Chapter 28: Fuel Behavior under Abnormal Conditions

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28.1 Introduction

As discussed in Chapter 9, under normal operating conditions, cladding and core structural materials operate around 300ºC while fuel pellets experience peak temperatures below 2000ºC at the pellet centerline. At those temperatures, zirconium alloys remain in their low-temperature alpha (hexagonal close packed) phase, and the vapor pressures of major fuel constituents remain low such that most fission products are retained within the fuel pellets. However, under accident conditions, higher temperatures and other conditions significantly alter their behavior.

Several abnormal conditions are of special interest in addressing the safety of nuclear power reactors. Licensing authorities generally require that accidents be postulated, that safety equipment be installed to mitigate these accidents, and that analyses be performed to demonstrate that the plant would survive these accidents without major releases of radioactivity. As a result, much research has been devoted over the years to the behavior of fuel materials under such postulated accident conditions.

From the point of view of fuel behavior, the two most challenging types of accidents postulated for LWRs are reactivity-initiated accidents (RIAs) and loss-of-coolant accidents (LOCAs). The particular RIAs of interest are the most severe overpower events and the LOCAs of interest are the most severe undercooling events that seem credible. As such, these accidents involve the most limiting conditions that fuel materials might be expected to experience.

Because the ability to postulate credible accidents is limited, another category of accidents called severe accidents is imagined. These accidents do not appear to be credible because of diverse and redundant safety systems that are incorporated into plant designs, but nevertheless they remain of interest for providing bounding estimates of accident consequences. This chapter describes the behavior of fuel materials under accident conditions and the methods used to evaluate their performance.

28.2 Reactivity-Initiated Accidents (RIAs)

This postulated accident is initiated by a large reactivity excursion, which is followed in turn by a power excursion, and possibly the expulsion of hot fuel particles into the coolant, and the conversion of that thermal energy into mechanical energy in a steam explosion. A reactivity excursion was the cause of the accident that happened at the Chernobyl plant in April 1986 in which the steam explosion ejected core materials, and started a fire. The postulated RIA accidents would initiate differently in BWRs and PWRs, but the progression of subsequent events would be similar in these two reactor types (but clearly of much smaller scale and with much smaller consequences than that of Chernobyl).

In a BWR, the RIA accident is postulated to occur after a control blade hangs up in the core as the drive assembly is withdrawn, and some time later the control blade drops out of the core at its maximum velocity, thus causing a rod-drop accident. The rod-drop accident that produces the largest reactivity excursion occurs under cold zero-power conditions. Although no rod-drop accidents have occurred in BWRs, broken and cracked shafts were found in the Oskarshamn and
Forsmark plants in 2008 due to higher core temperatures that led to metal fatigue and these might be considered accident precursors [1].

In a PWR, the accident is postulated to occur after a crack develops around an upper head penetration (nozzle) that houses a control-rod drive and the drive unit and its attached control-rod cluster are ejected under pressure when the housing fails. This is called a rod-ejection accident, and the one that produces the largest reactivity excursion occurs under hot zero-power conditions. This accident is not possible in a PWR under cold conditions because the vessel must be pressurized (and therefore hot) to cause the ejection. No rod-ejection accidents have occurred in PWRs either, but a precursor occurred in 2002 in the Davis Besse plant when a control-rod-drive nozzle remained attached only by its stainless steel liner following nozzle cracking and severe boric acid corrosion of the reactor head.

28.2.1 Reactivity-Initiated Accidents with Fresh Fuel

When nuclear power plants were first licensed reactivity initiated accidents were considered and the limits to avoid fuel damage determined.

The RIA accident evolution

Regardless of the speed of a falling BWR control blade or an ejecting PWR control rod, the nuclear reaction soon becomes prompt-critical and from then on the rate of power increase and subsequent Doppler-caused decrease are dependent only on nuclear properties of the core design. In general, the resulting pulse has a full width at half maximum of only a few tens of milliseconds, as illustrated in Fig.28.1.

![Figure 28.1. Fuel rod power during an RIA with an energy deposition of 170 cal/g.](image)

During this time, little heat is transferred from the fuel pellet to the cladding and the event can be considered adiabatic. Consequently, a power pulse in an RIA is usually characterized by the maximum fuel enthalpy in cal/g and by the pulse half-width in ms. The total pulse energy deposited in the fuel pellet during the transient can be calculated from the integral of the power curve.
Figure 28.2 shows the pulse width for typical enthalpy additions in a PWR. For a given total energy, the pulse is somewhat broader in a BWR because of the looser nuclear coupling in that reactor type [2].

Consider a pulse that deposits 200 cal/g (0.84x10^6 J/kg) in the UO_2 fuel pellets within a Zircaloy-clad fuel rod, which is at zero power and at room temperature. Assuming that the energy is deposited uniformly in the UO_2 pellets, the temperature of the pellets will rise to 2600K (2327°C) according to the enthalpy relation shown in Fig. 28.3 [3]. The thermal expansion coefficient of UO_2 is given by

\[
\frac{\Delta L}{L_o} = K_1T - K_2 + K_3 \exp(-E_D / k_B T)
\]  

(28.1)

where \(\frac{\Delta L}{L_o}\) is the strain caused by thermal expansion (equal to zero at 300 K), T is the temperature in K, \(E_D\) is a defect formation energy, \(K_1, K_2, K_3\) are constants and \(k_B\) is Boltzmann’s constant (8.62x10^-5 eV/K).

<table>
<thead>
<tr>
<th></th>
<th>UO_2</th>
<th>PuO_2</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>(K_1)</td>
<td>1.0 x 10^{-5}</td>
<td>9.0 x 10^{-5}</td>
<td>K^{-1}</td>
</tr>
<tr>
<td>(K_2)</td>
<td>3.0 x 10^{-3}</td>
<td>2.7 x 10^{-3}</td>
<td>unitless</td>
</tr>
<tr>
<td>(K_3)</td>
<td>4.0 x 10^{-2}</td>
<td>7.0 x 10^{-2}</td>
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</tr>
<tr>
<td>(E_D)</td>
<td>0.43</td>
<td>0.44</td>
<td>eV</td>
</tr>
</tbody>
</table>

Parameters for Equation (28.1) (from p.2-49 of ref [3])

For a 15x15 fuel design, the as-fabricated gap between the pellets and the cladding is about 2% of the cladding diameter, or approximately 12 microns. The gap thus accommodates much of the pellet expansion leaving a net strain in the cladding of less than 1%, which fresh zirconium-alloy cladding can readily accommodate. Therefore, the mechanical interaction between the pellets
and the cladding should not lead to cladding failure (through-wall penetration or fracture) for fresh fuel. As shown in subsequent sections, this is not the case for high burnup fuel.

