

Nuclear Fuel Performance

NE-533
Spring 2023

Last Time

- Growth and creep are the major mechanisms for dimensional instability in zirconium alloy cladding
- Growth results from the clustering of interstitials on prismatic planes
- Creep occurs in three stages: (1) creep down from water pressure; (2) creep out from fuel column; (3) fuel column axial stress
- Under irradiation, zirconium experiences irradiation induced hardening due to interstitial loops
- Dislocation channels form that don't have loops, resulting in localized deformation

Notes

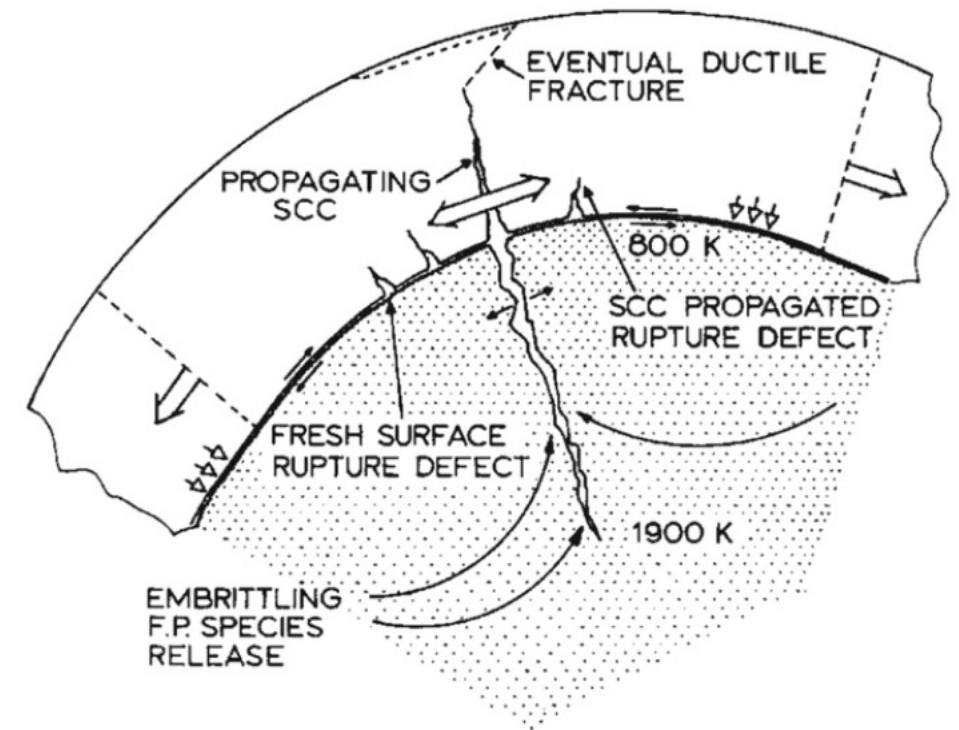
- MOOSE project part 2 due March 31
- Paper presentation on April 11
- Exam on section 3 March 30
 - covers all content through this lecture

Pellet Cladding Interaction (PCI)

- PCI includes both mechanical (PCMI) and chemical interaction (PCCI), often seen in a more general sense FCMI and FCCI
- The process of PCI involves the combination of high internal mechanical stress in the cladding and a corrosive environment resulting from the accumulation of volatile fission products in the fuel-clad gap
- These conditions may lead to the initiation and propagation of radial cracks, which can penetrate through the cladding
- Frequent PCI failures were observed in the 1960s and 1970s, leading to extensive research efforts
- Altering operational procedures, modifying the manufacturing process of the cladding, adding a composite layer and/or applying a protective coating to the inner cladding surface can help mitigate these failures

Physical Phenomena in PCI

- The fuel thermally expands and swells
- The cladding undergoes thermal and irradiation-induced creep
- Mechanical interaction between the fuel pellet and cladding generates radial compressive stresses in addition to tensile hoop stresses
- Volatile fission product gases are released into the gap
- The cracking of the fuel pellet together with non-uniform thermal expansion resulting in preferential contact at the pellet interfaces increases local mechanical stresses in the cladding

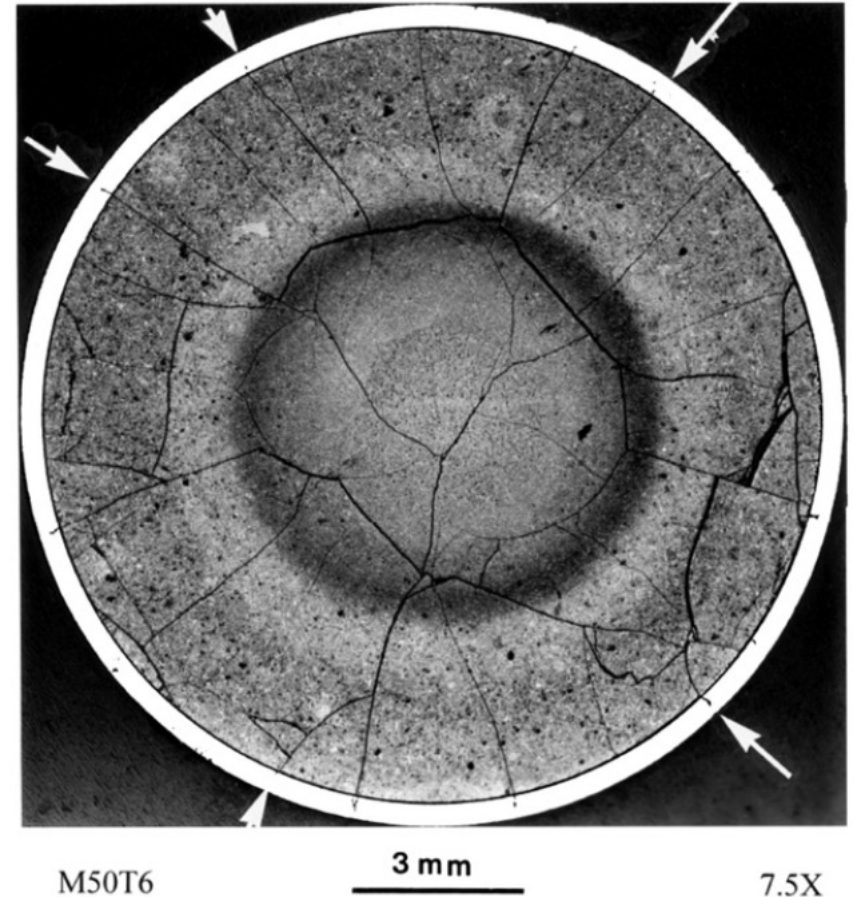


Stress Corrosion Cracking (SCC)

