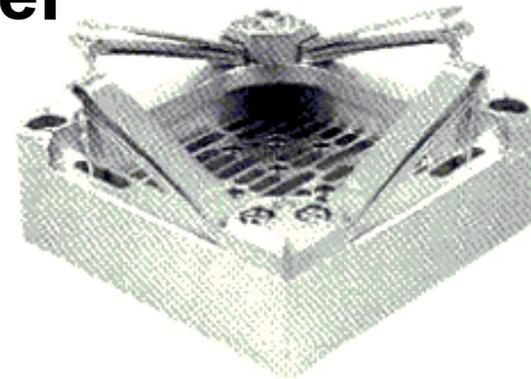
Radiation-induced Stress Relaxation

Stress Relaxation Approaches ~10% of pre-load

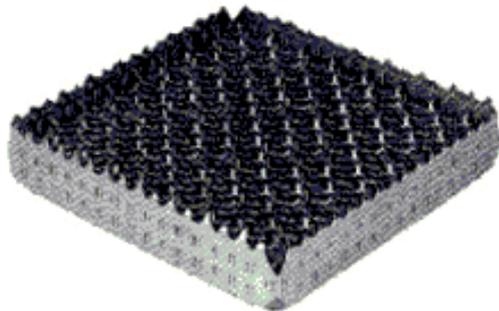


Stress-relaxation is an important factor for a number of LWR core internals

Spring components on fuel assemblies relax during service.



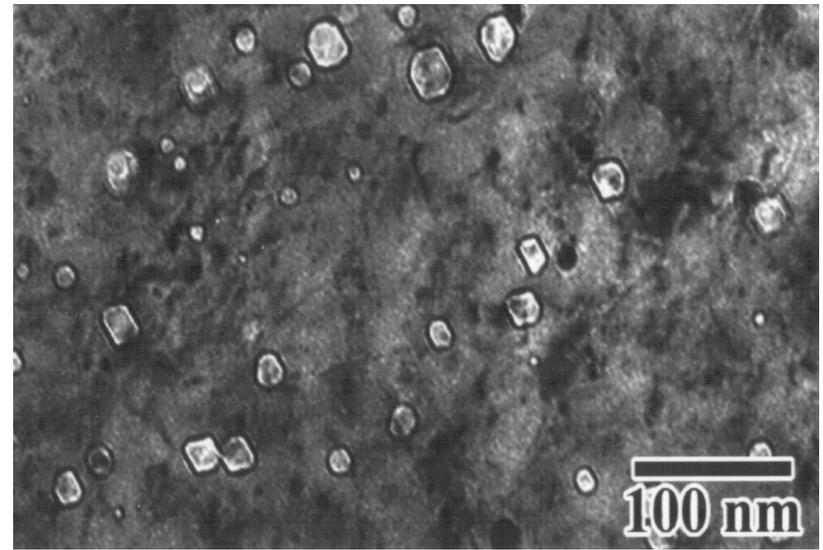
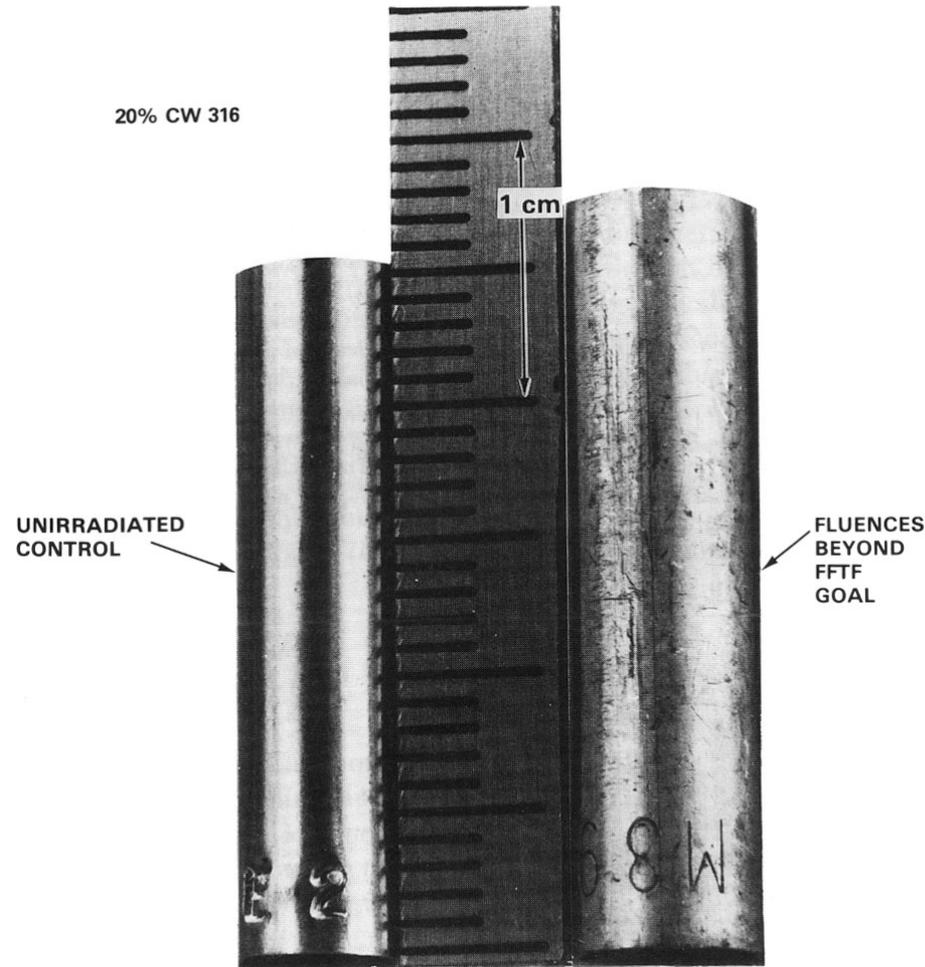
○ Top Nozzle



○ Grid Assembly

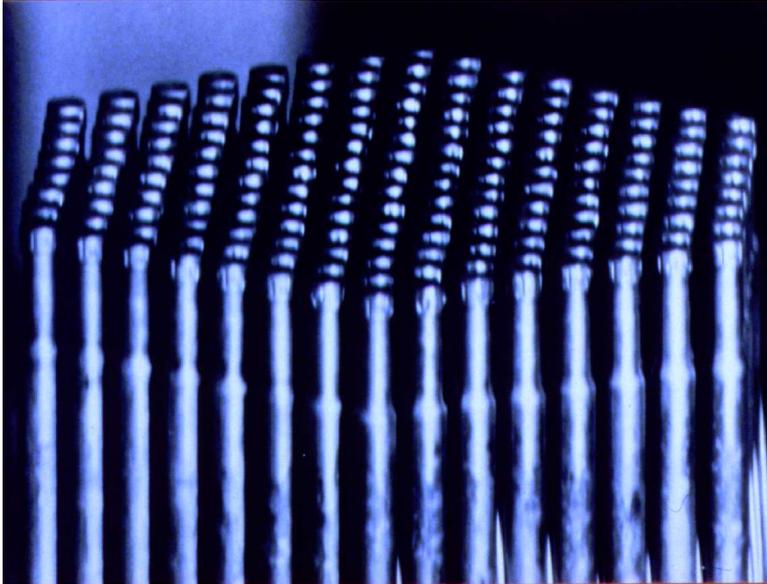
Easily Observed Swelling

- **Swelling: Volume increase in a material caused by void formation and growth**