![Graph of enthalpy of UO2 as a function of temperature to 4,000 K](after [3]).

Notice, however, that 2327°C in this example is well above the melting point of Zr (1852°C). Although the cladding temperature cannot rise significantly during the 10-ms period of the power pulse, the heat in the pellets has to go somewhere, and it eventually goes through the cladding into the coolant. During this process the cladding temperature rises significantly. This is shown in Fig. 28.4, where the cladding temperature was calculated with FRAPTRAN [4] considering a somewhat smaller pulse (108 cal/g peak fuel enthalpy) and including appropriate heat-transfer kinetics.
Actual cladding temperatures depend on the heat transfer across the pellet-to-cladding gap and from the outside of the cladding to the coolant, in addition to time, thermal conductivities, and heat capacities. Because these processes are complicated and difficult to model, the progression of fuel damage during RIAs is customarily determined empirically by testing. The state of the fuel and the cladding after one such test on a fresh fuel rod is shown in Fig.28.5. This test was performed in the PBF test reactor at an initial temperature of 265°C and resulted in a peak fuel enthalpy of 285 cal/g with a pulse width of 13 ms [5]. As indicated in Figure 28.4, in the early part of the transient the fuel cladding can fail in a brittle manner if not enough ductility remains after reactor exposure to survive the loading imposed by pellet cladding mechanical interaction resulting from fuel expansion. If the cladding survives the initial part of the transient, the high temperatures later achieved can cause cladding failure by oxidation and hydrogen pickup during the high temperature excursion can lead to further cladding embrittlement which can cause failure upon quenching.
High temperature behavior is illustrated in Fig.28.5. It is clear from this photomicrograph that very high cladding temperatures were achieved during the transient. Evidence of the beta phase is seen in the center of the cladding, indicating temperatures above the phase transformation at about 850ºC. Also, large oxide layers are seen on the inside and outside surfaces of the cladding, and these could not have developed during the transient unless temperatures were well above 1000ºC. Further, the eventual failure (quench fracture) is a brittle fracture (as deduced from the fracture surface), and cladding embrittlement was caused by oxygen diffusing into the bulk metal from the oxide on the surfaces. This amount of embrittlement in such a short time indicates temperatures well above 1200ºC. The high-temperature behavior of this Zircaloy cladding is thus similar to that which occurs in a LOCA, as described in Section 28.3. Finally, it is clear that the failure occurred at a low temperature during cool-down because there is no oxide present on the fracture surface.

**RIA limits for fresh fuel**

A traditional value of 170 cal/g total enthalpy has been used by the nuclear industry to indicate the level at which cladding failure will occur by overheating. Although the technical basis for this traditional value appears to have been lost, that value is consistent with test results from the Russian IGR and BIGR reactors in which failure occurred by a swelling and rupture rather than a mechanical mechanism [2].

While cladding failure is of some interest in assessing safety consequences of an RIA because of small releases of fission products, the major consequence of this accident comes from a possible steam explosion. Merely causing a crack in the cladding will not necessarily expel hot fuel
particles into the coolant such that a steam explosion could occur. From Fig. 28.3, however, it is seen that fuel enthalpies greater than 275 cal/g (1.15 x 10^6 J/kg) would result in some molten UO₂. At some enthalpy higher than this value, the thermal expansion that takes place during the phase change would cause molten or hot solid particles of UO₂ to be expelled from the rod into the coolant. The classic work by MacDonald et al. found damage levels for fresh and low-burnup fuel as shown in Fig. 28.6[7]. MacDonald concluded that a safety limit should have been set at about 230 cal/g rather than 280 cal/g as adopted by the Nuclear Regulatory Commission because of confusion between total energy deposition and peak fuel enthalpy (some energy is lost in the tests even in the first 10 ms).

<table>
<thead>
<tr>
<th>Total Energy Deposition (cal/g)</th>
<th>Peak Fuel Enthalpy (cal/g)</th>
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<tbody>
<tr>
<td>378</td>
<td>~305</td>
</tr>
<tr>
<td>338</td>
<td>~275</td>
</tr>
<tr>
<td>287</td>
<td>~240</td>
</tr>
<tr>
<td>240</td>
<td>~205</td>
</tr>
<tr>
<td>168</td>
<td>~145</td>
</tr>
</tbody>
</table>

Fig.28.6. Photographs of fresh fuel rods after RIA testing in the SPERT reactor (after [7]).

Fuel enthalpy resulting from an RIA is related to the worth of the ejected control rod as shown in Fig.28.7 [2]. Because control-rod worths are generally less than $2, it is seen from this figure that the fuel enthalpy change would usually be less than about 70 cal/g. If 25 cal/g is added for an RIA that starts at about 300°C in a PWR, the total is still less than the 170 cal/g enthalpy level at which cladding failure would occur and much less than the 230 cal/g level at which hot fuel particles might be expelled from a fuel rod. Therefore, no cladding failure, fuel dispersal, or steam explosion would be expected for an RIA involving fresh LWR fuel up to 30 GWd/ton.

More recently it has been shown that these limits may not be valid at higher burnups [8, 9]. This is because the cladding suffers embrittlement during reactor exposure (see chapter 22) causing cladding failures at energy deposition levels below the limits set for fresh fuel. These changes
which could include the effects of radiation damage and corrosion and hydriding (the latter being more important) are discussed in the next section.

![Graph showing maximum fuel enthalpy change for RIA in a PWR at hot zero power for various values of delayed-neutron fraction, beta (after [2]).]