- Alloys that are in a corrosive environment may develop cracks that would not otherwise develop in the absence of either stress or the corrosive environment – stress corrosion cracking (SCC)
- Stress corrosion cracks may propagate along intergranular (IG) and/or transgranular (TG) paths
- The propagation modes of these cracks may depend on the localized mechanical and chemical conditions that lead to SCC
- In order for SCC to initiate and propagate in any material, four conditions are simultaneously required:
 - A corrosive environment, a susceptible material, sufficient stress, and sufficient time

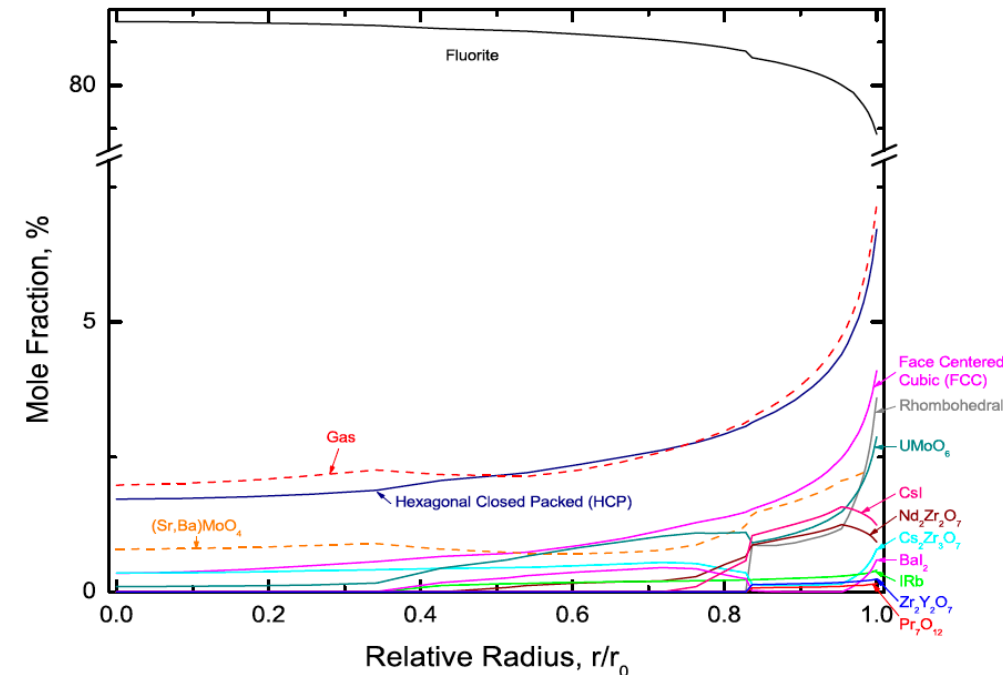
Corrosive Environment

- Chemically aggressive fission products accumulating in the fuel-clad gap form an important component of the corrosive environment
- Primary corrosive species are the volatile fission products, such as iodine (I), cadmium (Cd) and cesium (Cs)
- Corrosive species can diffuse down the temperature gradient through fuel cracks
- A higher local concentration of fission gases exists near the fuel crack and SCC is more likely to occur in the adjacent cladding region



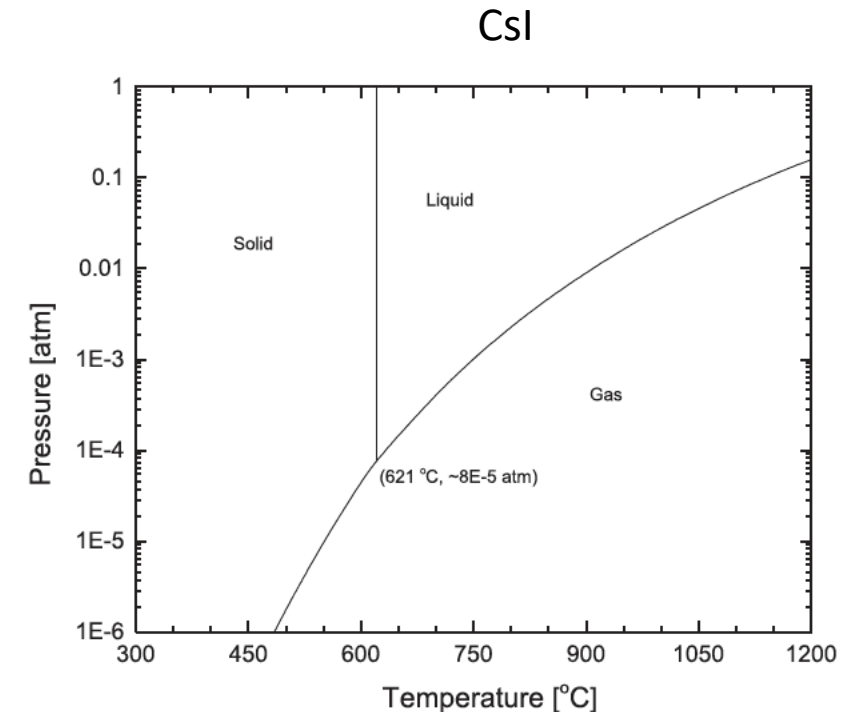
Corrosive Environment

- The ability of fission product atoms to be transported through the fuel to the fuel-clad gap depends on several factors, including its physical state
- The mobility of fission gases is markedly different than solid state diffusion
- The relative amounts of I and other chemical elements in the gaseous phase depend on the local fuel chemistry, which varies spatially
- The figure illustrates the predicted effects of fuel chemistry, in particular the predicted spatial distribution of phases in highly irradiated PWR UO₂ fuel, whereby the chemical behavior on the fuel surface is of particular interest