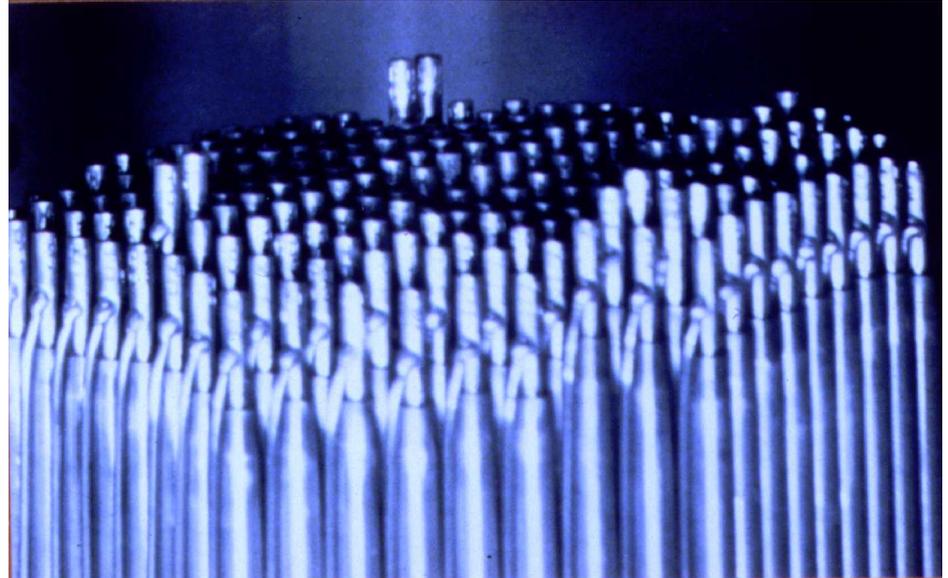


Easily Observed Swelling

FFTF Fuel Pin Bundles

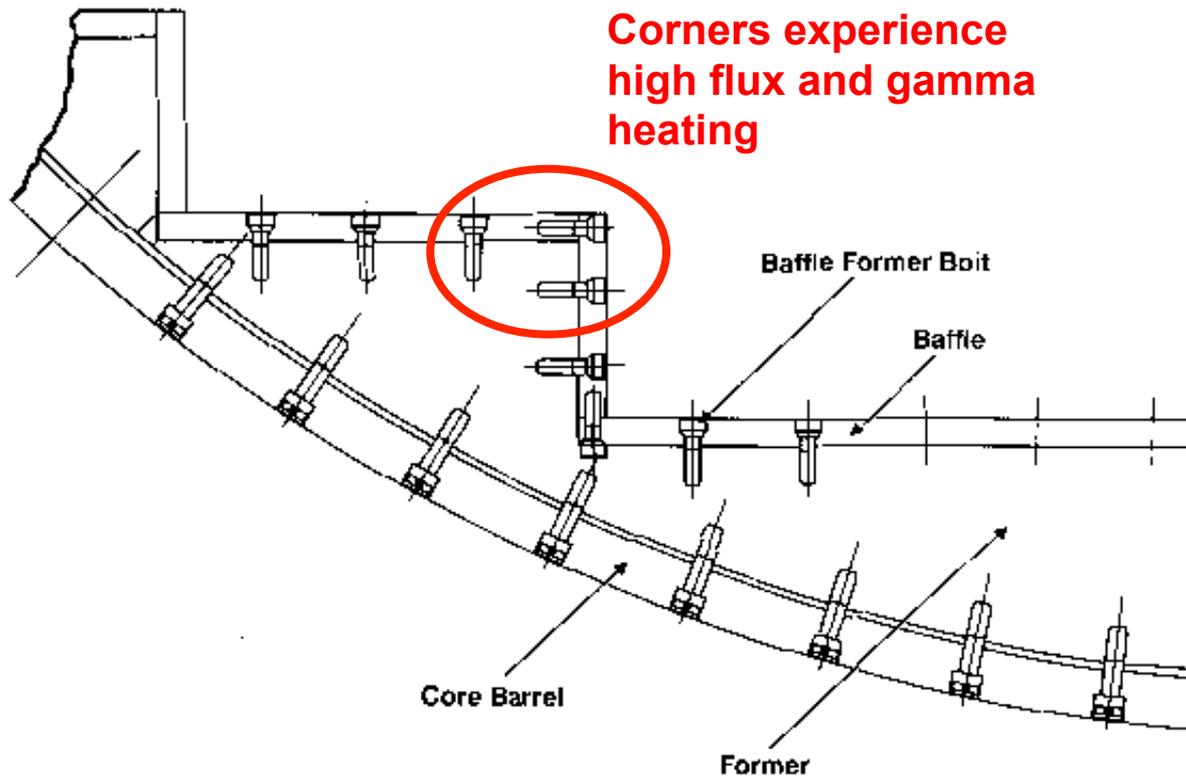


HT-9, no swelling

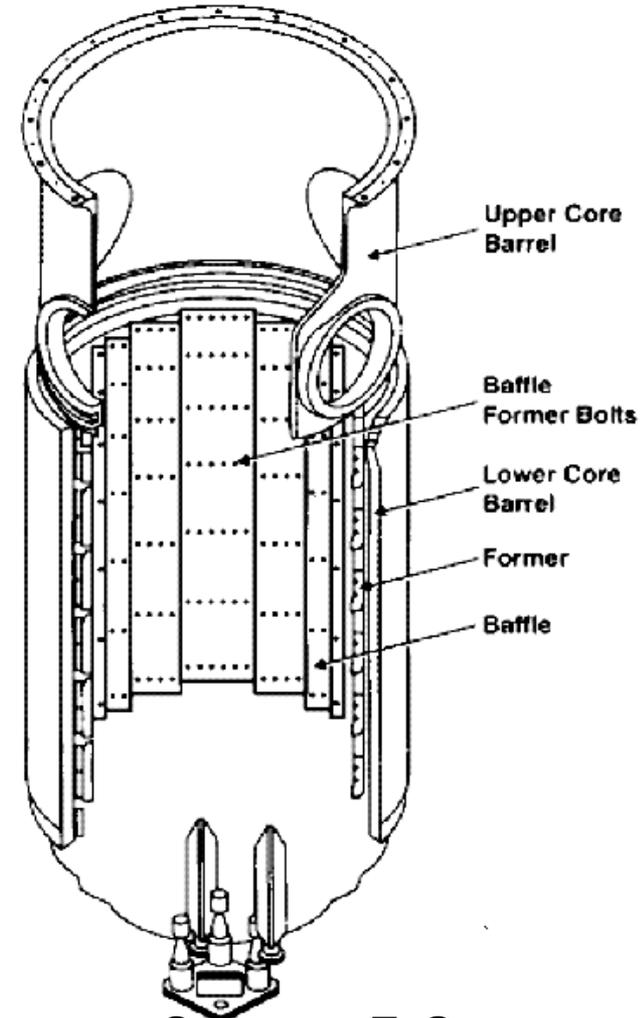


316-Ti stainless, swelling

Baffle bolts experience some of the highest fluences and temperatures in a PWR core



Corners experience high flux and gamma heating



A number of common transmutation reactions in reactors can influence irradiation performance

- A number of important reactions occur in reactor environments, varying with spectrum and materials
- Most create helium
 - $^{58}\text{Ni} + n_f \rightarrow ^{55}\text{Fe} + ^4\text{He}$
 - $^{60}\text{Ni} + n_f \rightarrow ^{57}\text{Fe} + ^4\text{He}$
 - $^{58}\text{Ni} + n \rightarrow ^{59}\text{Ni} + \gamma \rightarrow ^{56}\text{Fe} + ^4\text{He}$
 - $^{10}\text{B} + n \rightarrow ^7\text{Li} + ^4\text{He}$
- He production is of interest due to implications on embrittlement in fast reactors.