28.2.2 Reactivity-Initiated Accidents with High-Burnup Fuel

As of 2010, power reactors are achieving fuel burnups above 60 GWd/t (rod averaged); burnups above 40 GWd/t are considered high. Several properties of high-burnup fuel radically alter the evolution of an RIA in both BWRs and PWRs. Specifically: the fissile atom distribution is changed within the fuel pellets, the pellet-to-cladding gap is closed, often with pellet-cladding bonding, hydrogen is present in greater quantity the cladding, and fission gas bubbles are found within the fuel pellets. As shown above, one risk factor for RIA accidents diminishes with burnup: the possible energy deposition decreases because of lesser fissile material available. Thus the comments below are for a given energy deposition and the ability of a rod ejection to deposit a large enough amount of energy needs to be assessed separately. All of the other changes negatively affect the RIA response of the high burnup fuel in smaller or greater degree as discussed in the following.

Fissile atom redistribution and Rim Formation

Fissile atom redistribution can be seen in Fig.28.8(b) in a fuel rod with very high burnup [10]. When irradiation starts, the fissile atom density ($^{235}$U) is homogeneous through the pellet. When irradiation starts in parallel with the fissioning of $^{235}$U, $^{238}$U can absorb neutrons to become $^{239}$Pu. As the result of natural shielding, both this absorption reaction and the fission reaction occur preferentially at the periphery relative to the center of the fuel pellets. As burnup progresses this causes a build up of plutonium near the pellet surface. In this particular high-burnup fuel rod, the concentration of Pu atoms is more than doubled near the pellet surface relative to the centerline. The resulting increase in the number of fissions near the surface causes a much

---

1 Although radiation dose continues to accumulates further as burnup increases, the defect density is thought to saturate at relatively low exposures so that the burnup increases do not much change the irradiated microstructure.
greater concentration of fission gases and higher radiation damage. This particular region, called the rim region, exhibits a much different microstructure than that of the pellet centerline, showing a high density of high pressure bubbles, and very small grain size, as shown in figure 28.8a.

The formation of this rim has two important consequences for RIA response. First, a larger fraction of the total deposited energy will be deposited next to the pellet surface, and thus closest to the fuel cladding, maximizing the strain. Second, the large inventory of fission gases stored in the rim region is immediately available for release during the transient. The first of these factors increases probability of cladding failure. The second factor along with greater grain fragmentation in the rim can also lead to greater fuel dispersal.

As a consequence of the increased deposition of energy near the surface of the pellet, fuel temperatures near the pellet surface are very high early in the power transient (see Fig.28.9), and this near-surface peak temperature is much higher than the peak temperature in a fresh fuel rod. Because of high fuel surface temperatures the thermal expansion of the pellet is substantially enhanced. Figure 28.10 (left side) illustrates a typical thermal-expansion algorithm that is used in most computer models, especially for normal operating conditions when the peak temperature is at the pellet centerline. In this algorithm, the centerline node expands more than the other nodes and the expansion of each node is added to the next. Because UO₂ has no significant tensile strength, each nodal ring cracks and the incremental expansions add together adequately, without much sensitivity to the number of rings chosen.

Figure 28.10 (right side) shows the thermal-expansion algorithm that must be used for an RIA with high-burnup fuel. In this figure it is seen that the outer node expands more than the other nodes, and the expansion decreases going from the surface to the center. Conceptually, the outer node determines the expansion of the pellet as if the whole pellet were at the peak temperature. There are two practical problems with this algorithm. First, the temperature that is assigned to the outer node will be sensitive to the thickness of that node and hence the number of nodes chosen. Second, a very thin outer node will not have the ability to deform the cladding outwards without some inwards deformation of the node itself. Although this makes analysis difficult, it is seen that a high-burnup fuel pellet will expand considerably more than a fresh or low-burnup pellet for a given fuel enthalpy in an RIA.
Fig. 28.8. (a) SEM image showing the ultra-high-burnup rim structure, and (b) elemental distribution as a function of the distance from the fuel-to-cladding interface in a fuel rod with a burnup of about 105 GWd/t (after [10]).

*Fuel-cladding gap and Pellet-cladding Bonding*

The next factor to consider in high-burnup fuel is the pellet-to-cladding gap. As a consequence of fuel pellet swelling during normal operation combined with cladding creep-down due to a pressure differential during normal operation, the pellet-to-cladding gap will be closed. As a result, all the expansion of the fuel pellet will be transmitted to the cladding without any accommodation such as occurs in fresh fuel. Referring to an example above, it is seen that a 200 cal/g pulse would result in a cladding strain of more than 2.9% in high burnup fuel, whereas the same pulse would result in a cladding strain of less than 1% in fresh fuel.

Because of the difficulties in calculating pellet expansion, gap closure and cladding strain, empirical data can be used to obtain the strain values. Figure 28.11 shows data from RIA tests with high-burnup fuel in the Cabri test reactor in France and the Nuclear Safety Research
Reactor (NSRR) in Japan[2]. In general there is a trend from higher strain for PWR fuel with the largest operating pressure differentials (greatest cladding creepdown), to BWR fuel with modest operating pressure differentials, to test reactor fuel with no operating pressure differential. This figure shows measured plastic strain on the ordinate, so there would be an additional amount of elastic strain. Although there is considerable scatter in the data, it is clear that at 200 cal/g the total strain would be greater than 2.9% for any of these cases, in agreement with the above discussion. Although we concluded that fresh cladding could easily accommodate 1% strain during the power pulse that was considered, it is not clear that irradiated cladding could do likewise.

Fig.28.9. Edge-peaked pellet temperatures early in an RIA transient and center-peaked temperatures after significant heat transfer (after [8]).
Fig. 28.10. Thermal expansion algorithms for normal operation (highest temperature at pellet centerline) and for a zero-power RIA (highest temperature at the edge) in high-burnup fuel.