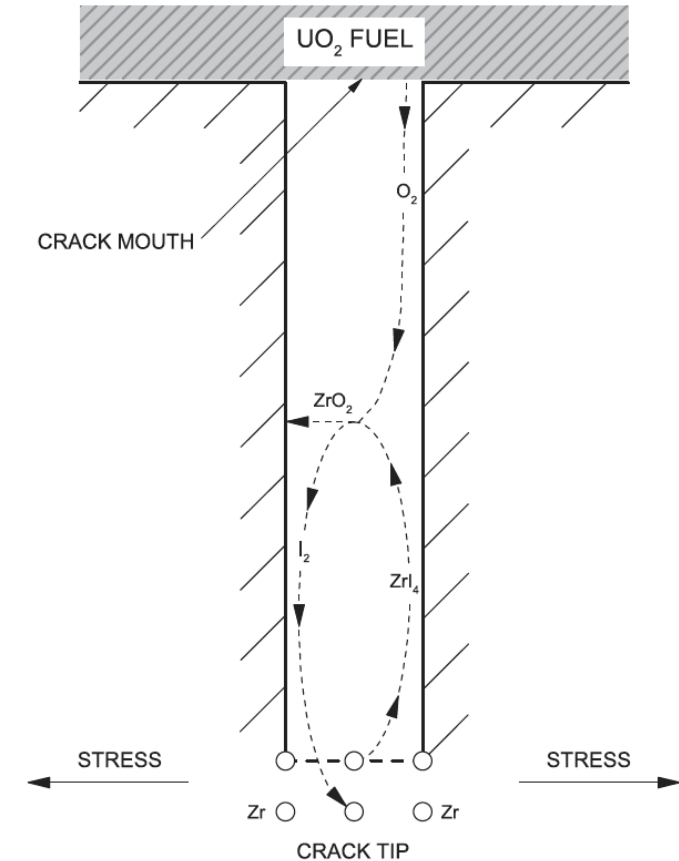
Corrosive Environment

- The dominant I containing solid phase that is predicted to be thermodynamically stable on the fuel surface is CsI
- Since typical fuel surface temperatures during normal operating conditions are of the order of 350–450 C and the hydrostatic pressure is greater than 1 atm, one would expect that solid CsI would be stable in the fuel-clad gap
- The radiolytic decomposition of CsI increases the iodine partial pressure in the gap by many orders of magnitude



Corrosive Environment

- The chemical interaction of liberated I (and possibly other elements) with the cladding is of great importance in crack initiation
- A zirconium iodide gaseous species migrates up the temperature gradient towards the crack mouth, whereby the high affinity of Zr for O may result in ZrO_2 formation, and decomposition may once again liberate I_2 gas
- The deposition of ZrO_2 on the crack walls is believed to create a passivation layer, which further localizes I encroachment at the crack tip



Susceptible Material

- The susceptibility of the cladding to SCC can be influenced by many factors, including the composition, microstructure, texture, and irradiation damage of the cladding, and the presence of an oxide passivation layer, which protects the metal from chemical attack
- All zirconium alloy cladding materials used in commercial power reactors are prone to PCI failure
- Minor compositional changes have been shown to offer slightly different performance characteristics

<i>Alloy</i>	<i>Sn (wt%)</i>	<i>Fe (wt%)</i>	<i>Cr (wt%)</i>	<i>Ni (wt%)</i>	<i>O (wt%)</i>	<i>Nb (wt%)</i>	<i>Structure</i>	<i>Reactor utilization</i>
Zircaloy-2	1.2–1.7	0.07–0.2	0.05–0.15	0.03–0.08	0.09–0.16	–	RXA	BWR
Zircaloy-4	1.2–1.7	0.18–0.24	0.07–0.13	–	0.09–0.16	–	CWSR, RXA	PHWR & PWR
ZIRLO	0.80–1.1	0.10	–	–	0.105–0.145	0.99–1.01	CWSR	PWR
OPT ZIRLO	0.66	0.11	–	–	0.105–0.145	1.04	PRXA	PWR
M5	–	0.03–0.05	0.015	–	0.118–0.148	1.0	RXA	PWR
E110	–	–	–	–	0.10	~1.0	RXA	PWR, RBMK & VVER

Susceptible Material

- The initial motivation of alloying zirconium with small amounts of tin was to offset the loss of corrosion resistance resulting from the introduction of nitrogen impurities during fabrication
- The control of impurities during manufacturing have significantly improved since the introduction of these alloys, making the addition of unnecessary
- The addition of niobium to these zirconium alloys increases the strength of the cladding while providing higher irradiation creep resistance and has exhibited elevated corrosion resistance, which is desirable for higher burnup fuel
- All Zr alloys are somewhat equally susceptible to SCC cracking following prolonged irradiation

Sufficient Stress

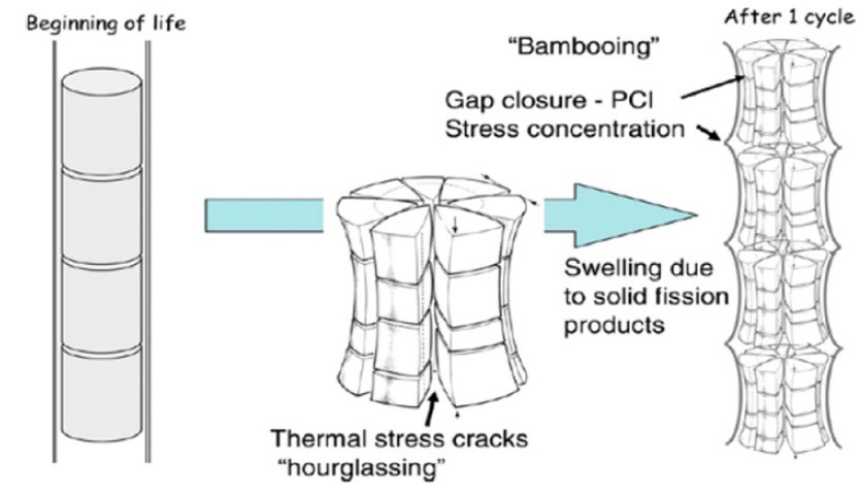
- The stress on the cladding depends on the external coolant pressure and creep, in addition to the stress imposed internally by the fuel
- The fuel pellet structurally deforms as a result of the following mechanisms: thermal expansion, solid and gaseous fission product swelling, thermal and irradiation-induced creep, irradiation-induced densification and cracking
- The fuel thermally expands almost immediately in response to an increase in temperature, whereas the contributions of creep and fission product swelling are longer term and depend on burnup
- UO₂ fuel is typically fabricated with an initial porosity of 3%–5% to accommodate fission products
- The benefits of this with respect to minimizing SCC are twofold: first, the effect of swelling is diminished by solid fission products filling internal voids; second, initial pores provide sinks for fission gases, thus impeding their release to the fuel-clad gap