Helium Embrittlement for fast reactors

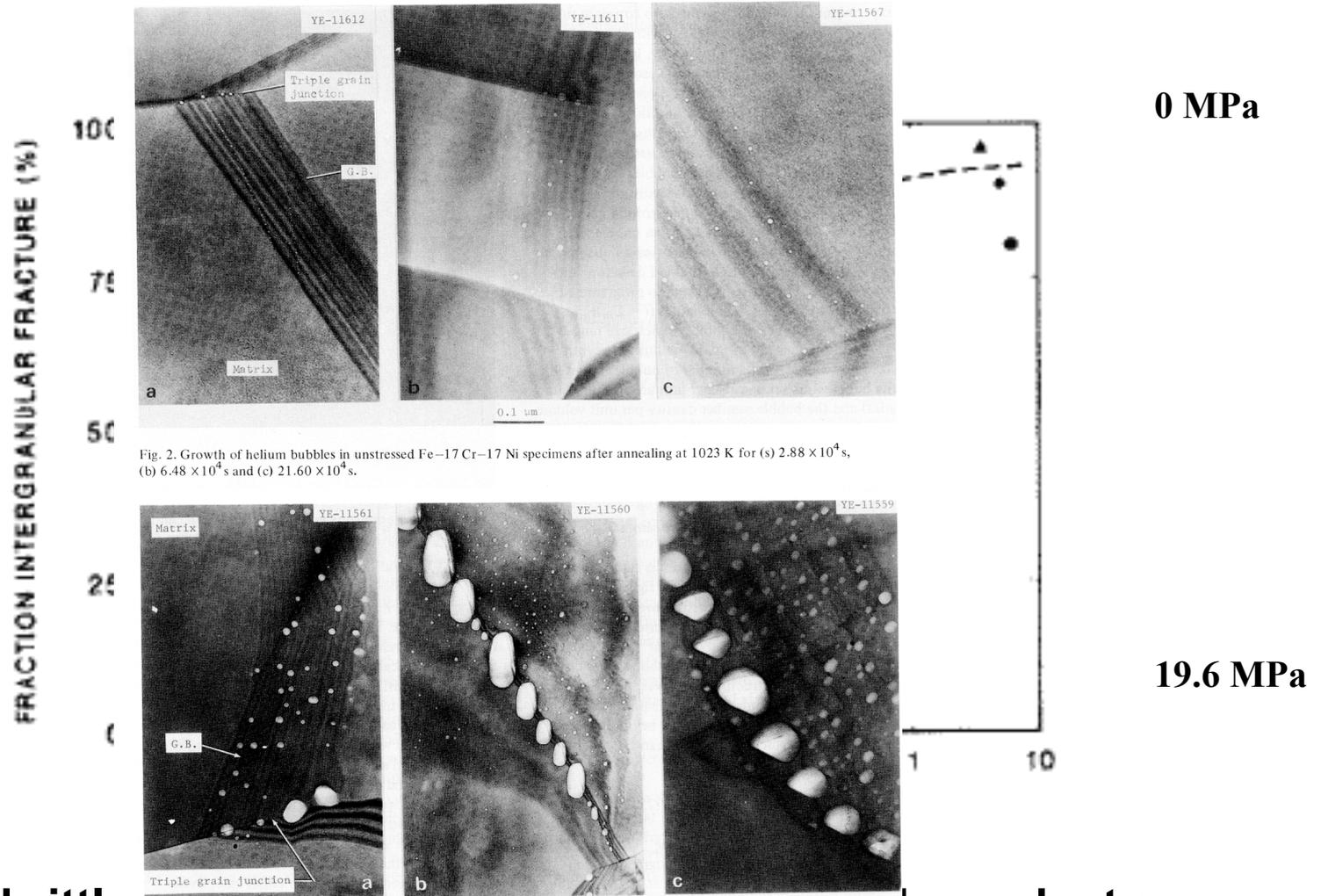


Fig. 2. Growth of helium bubbles in unstressed Fe-17Cr-17 Ni specimens after annealing at 1023 K for (a) 2.88×10^4 s, (b) 6.48×10^4 s and (c) 21.60×10^4 s.

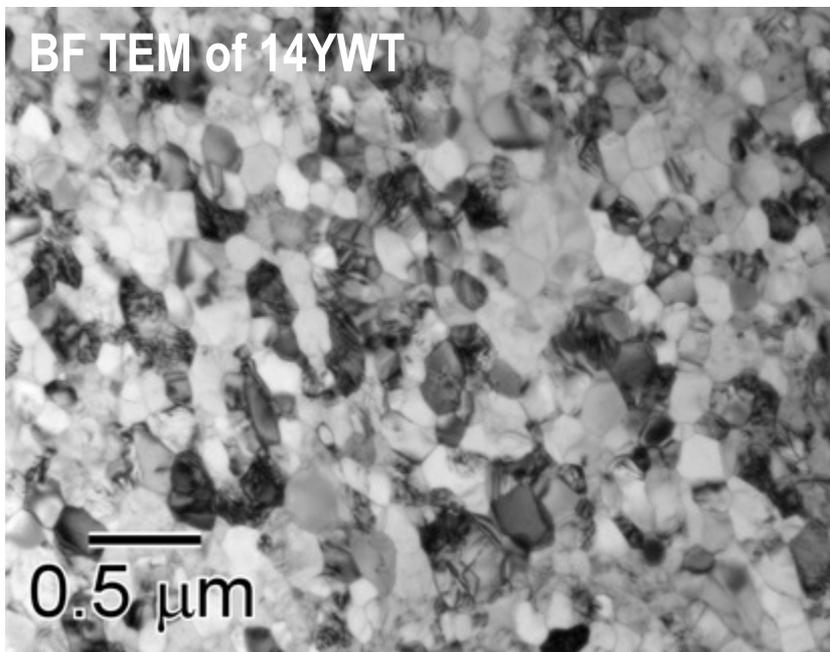
Embrittlement via intergranular fracture is dependent on helium content, temperature, and strain rate

van der Schaaf and Marshall, 1983

Nanostructured ferritic alloys are advanced ODS steels for high fluence applications

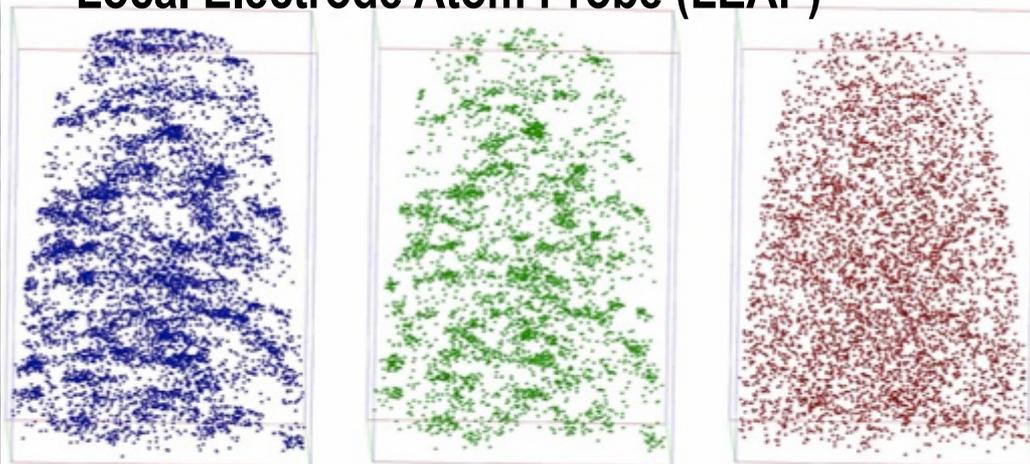
NFA contain high density of Ti-, Y-, and O-enriched nanoclusters

- NC discovered in 12YWT in 1999 (3D-APT at ORNL)
- NC observed in INCO MA957 in 2003 (3D-APT at ORNL and SANS)
- Neither are available...14YWT developed at ORNL early this century



Grain size = 136 (+/- 14) nm
Grain aspect ratio = ~ 1.2

Local Electrode Atom Probe (LEAP)



O
 $N_v = \sim 0.5 - 1 \times 10^{24} \text{ m}^{-3}$
 $r = \sim 1 - 2 \text{ nm}$

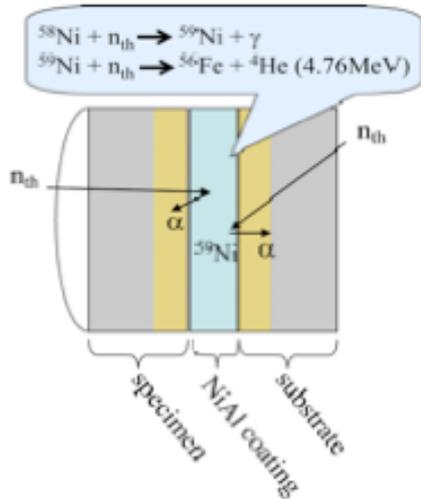
Ti = 43.9 +/- 6.7
Y = 6.9 +/- 5.8
O = 44.7 +/- 4.0