Fig. 28.11. Plastic strain measured in non-failed cladding as a function of maximum fuel enthalpy change for tests in Cabri and NSRR (after [2]).
The mechanical response of high burnup cladding is governed by the loading characteristics (Stress state, strain rate) and by cladding temperature, which varies during the transient. One of the crucial aspects of cladding failure during an RIA is at what temperature does the maximum load occurs. If the maximum load happens near the beginning of the transient, the cold cladding may not have enough ductility to survive the accident; if it happens later the greater ductility afforded by the higher temperature may help the cladding survive. Several failure mechanisms are thus possible, each with its own failure criterion, and overall cladding failure occurs if one of these criteria is satisfied.

**Cladding loading by PCMI**

As discussed above, the energy deposition causes the fuel temperature to rise, and the fuel to expand rapidly against the cladding. The resulting pellet-cladding mechanical interaction results in mechanical loading to the cladding tube. The cladding loading occurs very rapidly (at strain rates on the order of 1-10 s⁻¹).

The geometry of the problem and the pellet cladding bonding cause the cladding to be loaded in a biaxial state of stress which is somewhere between plane strain tension and equal biaxial loading. When there is zero bonding between the cladding and the pellet which can slide with no friction during PCMI, the state of stress is plane strain and when there is perfect bonding there is equal biaxial stress imposed on the cladding.

**Cladding Degradation during Reactor Operation**

Given the loading described above, cladding survival of the low temperature stage of an RIA depends on the remaining ductility. As mentioned in Chapters 17, 22, 25, zirconium alloy cladding changes during exposure to the reactor environment, suffering radiation damage and hydriding, both of which affect cladding ductility. It has been shown that both total hydrogen content and hydride distribution is crucial to determining cladding ductility.

The cladding mechanical response to a given loading is governed by its constitutive properties and by the failure mechanism. Because of radiation damage, corrosion and hydriding, these properties change with reactor exposure. As mentioned in Chapter 26, irradiation damage increases the yield stress, and decreases ductility and strain hardening. The hydrogen forms hydride particles that are more brittle than the matrix. As mentioned in Chapter 22, the hydrides redistribute themselves in response to temperature and stress gradients and thus they can form hydride rims or blisters, both of which reduce cladding ductility above and beyond the loss of ductility expected for a given hydrogen content[11, 12], as also shown in Chapter 26.

The most likely failure scenario is localized failure initiation in a weak spot of the cladding caused by an accumulation of hydrogen in the form of a blister or a rim. This is particularly clear in the case of oxide spalling, which creates a local cold spot, where hydrogen can precipitate and form hydride blisters. Such blisters are much less ductile than the alloy and fail early in the transient, creating a site for preferential cladding failure. Such a failure is shown in Fig. 28.12. Although the failure is apparently ductile because of its 45° shear angle, because the deformation is localized the overall cladding strain is small, and thus the failure is brittle.
For there to be significant consequences of the RIA in terms of fuel dispersal, the initial cladding failure has to propagate axially, and thus axial crack propagation and the variation of all these properties with axial distance is also important.

Figure 28.12: Zircaloy-4 cladding into which a rim of hydrides was introduced, and tested to failure at 300°C [13].

As mentioned in Chapter 22 during normal operation, corrosion of the zirconium-alloy cladding takes place and hydrogen is generated in the reaction. Much of the hydrogen is swept away in the flowing coolant, but a fraction of the hydrogen is absorbed in the cladding. At burnups around 60 GWd/t, typical cladding materials contain 100-800 wt. ppm hydrogen, depending on the cladding alloy. Only about 50 wt.ppm of that hydrogen will be in solution at normal operating temperatures, as can be seen from equation 22.32 [14]. The remaining hydrogen will be in precipitates that do not dissolve during the nearly adiabatic period of an RIA power pulse. Like all precipitates, the hydrides harden the cladding and reduce its ductility. The reduction in ductility will lead to cladding failure at lower and lower pulse energies as the hydrogen concentration increases (see Fig.28.13). This type of failure is referred to as pellet-cladding mechanical interaction (PCMI) failure and generally starts with crack initiation in the brittle hydride rim near the outside diameter and proceeds by a mixture of brittle and ductile failure modes through the rest of the cladding wall.

Two significant comments can be made about Fig. 28.13. First, such data are usually evaluated as a function of maximum fuel enthalpy change. This change corresponds to the expansion during the transient and does not include any expansion that might take place slowly as the temperature increases from room temperature to operating temperatures (in a PWR, for example). Irradiated cladding can usually accommodate expansion that takes place slowly by some combination of cladding creep-out and realignment of pellet fragments. Second, at very low hydrogen concentrations the cladding has enough ductility to survive the period of the power pulse and ultimately fails around 170 cal/g (total) by the high-temperature mechanism described above for fresh fuel.
Finally, fuel dispersal that could lead to a steam explosion would occur by a totally different mechanism in high-burnup fuel than in fresh fuel. In fresh fuel, it was necessary to have fuel melting to produce a sudden expansion that could expel hot fuel material. Fission gas bubbles, which accumulate on grain boundaries (seen as pores in Fig.28.8(a)), can cause such an expansion in high-burnup fuel. Rapid heating of fuel material, especially in the gassy ultra-high-burnup rim region near the pellet periphery, results in breakup of the fuel pellets and entrainment of particles in gas that escapes through the cladding failure. Although data are sparse, most tests with high-burnup fuel in which the failure occurred by the PCMI mechanism experienced fuel dispersal that produced significant pressure pulses in the test apparatus. Referring back to the discussion of Fig.28.7, most fuel rods in a reactor would undergo less than 70 cal/g fuel enthalpy change during an RIA, but PCMI failures in that range are seen in Fig.28.13. Therefore, some irradiated fuel with high hydrogen content might experience cladding failure, fuel dispersal, and the potential for a steam explosion in an RIA if such fuel were located near high-worth control rods.

### 28.3 Loss-of-Coolant Accidents (LOCAs)

A LOCA can be initiated by a pipe break or a leak in a component. A LOCA occurred at Three Mile Island in 1979 when coolant was lost through a relief valve that did not close after a series of unplanned events. This accident evolved into a severe accident when an emergency core
cooling pump, which had automatically started, was turned off because the operators mistakenly thought that the coolant was going “solid.” Many lessons were learned from the Three Mile Island accident, and improved instrumentation and procedures should significantly increase the likelihood that a future LOCA would be successfully terminated. However the fact that it did occur illustrates the need to consider the consequences of such accidents in reactor licensing in order that any effects are mitigated. This section will address LOCAs up to the point that they are successfully terminated by an emergency core cooling system and the effects of this accident on the fuel integrity, especially at high burnup.