Reducing Internal Pressure

- The initial grain size of the fuel, which evolves with burnup, affects fission gas release, among other factors
- Since intragranular fission gas diffusion occurs at a much slower rate than intergranular diffusion, larger grain sizes impede the overall release of fission gases to the fuel surface
- Reducing fission gas release with large grained fuel is less effective with increasing linear powers from 50-65 kW/m
- As an undesired consequence to improved fission gas retention with large grained fuel, fission product swelling can be more pronounced

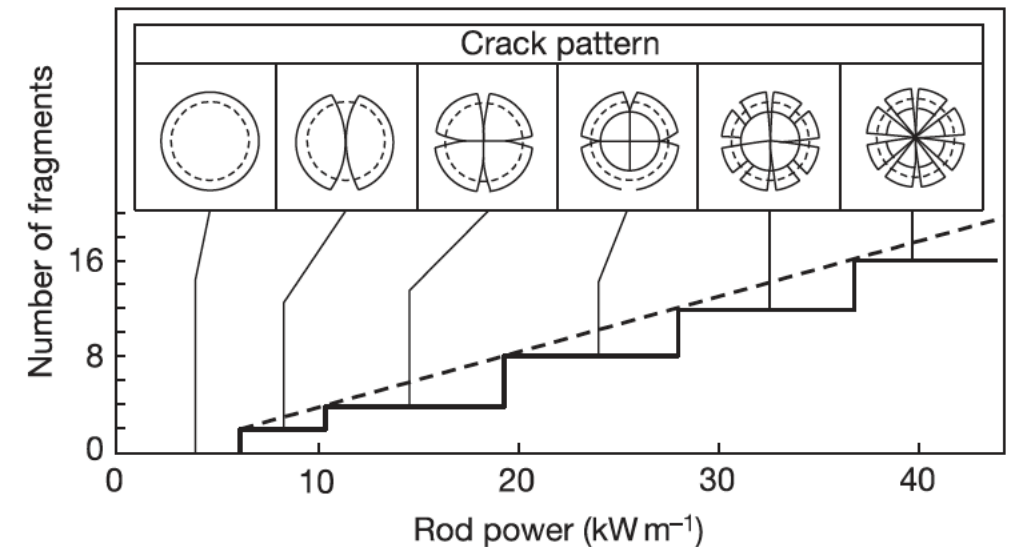
Pellet Deformation

- The large thermal gradients in the radial direction, and a lesser extent in the axial direction, contribute to non-uniform thermal expansion, which results in a shape that resembles an hourglass
- Pellet cracking due to thermal stresses further contributes to the hourglassing effect
- The edges of cylindrical pellets induce large local stress concentrations in the cladding when the pellet-clad gap closes with the enhanced risk of perforation



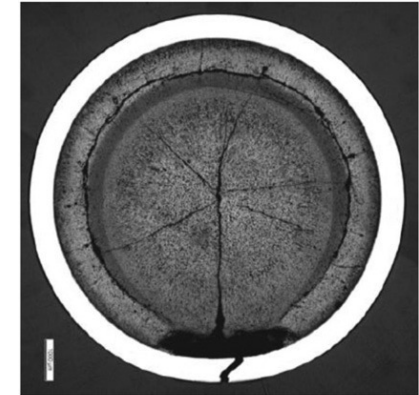
Fuel Cracking

- Fuel pellets experience varying degrees of fracture due to large internal stresses induced by thermal expansion that exceed the fracture strength of UO_2
- The fracture strength varies from 80 to 150 MPa and is strongly influenced by pellet microstructure, which decreases with respect to porosity, pore size, and grain size
- The number of fuel cracks generally increases with larger thermal gradients, thus the number of cracks in the fuel increases with respect to linear power

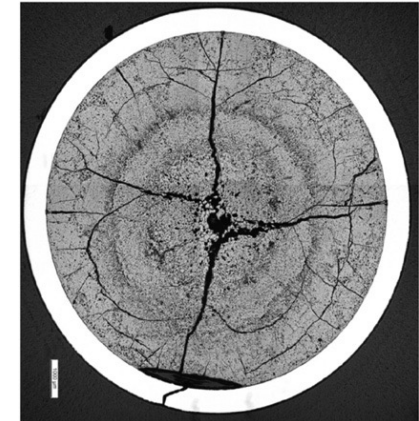


Missing Pellet Surface

- Several failures have been experienced in LWRs in the early 2000s due to physical defects in the fuel, often due to chipping, which is often referred to as a Missing Pellet Surface (MPS)
- The cladding eventually creeps down onto the fuel, except in the vicinity of the MPS
- A local stress concentration is experienced in the cladding adjacent to the MPS as a result of the bending moment that is induced by non-uniform contact coupled with an expanding pellet
- The increased local concentration of corrosive fission product species together with enhanced local stresses elevates the risk for SCC failure