Balance = Fe, Cr

Source: D. Hoelzer

Source: D. Hoelzer

NFA alloys exhibit extreme radiation tolerance

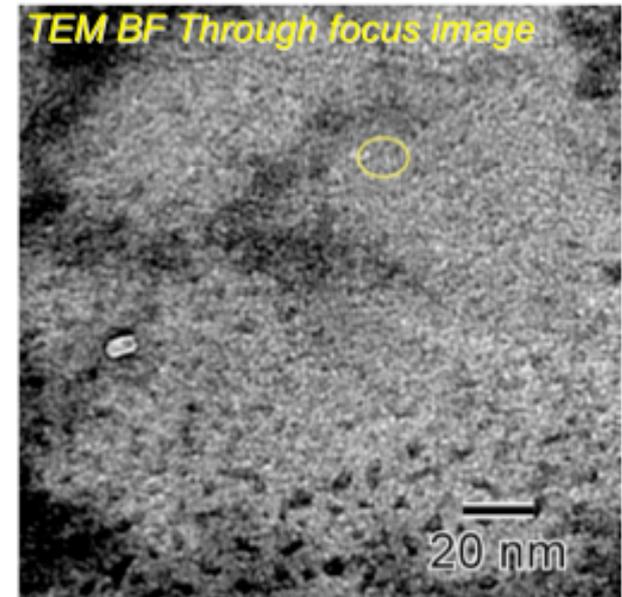
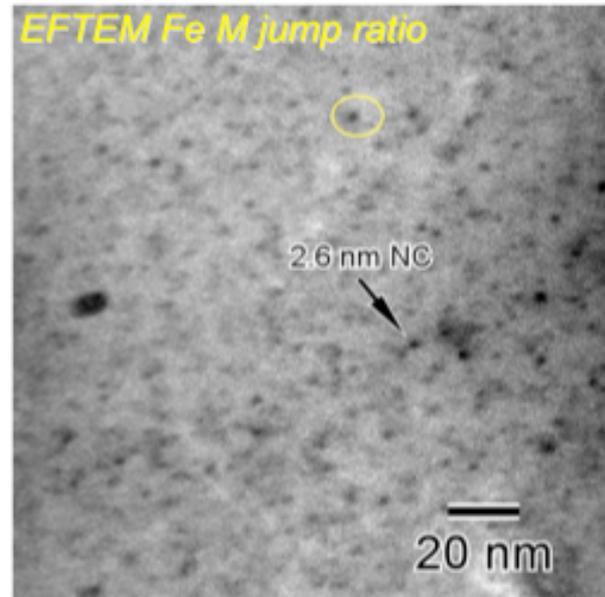


L.K. Mansur and W.A. Coghlan,
ASTM STP 1046, 1989

Lift-Out FIB
Specimen



- Simultaneous neutron and He implantation
- HFIR: 9 dpa and up to 380 ppm He at 500°C



J. Bentley et al., Microsc. Microanal., V13(Suppl 2), 2007, CD1072

T. Yamamoto et al., JNM, 367-370, 2007

- Ti-, Y-, and O-enriched NC are stable during irradiation
- He trapping - If cavities exist, they are too small (≤ 2 nm) to detect reliably using standard through focus imaging

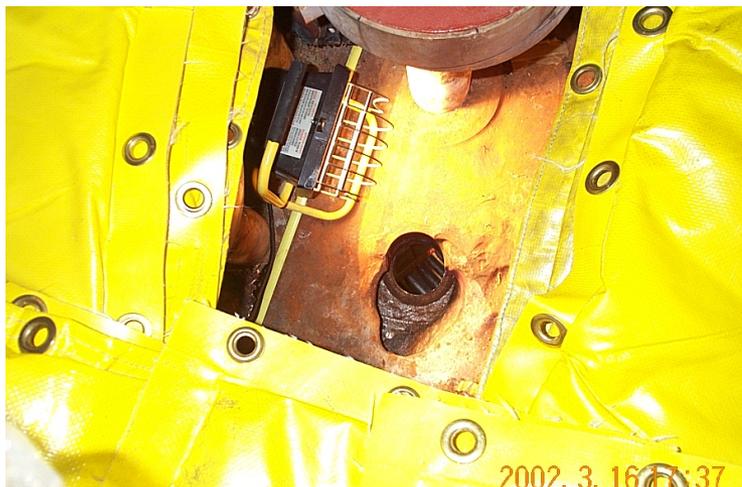
Corrosion also plays an important role in the economics of nuclear power

Ten most expensive costs of corrosion for Oconee units 1, 2, and 3 (PWR)

Work Activities	Cost	Cost, %	% attributed to corrosion	Cost of Corrosion
Steam Generators	\$22,757,765	8.26	95	\$21,619,877
Maint. Engg. Supp.	\$13,204,783	4.79	33	\$4,357,578
Radiation Protect.	\$12,116,142	4.40	80	\$9,692,912
Mechanical Comp.	\$10,709,285	3.89	33	\$3,534,064
Maint. Funct. Supp.	\$10,675,567	3.87	33	\$3,522,937
Work Control	\$6,073,111	2.20	33	\$2,004,127
Chemistry	\$5,570,659	2.02	60	\$3,342,395
Piping	\$2,391,285	0.87	60	\$1,434,771
Coatings and Paint	\$2,279,358	0.83	45	\$1,025,771
Decontamination	\$1,216,689	0.44	80	\$913,351
Other	\$188,590,607	68.43	9	\$17,122,624
TOTAL	\$275,585,251		25	\$68,896,313