In BWRs and PWRs, a LOCA is postulated to begin with a blowdown of steam through the pipe break or leak as water evaporates rapidly (flashes). The increase in coolant voids shuts down the nuclear reaction, but heat stored in the fuel and decay heat from existing radionuclides must be removed to prevent runaway core temperatures. As the pressure in the vessel drops and core temperatures increase, emergency cooling systems are activated to add water to the vessel and try to turn this temperature increase around. Figure 28.14 shows cladding temperatures and fuel rod pressures during a typical postulated LOCA that is successfully terminated by the emergency core cooling system [15].

![Fig.28.14. Cladding temperature (solid line) and rod pressure (dashed line) during a postulated LOCA (after[15]).](image)

Emergency core cooling systems are designed to provide enough cooling that fuel damage is limited, and the adequacy of these systems is determined by the extent of predicted fuel damage in a safety analysis. After a lengthy public hearing, the U.S. Atomic Energy Commission in 1973 concluded that fuel damage would be adequately limited if the cladding retained some ductility. Although limited ductility will not prevent fracturing, even marginal ductility will ensure that the cladding will exhibit its full tensile strength and be less susceptible to scratches and flaws than brittle material. This criterion was thought to be the best way to avoid fragmentation and keep fuel pellets within fuel rods in a coolable array. The ductility concept
was adopted by most countries with nuclear power reactors, and cladding ductility under LOCA conditions has been extensively studied.

28.3.1 Loss-of-Coolant Accidents with Fresh Fuel

In this discussion, events are considered that take place as cladding temperatures rise and then fall during a typical postulated LOCA transient. These events are indicated in Fig.28.15 and discussed in the following sections.

![Temperature Progression and Associated Phenomena](Fig.28.15)

**Phase Change**

The first event for zirconium-alloy cladding (Zircaloy-4 will be assumed for much of this discussion since much of this knowledge has been derived for Zircaloy-4 as other alloys are still being studied) is a phase change from the alpha phase (hexagonal close packed structure) to the beta phase (body-centered cubic structure). The transition begins at about 800°C and is complete at about 975°C. More is said about subsequent phase transformations in the paragraphs below.

**Ballooning and Rupture**

Because fuel rods are pressurized with He to improve internal heat transfer, fuel rods have a high internal pressure when they are at operating temperatures. By design, these fuel rod pressures are limited in relation to system pressures (~2250 psi in PWRs and ~1035 psi in BWRs) to ensure that cladding does not creep away from the fuel pellets thus degrading heat transfer. During a LOCA, system pressure is lost and the differential pressure across the cladding wall is therefore large. In parallel the yield stress reduces with temperature so that the cladding material softens enough to balloon and rupture in the same temperature range as the phase transformation. Figure 28.16 shows the temperatures and corresponding hoop stresses at which rupture occurs for Zircaloy-4 [16]. The engineering hoop stress $\sigma$ is related to the internal fuel rod pressure by

$$\sigma = \frac{d\Delta p}{2t}$$  \hspace{1cm} (28.2)
where $\Delta p$ is the pressure difference, $d$ is the cladding tube diameter and $t$ is the cladding thickness.

Cladding rupture under these circumstances is an unstable process. As the cladding deforms and reaches its ultimate tensile strength, subsequent expansion results in a reduced stress and hence runaway deformation. This will generally occur at just one local area that becomes a hot spot as the result of some non-uniformity in a fuel pellet. Because of the instability, it is not surprising that measured burst strains vary considerably from test to test, but important trends are present. Figure 28.17 shows a large number of measured burst strains that reveal a maximum at around 800°C [16]. For this material (Zircaloy), 800°C marks the beginning of the phase transformation from alpha to beta. Thus it is seen that strains are largest when the material ruptures in the pure alpha phase, but the strains are reduced substantially when rupture occurs with a mixture of alpha and beta phases.
Fig. 28.17. Maximum circumferential strain as a function of rupture temperature for Zircaloy cladding heated in steam (Fig. 8 of Ref. [16]). Different symbols correspond to different tests in the database shown in Ref. [16].

Fig. 28.18 shows an unirradiated fuel rod that has ruptured in a simulated LOCA transient [17]. The size of the cladding balloon at the time of rupture is important for two reasons. First, the swollen fuel rods reduce the flow area for coolant during the subsequent course of the LOCA. However, the increased surface area for cooling along with the turbulence introduced by the balloon usually result in a little extra cooling in the balloon region rather than a temperature increase.

Fig. 28.18. Ballooned and ruptured region of unirradiated Zircaloy-2 after undergoing LOCA conditions. (after [17]).
The second effect of the balloon size is that balloons will have a thinner wall than the undeformed cladding, thus leading to higher oxidation when measured as a percentage of the wall thickness (ECR). In fact, oxidation will also take place on the inside of the cladding as well as on the outside because steam will enter through the rupture opening. This enhanced two-sided oxidation plays a significant role in most safety analyses.

### Oxidation

When zirconium is exposed to steam, the zirconium is oxidized at a more rapid rate than during normal operation (corrosion). As the cladding temperature increases above 1000ºC, the cladding surface oxidizes rapidly enough that more than 10% of the cladding wall thickness can be consumed during the period of a LOCA before the core is cooled by emergency systems. The high-temperature tetragonal oxide structure is normally adherent and forms a protective layer on the cladding surface. Thus hydrogen that is liberated in the high-temperature reaction does not get into the metal and is swept away with the flowing steam.