PWR



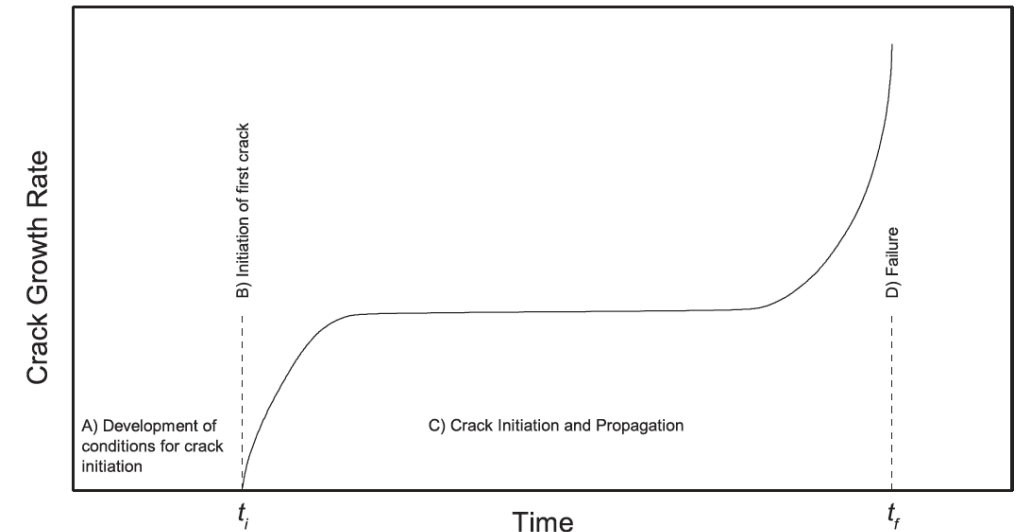
BWR



PHWR

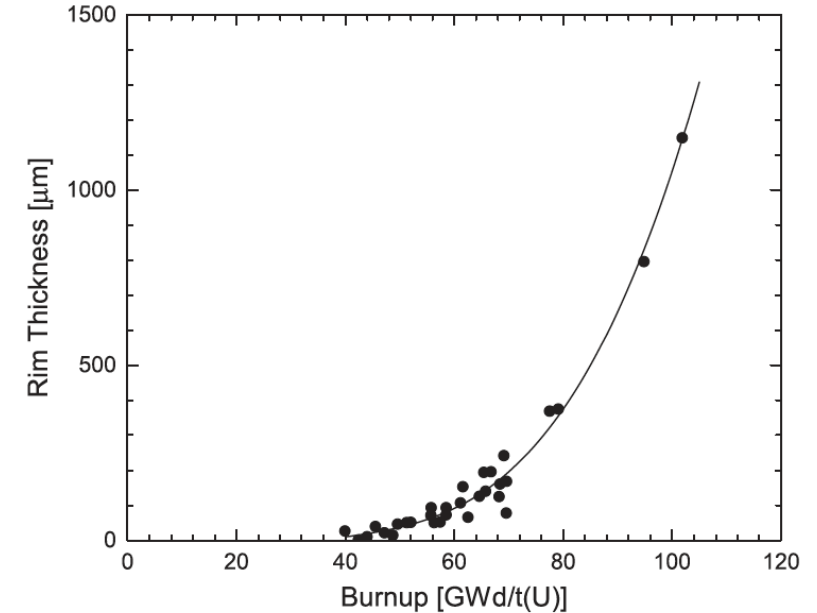
Sufficient Time

- A sufficient duration of time is required for SCC to develop in the cladding
- The SCC process can be divided into four stages:
 - Development of the corrosive environment and the surface conditions required for SCC to initiate,
 - Initiation of SCC,
 - Propagation of SCC, and
 - Failure
- The SCC-induced crack will typically propagate through the majority of the cladding wall, and then the remaining ligament typically fails by ductile shear



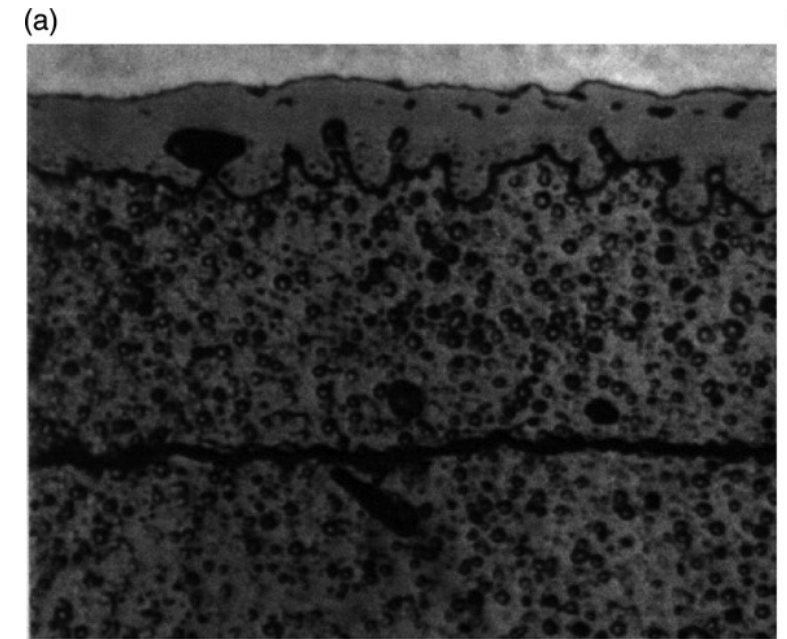
Effect of Burnup

- The period of time to establish the conditions for SCC is related to burnup, and is complicated by the numerous mechanisms associated with changes in both the fuel and cladding during the course of irradiation
- The mechanisms with relevance to SCC that become more pronounced with burnup include irradiation damage to the cladding, fission product swelling, fission gas release, and formation of a High Burnup Structure
- The local burnup in the rim region can be 2–3 times greater than the integral burnup in highly irradiated fuel, which means that the local concentrations of fission and activation products in the rim region are considerably higher, which have a direct influence on the fuel surface chemistry