Assessment of the Event

RPV Head Degradation- Nozzle 3

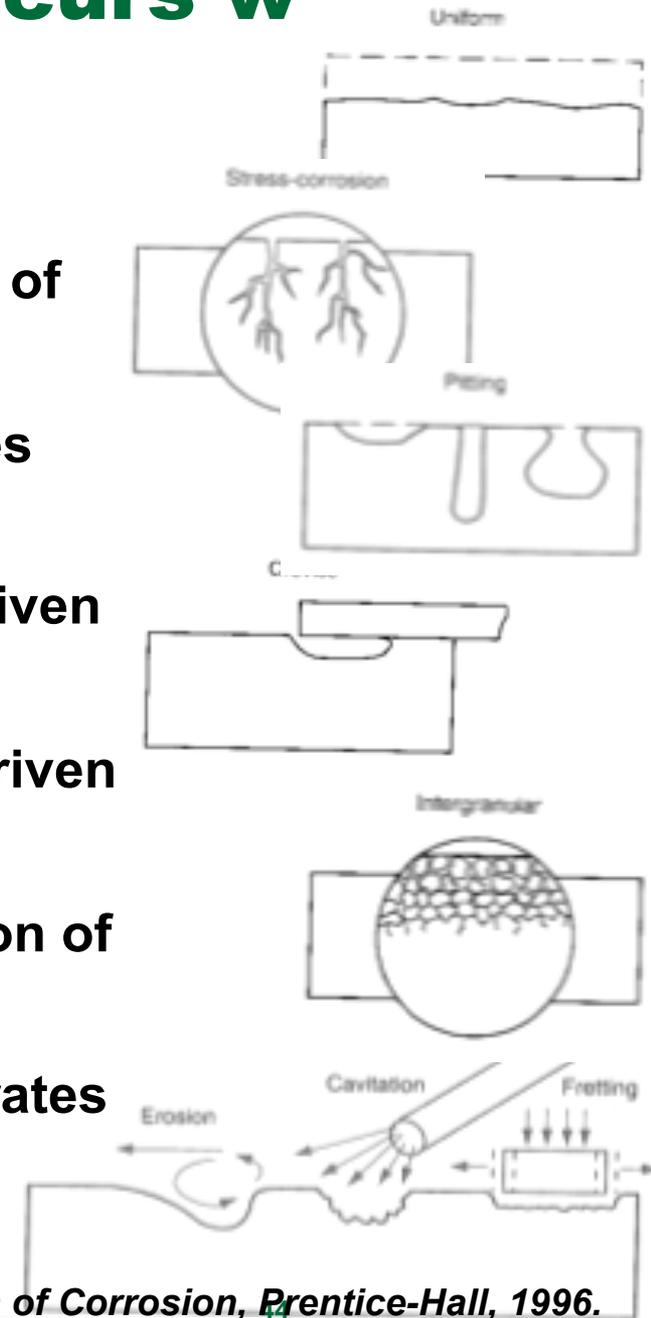


Today's LWR environments are continuously managed

	BWR-NWC	BWR-HWC	PWR
Coolant Temp (°C)	288	288	320
Coolant Press. (psig)	1020	1020	2420
pH (at 25°C)	6.0	6.0	7.0-7.2
Oxygen (ppb)	300-2000	<10	<5
Hydrogen (ppm)		0.4-3	3-5 (35 cc/kg)
ECP (mV _{SHE})	+150	<-230	-770
Conductivity (μS/cm)	<0.1	<0.1	20.5
B content (ppm)			1000
Li content (ppm)			2-3
SO ₄ ⁻ content (ppb)	< 3	< 3	< 3
Cl ⁻ content (ppb)	< 1	< 1	< 1

Corrosion of many forms occurs with nuclear power plants

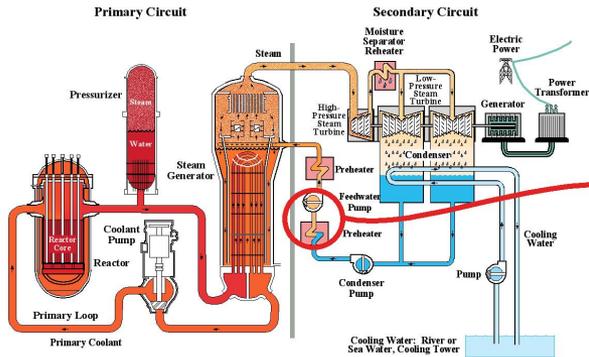
- **General corrosion**
- **Stress Corrosion Cracking:** combination of stress and environment
- **Pitting:** Localized corrosion driven by species and electrochemical differences
- **Crevice Corrosion:** Localized corrosion driven by species and electrochemical differences
- **Intergranular attack:** localized corrosion driven by material and microstructural differences
- **Erosion-Corrosion:** driven by a combination of factors
- **Flow-assisted corrosion:** driven by flow-rates and corrosion processes



Flow-accelerated corrosion caused the Mihama-3 incident

(a)

Schematic of PWR Plant



(c)

- 22 m/s, 0.93MPa, 142°C, 8.6-9.3pH
- 185,700 hr. operation

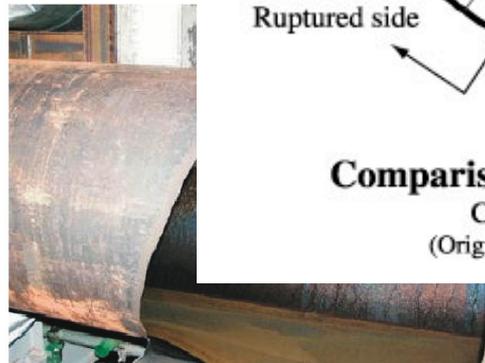
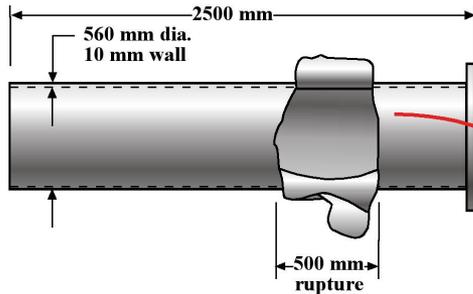
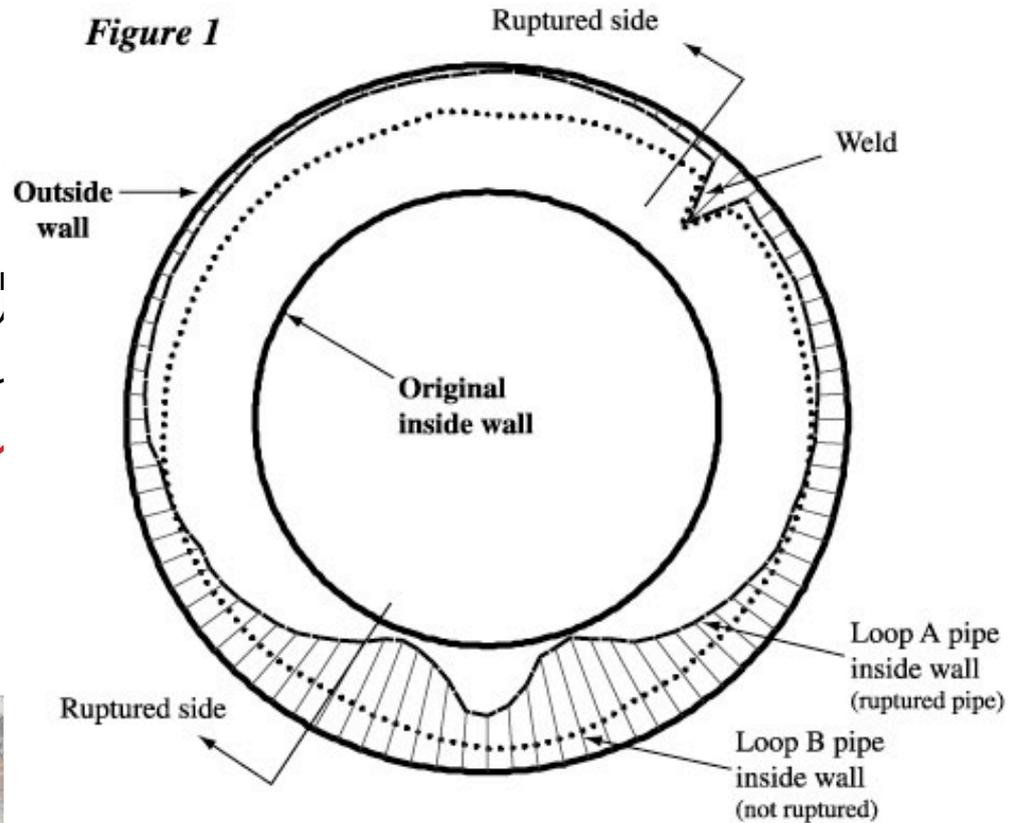