As the protective oxide builds up on the surface of the metal, it slows down the rate of oxidation by increasing the distance that oxygen has to diffuse to reach the metal surface. In the important temperature range of 1000-1500ºC, the weight gain from the ingress of oxygen per unit time is found by experiment to obey parabolic kinetics, as described in Chapter 22

\[ w = K t^{1/2} \] (28.3)

where \( w \) is weight gain in grams per square centimeter of surface area and \( K \) is a temperature-dependent coefficient of the form

\[ K = K' \exp\left(-\frac{Q}{RT}\right) \] (28.4)

An early study of oxidation of zirconium was performed by Baker and Just [18] and the results fit to equation (28.4). Although the Baker-Just correlation has been found to overestimate oxidation by as much as 30% around 1200ºC, this equation is given below because it has been so widely used:

\[ w_{BJ}[g/cm^2] = 2.02 t^{1/2} \exp\left(-11,449/T\right) \] (28.5)

where \( T \) is the temperature in K and \( t \) is time in seconds. A more recent and more accurate study of the oxidation of Zircaloy-4 was performed by Cathcart, Pawel, et al.[19]. The so-called Cathcart-Pawel correlation is

\[ w_{CP}[g/cm^2] = 0.602 t^{1/2} \exp\left(-10,050/T\right) \] (28.6)

A related parameter that is often used is ECR, or equivalent cladding reacted. ECR is defined as the percentage of the cladding thickness that would be oxidized if all the oxygen stayed in the oxide layer as ZrO₂. This is an artificial parameter because some of the oxygen diffuses into the...
metal, but it is useful, and is directly related to weight gain by simple geometric factors, along with factors based on the density of Zr (6500 kg/m³ = 6.5 g/cm³) and the atomic masses of Zr and oxygen. The conversion is given in the equations below for one-sided oxidation

$$ECR_{1s} = 43.9 \left[ \frac{w}{h} \left( 1 - \frac{h}{d_o} \right) \right], \quad (28.7)$$

and two-sided oxidation.

$$ECR_{2s} = 87.8 \frac{w}{h} \quad (28.8)$$

where ECR is in percent, $h$ is cladding thickness in cm, and $d_o$ is cladding outside diameter in cm.

The oxidation reaction of zirconium in steam is exothermic, with a heat of reaction of 140.5 kcal/mole of Zr ($6.45 \times 10^6$ J/kg) [3]. This reaction heat (also called metal-water reaction heat) can be greater than the decay heat from radioactive nuclides at higher temperatures, and the reaction heat will then dominate the heat-transfer situation. If $dw_{Zr}/dt$, in units of g.cm⁻².s⁻¹, is the rate of Zr mass reacted with steam, then the power, $P$, in kW/cm generated by the metal-water reaction per centimeter length of cladding is

$$P = 20.3 d_o \frac{dw_{Zr}}{dt} \quad (28.9)$$

If it is assumed that all of the oxygen formed stoichiometric ZrO₂, then the mass of zirconium reacted is simply

$$w_{Zr} = \frac{91.2}{32.0} w \quad (28.10)$$

where 91.2 is the atomic mass of zirconium, 32.0 is the atomic mass of diatomic oxygen, and $w$ is the weight gain in grams per square centimeter of surface area. and thus

$$P = 57.9 d_o \frac{dw}{dt} \quad (28.11)$$

An example of linear power for various temperatures and various oxidation levels is shown in Fig.28.19.
Fig. 28.19. Linear power generation from metal-water reaction for cladding with a diameter of 1.25 cm and various oxide thicknesses (after [3]).

**Diffusion of Oxygen into the Metal**

At the same time that oxidation is proceeding rapidly, oxygen from the cladding surface is diffusing rapidly into the cladding metal. Zirconium alloys in the beta phase cannot hold much oxygen, however. Therefore, as oxygen pours into the metal, near-surface regions with high oxygen concentrations change phase again – this time back to the alpha phase, which is stabilized by the oxygen (the so-called oxygen-stabilized alpha phase). The oxygen solubility limit in the alpha and beta phases is temperature dependent as shown in Fig. 28.20[20]. At 1200°C, for example, the beta phase can hold only about 0.7 wt% oxygen.

Finally, the emergency core cooling system will reflood the core, temperatures will begin to come down, and all the cladding metal will return to the alpha phase. The imprint of the high-temperature phases can actually be seen in the microscope at room temperature as shown in Fig. 28.21[17]. When the cladding temperature reaches its wetting (Leidenfrost) temperature in the range of 600-800°C, the cladding will quench and the temperature will fall quickly to the average water temperature. Because the containment is sealed, the containment pressure will rise somewhat thus elevating the boiling point of water to about 135°C, so the cladding does not return exactly to room temperature. At this lower temperature, oxygen diffusion is effectively stopped and the high-temperature oxygen distribution is frozen into the cladding as illustrated in Fig. 28.22.
Fig. 28.20. Pseudo-binary Zircaloy-oxygen phase diagram (after [20]).

Fig. 28.21. Unirradiated Zircaloy-2 after oxidation in steam at 1200ºC for 600 seconds, as seen in a microscope at room temperature. (after [17])
Zircaloy metal containing more than about 0.6 wt% oxygen is found to be brittle. Therefore, only the low-oxygen material, which was in the beta phase when it was at high temperature, is ductile. To determine the LOCA conditions under which fuel rod cladding becomes brittle, mechanical tests are usually performed at 135°C on short lengths, or rings, of cladding specimens that have been exposed to high-temperature steam for different lengths of time. A ring-compression test, as illustrated in Fig.28.23, is generally used for this purpose. During compression, maximum tensile stresses are generated at the 12:00 o’clock and 6:00 o’clock positions on the inside diameter (ID) and at the 9:00 o’clock and 3:00 o’clock positions on the outside diameter (OD). Zero ductility is assumed when failure occurs at one of these locations with a very small (1%) permanent strain.

The transition from ductile to brittle failure could then be correlated with time, but the time would be strongly dependent on the steam temperature. Instead, the ductile-to-brittle transition is usually correlated with the calculated (not measured) ECR using either the Baker-Just equation or the Cathcart-Pawel equation. This is in effect a variable transformation that combines both time and temperature. Using this method, the transition from ductile-to brittle behavior is found at approximately 17% ECR as calculated with the Cathcart-Pawel correlation for modern zirconium alloy cladding materials, and this result is nearly independent of temperature. In other
words, enough oxygen diffuses into the metal to cause embrittlement in the same time that it takes Zircaloy to oxidize to 17% ECR. Because this is just a mathematical transformation, the same Zircaloy oxidation equation is used to evaluate embrittlement in all zirconium alloy cladding materials.