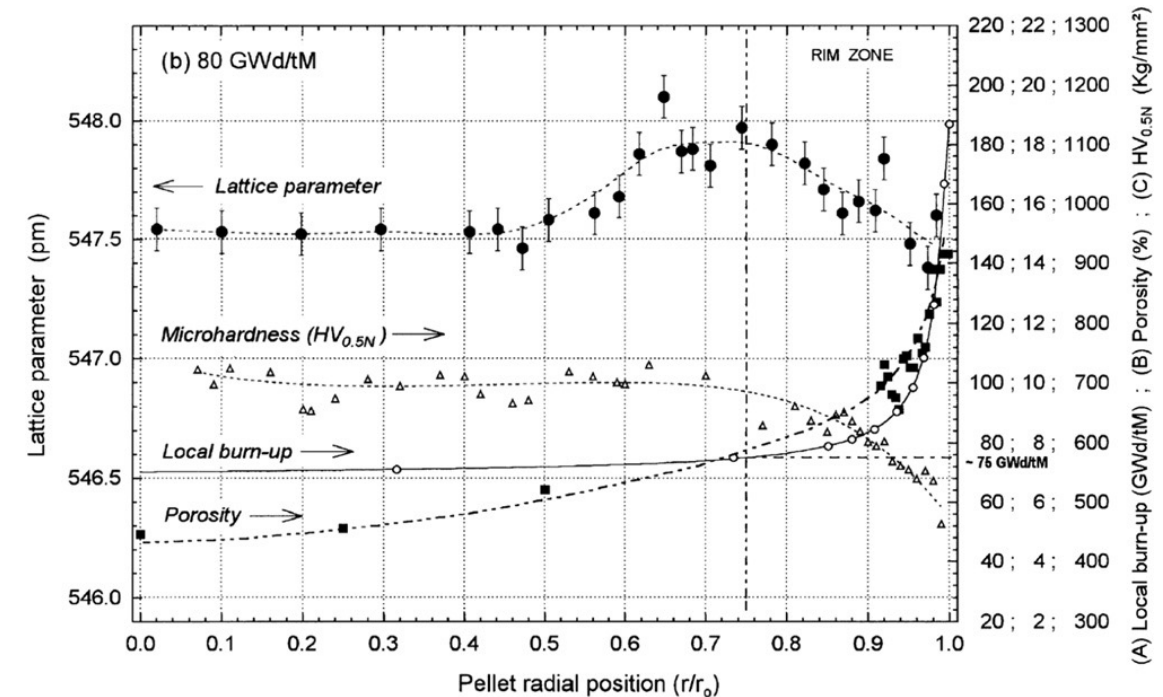
Rim/HBS Region

- In medium burnup fuels, an internal zirconia layer 6–12 mm thick forms on the clad inner wall as soon as pellet-clad contact occurs
- The coverage of the clad internal surface by zirconia tends to extend progressively with further irradiation and gap closing
- High burnup fuel shows the development of a very effective pellet-clad bonding characterized by an intimate mixing of U and of the internal zirconia layer
- Pellet-clad bonding, has been observed and seems to be controlled by the irradiation duration at closed pellet-clad gap



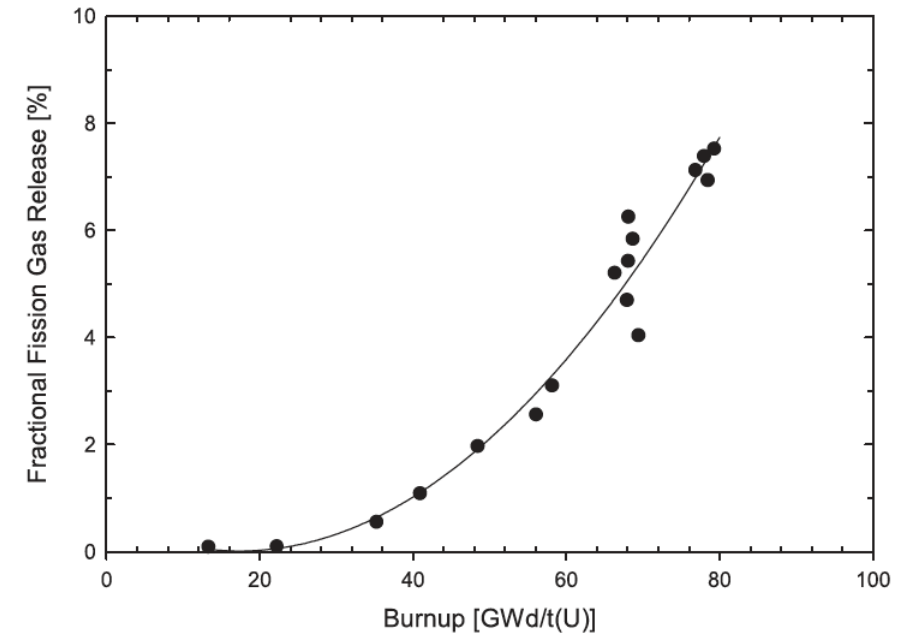
Rim/HBS Region

- The HBS region has a much higher porosity (up to 30% locally) than the bulk of the fuel, which affects the mechanical properties
- Microhardness measurements show a reciprocal trend of the strength with porosity
- The softening of the fuel surface might be beneficial in reducing mechanical stresses imposed by the fuel on the cladding at the point of contact



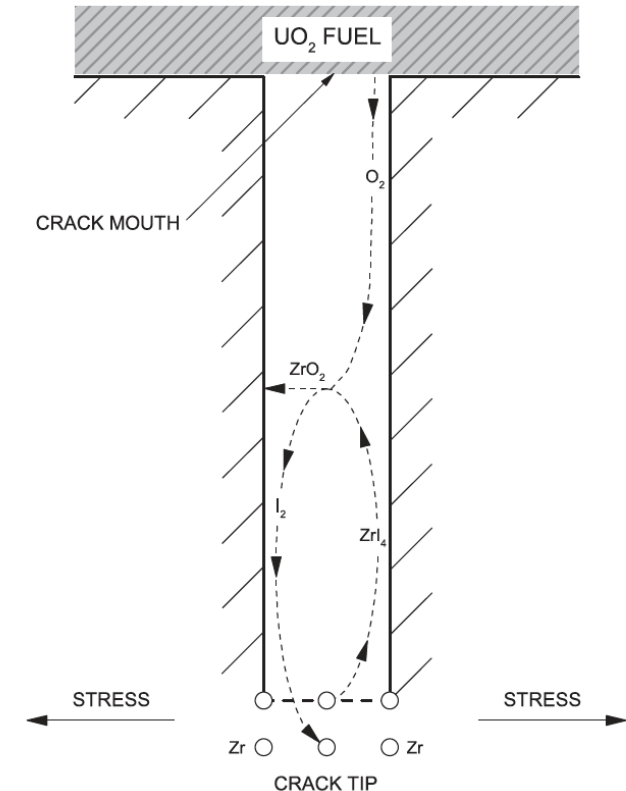
HBS and Fission Gas Release

- The large increase in porosity within the HBS in high burnup fuel also affects fission gas release, providing local intergranular accommodation for retaining fission gases
- Although the formation of the HBS promotes local fission gas retention, the absolute amount of fission gases that are released to the gap increases with burnup