Figure 1

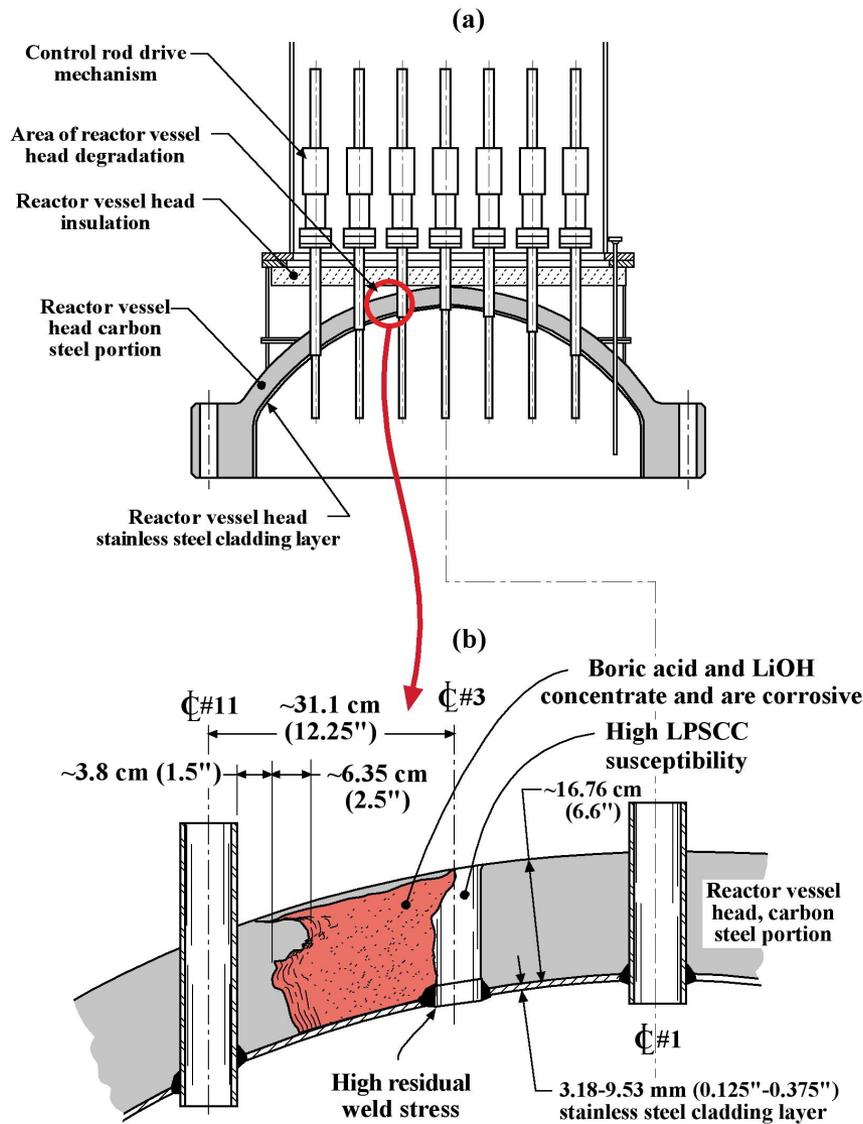


Comparison of thinning of A & B loop pipes

Cross-section viewed from upstream
(Original outer diameter 560mm, thickness 10mm)



SCC in one component can lead to other forms of corrosion



(c) Nozzle #3 with insulation removed and shielding installed 03-16-02



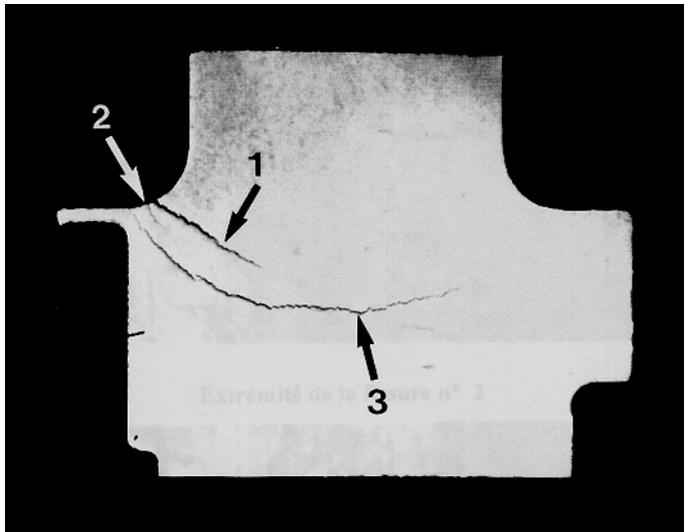
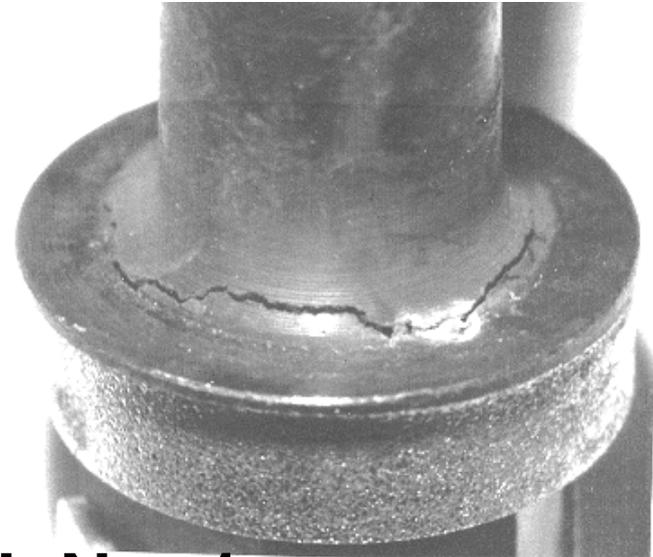
(d) Nozzle #3 cleaned



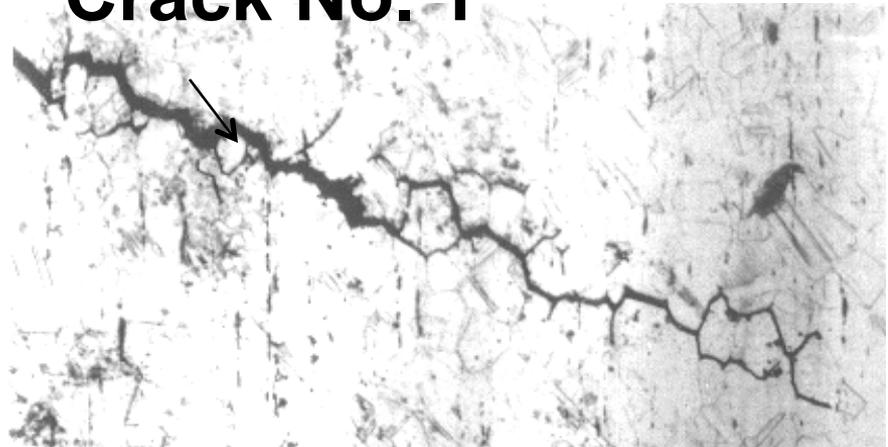
(e) LPSCC crack in cladding



IASCC: baffle former bolts in PWR



Crack No. 1

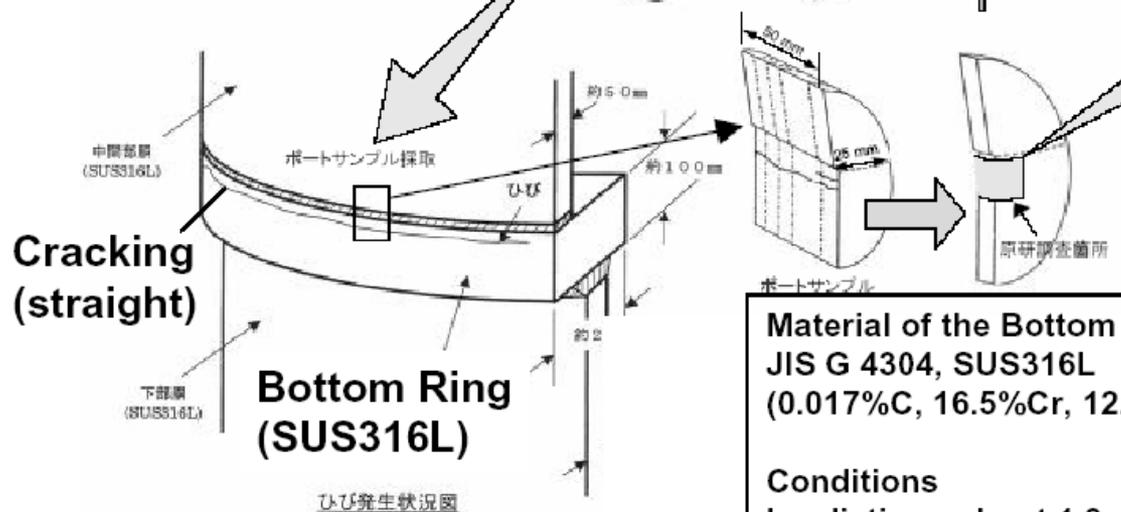
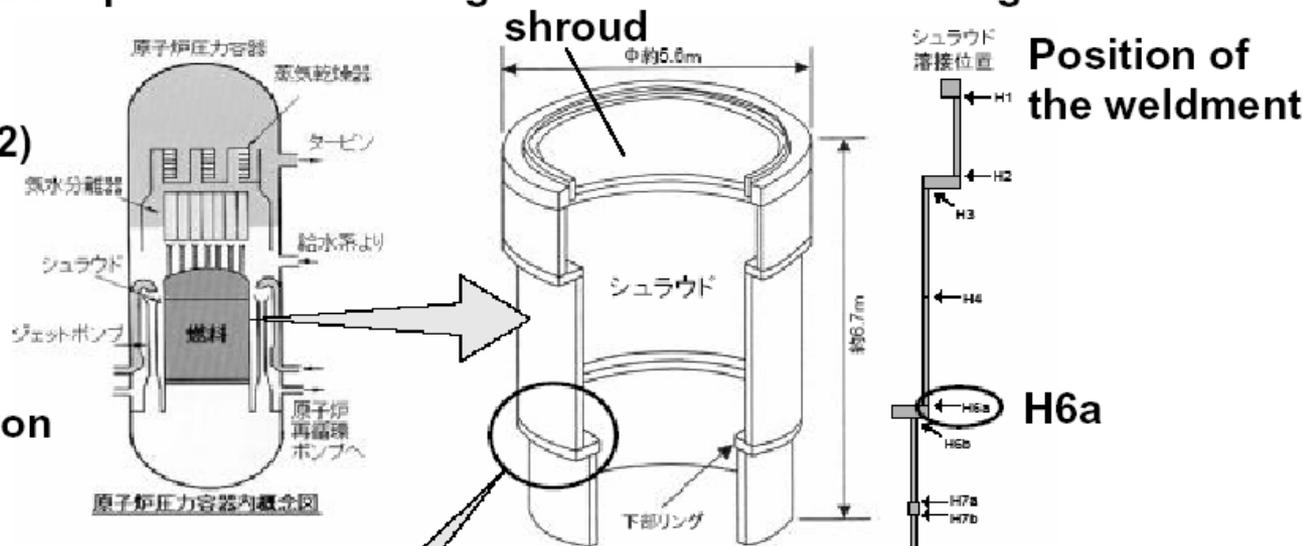