It is interesting to note that rate of oxidation and embrittlement of cladding materials are both controlled by the diffusion of oxygen, but the diffusion mechanisms are very different. In one case, oxygen diffuses through the oxide and in the other case oxygen diffuses through the metal. Oxides such as ZrO$_2$ have ionic crystal structures such that local charge neutrality must be maintained. Therefore, if an aliovalent impurity such as Nb with a valence of +5 is incorporated into the oxide, which normally has Zr cations with a valence of +4, the local lattice defect concentration is modified and the diffusion rate is altered. Note that Sn has a valence of +4 just like Zr, so Sn impurities do not significantly affect diffusion in ZrO$_2$. Consequently alloys like M5 (Zr-1%Nb) would be expected to oxidize at different rates than Zircaloy (Zr-1.4%Sn).

On the other hand, diffusion of oxygen in Zr metal takes place by an interstitial mechanism. The number of interstitial locations available for oxygen diffusion is fixed and not affected by substitutional impurities such as Sn and Nb. Therefore, the rate of embrittlement of different zirconium-based cladding alloys is not expected to be sensitive to alloy composition.

### Hydrogen Absorption

There are two special circumstances in which hydrogen can be absorbed in the cladding metal during a LOCA transient. One involves breakaway oxidation, which is similar to breakaway corrosion described in Chapter 22 and the other involves ruptured balloons. The significance of hydrogen absorption is that hydrogen acts like a catalyst and accelerates the embrittlement process, partly by increasing the rate of diffusion of oxygen in the metal.

It was stated above that the reaction of zirconium with steam during a LOCA normally produces a protective oxide layer on the cladding surface. But this is not always the case. The normal protective tetragonal form can transform to a monoclinic form under some circumstances. The monoclinic form of ZrO$_2$ is fragile and contains many cracks that give hydrogen access to the metal. Very soon after the monoclinic oxide forms at LOCA temperatures, sufficient hydrogen enters the metal to cause embrittlement long before the 17% ECR level is reached.

Several conditions have been identified that lead to early breakaway oxidation[17, 21]. A rough surface on the cladding promotes the formation of a monoclinic oxide by providing alternate compressive and tensile stress states for crystal growth. Fluorine, which has been used in some etchants, also promotes breakaway. Impurities in zirconium produced by the Kroll process seem to be beneficial in preventing breakaway compared with pure electrolytic zirconium. The origins of breakaway are not well understood, however, and there may be other factors as well.

Figure 28.24 shows two cladding materials that have the same nominal composition, but were fabricated differently. The older Russian E110 alloy is seen to experience breakaway oxidation in a few hundred seconds compared with the French M5 alloy, which shows no evidence of breakaway after 2400 seconds. After learning about the factors mentioned above, the Russian
manufacturer, TVEL, altered the manufacturing process to avoid early breakaway in the E110 alloy [22].

![Image of E110 cladding showing early breakaway oxidation at 1000ºC compared with modern M5 cladding that is resistant to breakaway oxidation.](after [17] and related unpublished figures)

28.3.2 Loss-of-Coolant Accidents with High-Burnup Fuel

The same properties of high-burnup fuel that altered RIA behavior (Sect 28.2.2) also alter LOCA behavior, but in an entirely different way.

**Effect of Hydrogen**

All of the hydrogen that existed as hydride precipitates in high-burnup fuel during normal operation will go into solution at LOCA temperatures as can be seen from equation 22.32. As mentioned above, hydrogen in the cladding metal increases the rate of oxygen diffusion at LOCA temperatures and accelerates the embrittlement process such that embrittlement occurs in less time than it takes to oxidize Zircaloy to 17% ECR. Figure 28.25 shows this effect for cladding with various hydrogen concentrations, using the calculated ECR (Cathcart-Pawel correlation) as the measure of time at temperature. Data points on this figure that are not labeled correspond to unirradiated cladding samples that were either as-fabricated or artificially charged with hydrogen. Each point on this figure represents a series of ring-compression tests that were taken to locate the embrittlement threshold. Notice that the uncertainty seems to be larger at the lower hydrogen concentrations and that the trend appears to apply equally to all of the zirconium alloys as expected.
ID Oxygen Pickup

Closure of the pellet-to-cladding gap occurs early during the burnup process, and long-term contact between the pellet and cladding at operating temperatures results in diffusion welding, or bonding. This occurs earlier in PWRs than in BWRs because of different design pressures and cladding thicknesses, and the bonding is fully developed by about 50 GWd/t in PWRs and 60 GWd/t in BWRs. Bonding can be seen microscopically (Fig.28.26), and tangible evidence is also found by technicians who remove fuel from high-burnup specimens for testing purposes. When bonding is present, impact drilling is usually required to remove the fuel, and the bonding interaction layer along with attached pieces of fuel remain attached to the cladding.
Fig. 28.26. Interaction bonding layer between UO₂ fuel and Zircaloy-4 cladding in high-burnup specimen at 83 GWd/t (after [24]).

The bonding layer is found to be largely ZrO₂, and insights to this bonding process can be gained from Fig. 28.27. This Ellingham plot shows the free energy of formation of oxides with various metals that are relevant to nuclear fuel materials. Notice that the formation energy is nearly the same for Zr, U, and Pu (prevalent in high-burnup fuel) and that all form dioxides. Thus oxygen can be exchanged freely among them when the cladding and the fuel are in contact.
The consequence of fuel-to-cladding bonding for LOCA behavior is that an oxygen source is then present on the cladding inside diameter (ID) as well on the outside diameter (OD). This is illustrated in Fig.28.28.

Fig.28.27. Free energy of formation of oxides of zirconium alloy constituents and some impurities. (after [17]).
During the high-temperature portion of a LOCA, cooling is poor and the heat flux through the cladding is small. As a result, the cladding is nearly isothermal when it is at its highest temperatures and most oxygen diffusion is taking place. Therefore, the oxygen diffusion rate is the same on the ID and the OD. Further, since the quantity of oxygen needed to embrittle the metal is rather small (see Fig. 28.22), the thicknesses of the OD and ID oxide layers are relatively unimportant – there is usually enough oxygen. Thus, when a well-developed bonding layer is present, it is expected that the same amount of oxygen will enter the metal from the ID and the OD.