Incubation Time

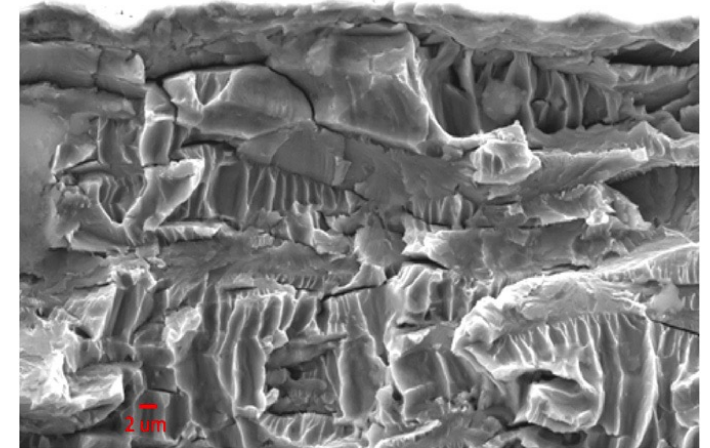
- The corrosive environment, represented by a sufficient inventory of chemically active fission gases in the gap, not only depends on burnup, but the ability of these gases to chemically attack the cladding
- This environment requires that the normally protective oxide coating on the inner surface of the cladding is breached, thus permitting corrosive species to chemically react with the bare cladding
- The incubation time reflects the time required for a flaw in the protective oxide to be developed and for sufficient ZrI_4 to form in the cladding, resulting in the development of cracks



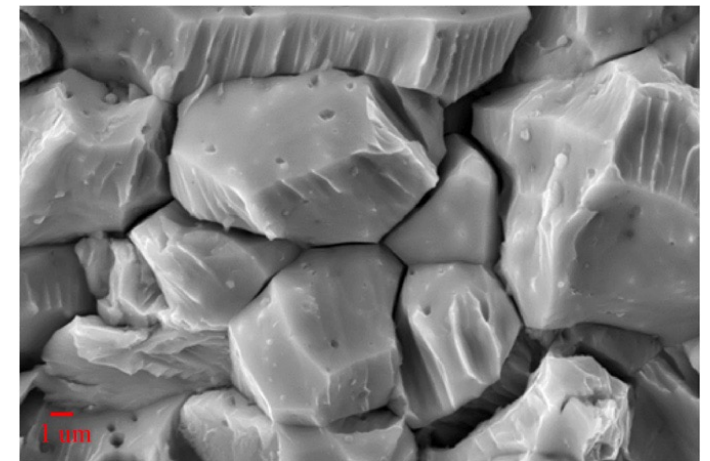
Crack Propagation

- Once a crack has initiated, it can propagate through the cladding wall with a sufficiently high applied load
- Both intergranular and transgranular propagation modes are possible
- The propagation rate is a linear function of the stress intensity factor, K_{SCC} , and is independent on the propagation mode for sufficiently high K_{SCC}
- The increase in iodine content generally increases the crack propagation rate
- Increasing temperature results in decreasing the susceptibility to PCI failure, while neutron irradiation has been found to increase susceptibility

Transgranular



Intergranular



Through-Cracks

- Following the formation of a through-wall crack and the ingress of water into the fuel-clad gap, the cracking process is arrested since the corrosive species (notably I, Cs, and Cd) have been discharged
- The ingress of water in the fuel-clad gap may result in clad hydriding on the inner surface
- The initial SCC crack can oxidize, and volume expansion may lead to resealing the primary failure
- All PCI cracks are pin-hole defects, whereas observable cracks are secondary due to clad hydriding or ductile tearing
- The time to failure depends on many parameters, but is generally determined by the local linear power, the change in linear power, and the local burnup

Reactor Susceptibility

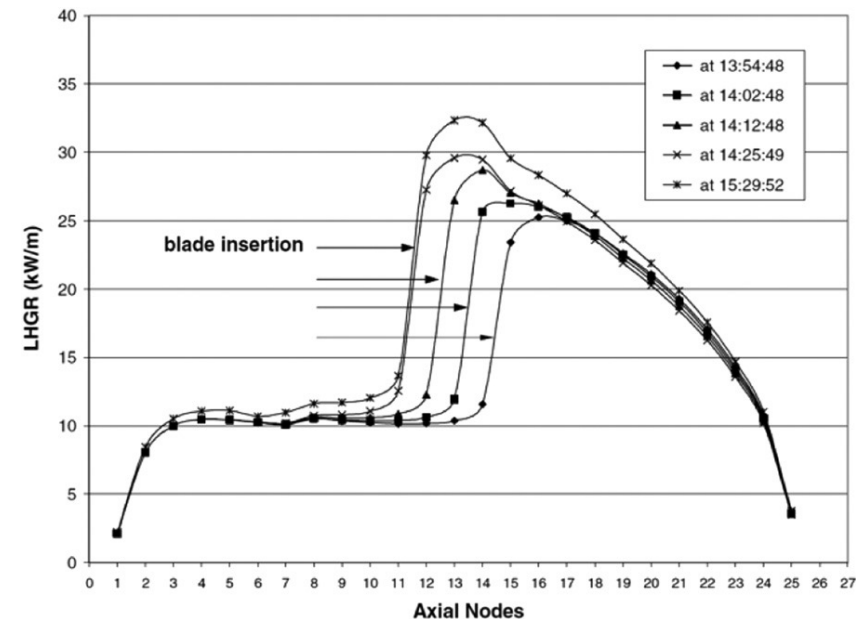
- All current PWR, BWR and CANDU reactors utilize UO₂ fuel, zirconium alloy cladding, and are water cooled
- The degree of susceptibility of each reactor and fuel design to PCI rests on numerous design specifications
- PWRs have smaller fuel diameters, BWRs have the thickest cladding
- The geometric design has an influence on the stresses in the fuel and cladding
- Linear power affects the temperature, which impacts a variety of other phenomena
- Discharge burnup influences the inventory of fission products in the fuel/cladding interface