3. Research ~Examples of the Investigation Results for Cracking~

3.1 Shroud

(Unit3
-Fukushima2)

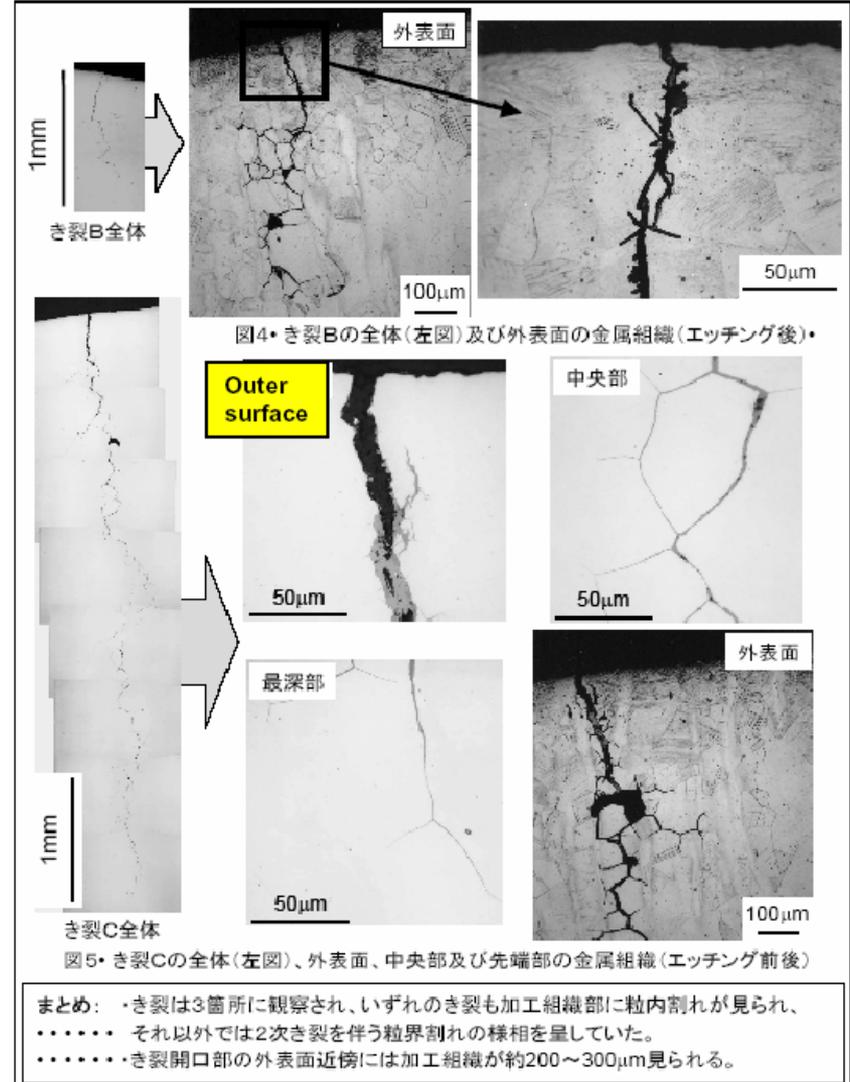
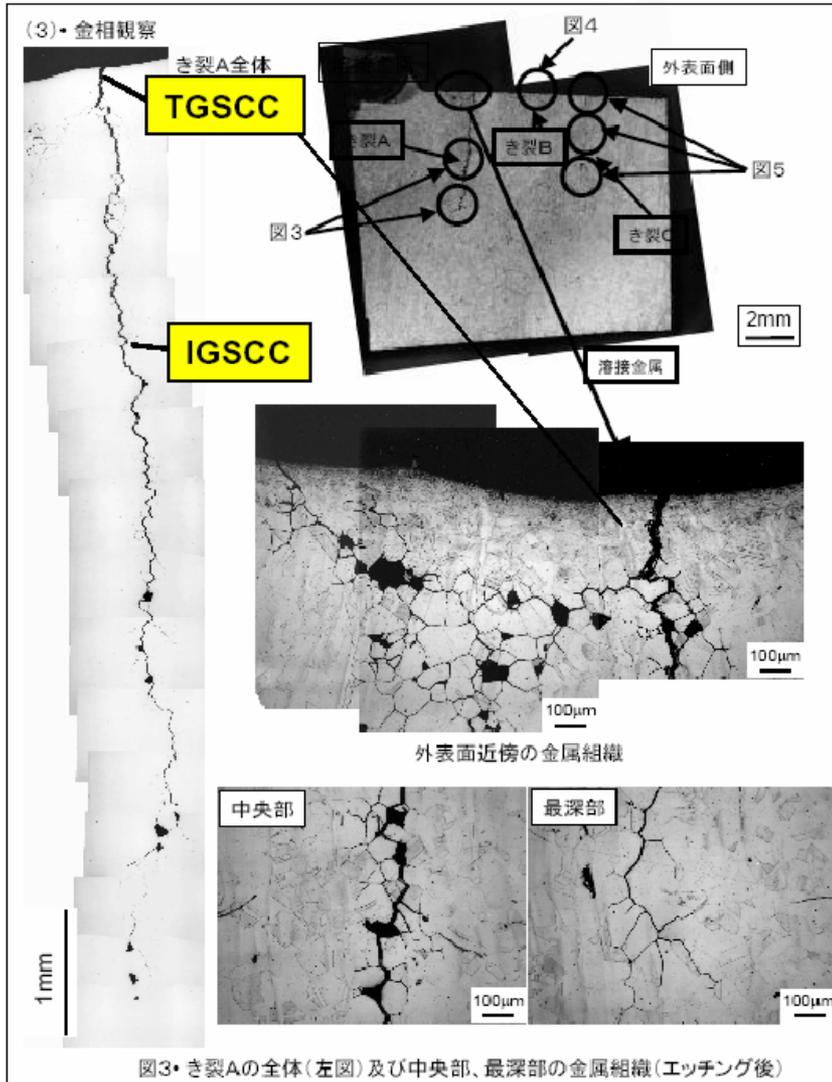
Cross section
of the RPV