Table 2 Pertinent fuel parameters typical of BWR, PWR, and PHWR reactor designs are compared below. Note that these values vary depending on a number of different factors, but are intended to give a broad impression of the relative differences

Parameter	PWR	BWR	PHWR
Cladding thickness (mm)	0.57–0.7	0.61–0.86	0.38–0.42
Cladding outer diameter (mm)	7.8–10.9	9.6–12.3	13.1–17.2
Initial gap thickness (μm)	~157		40–130
Fuel pellet diameter (mm)	7.6–9.4	7.84–10.4	12.1–14.3
Fuel pellet length-diameter ratio (unitless)	0.90–1.7	0.78–1.2	0.92–1.6
Initial fuel enrichment (%)	1.9–4.95	1.8–4.9	0.71 (natural)
Initial fuel porosity (%)	3.5–5	3–5	3
Chamfer	Yes	Yes	Yes
Dish	Two	Two	One
He pre-pressurization (atm)	7–24	5–10	1
Plenum	Yes	yes	no
Avg. linear power (kW m ⁻¹)	13–19	16–18	20–45
Peak linear power (kW m ⁻¹)	33–40	40–47	50–58
Avg. discharge burnup (GW d t(U) ⁻¹)	31–55	17–44	6.1–9.1
Peak discharge burnup (GW d t(U) ⁻¹)	39–65	31–50	15

Reactor Susceptibility

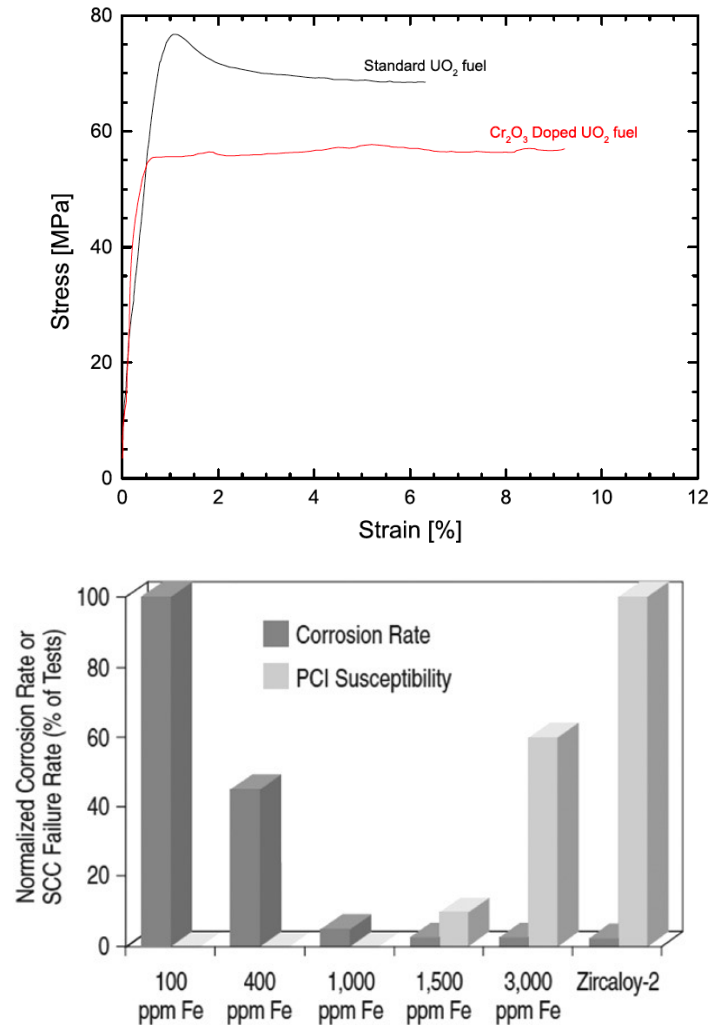
- In general, PCI failures are typically experienced (by any reactor) during a large change in power; thus, the manner in which power changes dictates to a large degree the likelihood of PCI failure
- Unlike BWRs, the neutron flux in a PWR is not primarily controlled by the insertion and extraction of control rods during operation
- Due to smoother control of reactivity and a lower linear power, PCI failures are significantly less frequent in PWRs than other major commercial power reactor designs
- PCI failures are more of a concern in BWRs
- Control blade maneuvers in BWRs create local power transients that often lead to PCI failures in fuel rods adjacent to these blades



The change in LHR resulting from the successive removal of three control rod blades

PCI Mitigation

- There are two primary approaches to mitigate PCI failures: 1) changes in the design of various components – notably, the fuel pellet, fuel cladding and fuel assembly; 2) the manner in which the reactor is operated can be altered to minimize PCI failures
- The design of fuel has changed to better optimize performance and reliability, including modifying the fuel pellet geometry, microstructure (i.e., grain size and porosity), and composition (i.e., initial O/M, minor additives)
- Many design changes of the cladding have been investigated, including the development of small grain sizes and texture control, alloy composition, inclusion of an inner liner and the application of a pellet-clad interlayer



PCI Mitigation

- Fuel assembly designs for all reactor types are constantly evolving as assemblies/bundles are improved to increase operational economics
- A continuing trend in design evolution is sub-division of the fuel into smaller diameter elements/rods to increase the total number of elements/rods, which increases assembly/bundle power without a corresponding increase in UO₂ temperature, thus mitigating thermally driven fuel failure mechanisms
- Other changes in general fuel assembly/bundle design include variations on fill gas pressure, presence and design of plenums to collect fission gases, changes to appendage design to improve CHF, general optimization of rod end regions in the reactor to mitigate end-flux-peaking
- The three variables that are controlled from an operational point of view are the linear power, change in linear power, ramp rate and discharge burnup

Summary

- Pellet-clad interaction (PCI) takes two forms
 - Pellet-clad chemical interaction, PCCI (bonding occurs)
 - Pellet-clad mechanical interaction, PCMI (pellet pushes and drags cladding)
- In order for SCC to initiate and propagate in any material, four conditions are simultaneously required:
 - A corrosive environment, a susceptible material, sufficient stress, and sufficient time
- BWRs more likely to have PCI failures than PWRs
- Two types of mitigation strategies to limit PCI failures