Material of the Bottom Ring
 JIS G 4304, SUS316L
 (0.017%C, 16.5%Cr, 12.5%Ni, 2.2%Mo)

Conditions
 Irradiation : about 1.3×10^{22} n/m²(E>1MeV)
 Temperature : about 288 deg.
 Dissolved Oxygen : about 250 ppb

Cracking of the Shroud (2F-3) ~ Microscopic observation ~





Types of Liquid Metal Corrosion

- **Solution of solid metal(s) into LM**
- Diffusion of LM atoms into solid metal lattice
- Formation of intermetallic compound on surface of solid metals
- **Mass transfer**
- **Leaching of constituents from alloy surface due to selective solution**
- Subsurface precipitation of intermetallic compound caused by inward diffusion of LM atoms and interaction with alloy components
- **Impurity reactions**
- (No radiolysis and associated corrosion)

Concrete (Originally Based on Lime Hardened by Atmospheric Carbonation) has been Utilized as a Construction Material for Several Thousand Years



**Great Pyramid at Giza
(~2500 BC)**



**Colosseum
(82 AD)**



**Pantheon
(126 AD)**



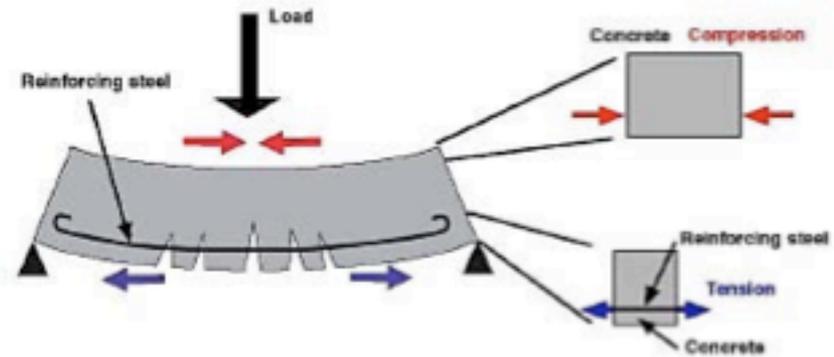
**Arch of Severus
(205 AD)**

- **Why did these structures survive?**
 - Careful materials selection
 - Mild climatic conditions
 - Lack of steel reinforcement
 - Construction method
- **These structures were not fabricated using current “hydraulic portland cement” (circa 1824)**

NPP Safety-Related Concrete Structures are Composed of Several Constituents that, In Concert, Perform Multiple Functions



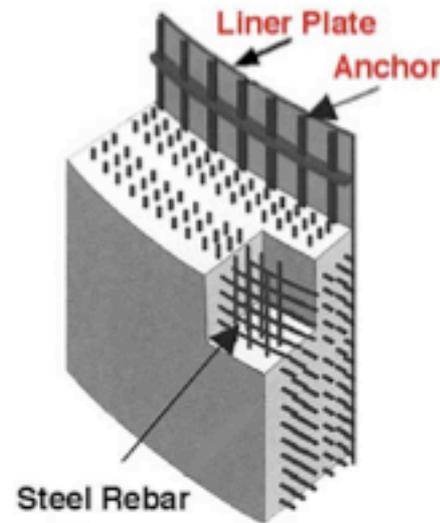
Concrete



Mild Steel Reinforcement



Post-tensioning tendons

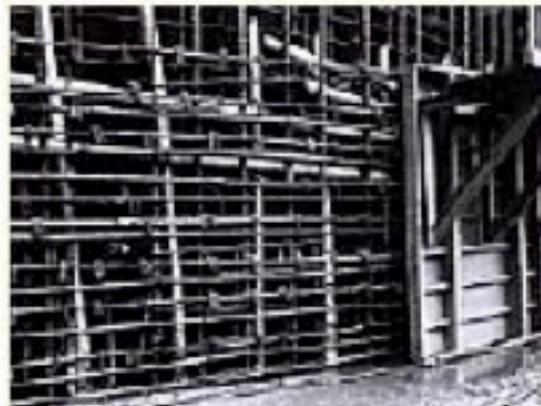


Steel Liner Plate

All NPPs Contain Concrete Structures Whose Performance and Function are Necessary to Protect the Safety of Plant Operating Personnel and General Public



Trojan NPP

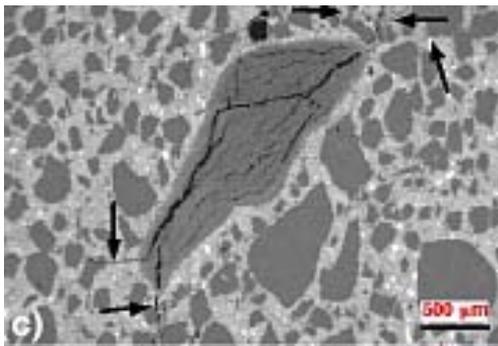


**Steel Reinforcement
In Wall Near Base**

- Concrete structures are essentially passive under normal operating conditions, but play a key role in mitigating the impact of extreme/abnormal operating and environmental events
- Structural components are somewhat plant specific, may be difficult to inspect, and usually can not be replaced
- Structures are subject to time-dependent changes that may impact their ability to withstand various demands from operation, the environment, and accident conditions
 - Excessive degradation can lead to failure
 - Failure often affects serviceability, not safety

Summary of Common Causes of Defects in Concrete Members

Unsuitable Materials	Improper Workmanship	Environmental Exposure	Structural
Aggregate unsound or reactive contaminated Cement wrong type manufacturing error contaminated Admixture wrong kind contaminated Water organic contaminants chemical contaminants dirty Reinforcement wrong kind incorrect size	Faulty Design Incorrect Concrete Mix low cement content high water content incorrect admixture dose batching errors High Slump Unsuitable Formwork/Shoring Misplaced Reinforcement Handling/Placing Concrete segregation careless placing inadequate or over vibration poor finishing Incomplete Curing	Concrete Chemical Attack efflorescence or leaching sulfates acids or bases delayed ettringite formation alkali-aggregate reactions Physical Attack salt crystallization freezing and thawing thermal exposure/thermal cycling abrasion/erosion/cavitation irradiation fatigue or vibration biological attack Steel Reinforcement carbonation, chlorides and stray currents	Loads Exceed Design Accident Settlement Earthquake



Cracking Due to Alkali-Silica Reaction



Carbonation



Chloride Ingress

Reinforcement Corrosion

Source: D. Naus

Crystal River Unit 3 Containment Delamination



Steam Generator Replacement Opening

- At liner – 23' 6" by 24' 9"
- At concrete opening – 25' 0" by 27' 0"



Summary

- **Material performance is essential to reactor performance, economics, and safety.**
- **A modern reactor design utilizes many different materials and material systems to achieve safe and reliable performance**
- **Material performance in these harsh environments is very complex and many different forms of degradation may occur (often together in synergistic fashions)**
- **New materials science techniques may also help understand degradation modes and develop new manufacturing and fabrication techniques.**
- ***Nanotechnology may be able to solve some of these issues...but***

The incorporation of advanced alloys is not a trivial task!

- **Many factors are important for reactor service.**
 - Availability
 - Cost
 - Fabrication/Processing
 - Reproducibility/Uniformity
 - Irradiation resistance
 - Mechanical performance
 - Creep performance
 - Corrosion performance
 - Thermal properties
 - Joining
 - Fatigue
 - Fracture toughness
- **Any new alloy must also meet all regulatory (or code qualification) standards.**

The DOE-NE NEET program currently has open calls for advanced materials and manufacturing

- **Under this competitive process, materials are sought that provide**
 - Improvement in mechanical performance by a factor of 5-10 over traditional materials
 - Increase in maximum operating temperature of greater than 200° C over an 80 year lifetime
 - Increased radiation tolerance to beyond 300 dpa
- **Materials that support multiple designs or missions would be favored over single-applications**
- **High-risk/reward and transformational concepts are appropriate for NEET.**
- **Evolutionary gains are appropriate for the individual programs.**
- **DE-FOA-0000426 (materials)**
- **DE-FOA-0000427 (manufacturing)**