Confirmation of this is seen in Fig. 28.29. These micrographs were taken on a high-burnup fuel rod after testing under LOCA conditions in the Halden test reactor (pre-test micrograph in Fig. 28.26). In Fig. 28.29, the microscopist has measured and labeled the thickness of the oxygen-stabilized alpha layer at the OD and ID at two azimuthal locations. In all cases this alpha layer was found to have the same thickness (about 20 microns in this specimen). Because the interior edge of the alpha layer is fixed at an oxygen concentration of about 6.7 wt% (see Fig. 28.22), the oxygen concentration profile is the same on the ID and OD thus demonstrating that the same amount of oxygen entered from both directions.
Fig. 28.29. Grain morphology and alpha layer at OD and ID of high-burnup fuel rod exposed to LOCA conditions. (after [24]).

**Fuel Relocation**

Because UO₂ is a brittle ceramic, reactor start-up and subsequent power changes cause fuel pellets to crack even at moderately low burnups. Figure 28.30 shows pellet fragments in two such irradiated fuel rods that have been exposed only to normal operation. If these pellet fragments could move axially into the enlarged volume of a cladding balloon during a LOCA, the heat generation rate within the balloon due to radionuclide decay and stored heat would increase. This temperature increase would accelerate the embrittlement process in the balloon region.
As cladding temperatures rise during a LOCA, unstable deformation, ballooning, and rupture will all occur within a few seconds once the rupture temperature and pressure conditions are met (Fig. 28.16). Immediately thereafter, cracked pellet fragments from just above the ballooned region are found in tests to move into the enlarged volume of the balloon. This movement, or relocation, is driven by gravity, and an example is shown in Fig. 28.31.

Randomly oriented granules in a rubble bed or heap are characterized by a packing fraction that is less than unity. It is clear from Fig. 28.31 that the fragmented fuel particles that have relocated into the balloon have a lower overall density than the original fuel pellets from which the fragments came. Therefore, a certain minimum balloon size would be required to realize an
actual increase in fuel mass within the balloon region. Figure 28.32 shows that the relative volume increase needed to accommodate additional mass in the balloon is about 18% for a large number of simulated LOCA tests with irradiated fuel rods. This corresponds to a packing fraction of about 78% on average. A diametral strain of less than 9% will give a volume increase of 18%, so it is clear from Fig.28.17 that essentially all balloons that occur during a LOCA will be large enough to accommodate extra mass by fuel relocation.

![Graph](image)

**Fig.28.32.** Pellet stack reduction as a function of volume increase in cladding balloons for pre-irradiated rods. (after [25])

A special case of fuel relocation can occur at very high burnups such as that shown in Fig.28.8. Porosity is very high in the rim region (22% in the case shown), and the pores are filled with fission gas at high pressure. When the fuel rod pressure is released at the time of cladding rupture, the unopposed pressure within the pores fractures the brittle fuel material and very small fuel particles are entrained in the fission gas and fill gas that escapes through the rupture opening.

Figure 28.33 shows the gamma scan of a fuel rod segment that had been subjected to LOCA conditions in the Halden test reactor. This fuel had a burnup of about 92 GWd/t, and more than 40% of the fuel volume had the rim structure. In this test, the entire upper portion (19 cm) of the fuel above the rupture opening was lost during rod depressurization. Similar out-of-pile tests have been run at Argonne National Laboratory on fuel that had a burnup of about 57 GWd/t with the observed loss of fuel amounting to less than that in one fuel pellet. Earlier LOCA tests at the Karlsruhe Nuclear Research Center on fuel with burnups no greater than 35 GWd/t resulted in no loss of fuel. Thus very high burnup fuel with a well developed rim structure may be susceptible to significant loss of fuel particles during a LOCA transient.
Fig. 28.33. Gamma scan of very high burnup (~92 GWd/t) fuel rod showing major loss of fuel material after LOCA testing (after[26]).

### 28.4 Severe Accidents

If overpower events or undercooling events are not mitigated by adequate operating limits and emergency systems, then fuel temperatures could escalate to temperatures around 2000°C and above. At these high temperatures, new materials-related phenomena occur [27]. Table 1 lists melting and boiling points of many core materials and fission products, and these values provide insights into material behavior and the release of radioactive species during an accident.

#### Table 28.1. Melting and boiling points of materials of interest.

<table>
<thead>
<tr>
<th>Material</th>
<th>Melting Point (°C)</th>
<th>Boiling Point (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volatile Fission Products</td>
<td></td>
<td></td>
</tr>
<tr>
<td>--------------------------</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>I₂</td>
<td>114</td>
<td>185</td>
</tr>
<tr>
<td>HI</td>
<td>-51</td>
<td>-35</td>
</tr>
<tr>
<td>CsI</td>
<td>626</td>
<td>1280</td>
</tr>
<tr>
<td>CsOH</td>
<td>315</td>
<td>990</td>
</tr>
<tr>
<td>Te</td>
<td>450</td>
<td>988</td>
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<table>
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<th>Refractory Fission Products</th>
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<tr>
<td>BaO</td>
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<td>2808</td>
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<tr>
<td>Ru</td>
<td>2250</td>
<td>4150</td>
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<tr>
<td>SrO</td>
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<td>3249</td>
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<td>La₂O₃</td>
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<td>&gt;4000</td>
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<th>Control Rods</th>
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<tr>
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<td>961</td>
<td>2163</td>
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<tr>
<td>In</td>
<td>157</td>
<td>2073</td>
</tr>
<tr>
<td>Cd</td>
<td>321</td>
<td>767</td>
</tr>
<tr>
<td>B₄C</td>
<td>2470</td>
<td>&gt;3500</td>
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<tr>
<td>Hf</td>
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<th>Cladding Alloys</th>
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<tr>
<td>Zr</td>
<td>1852</td>
<td>4409</td>
</tr>
<tr>
<td>Sn</td>
<td>232</td>
<td>2603</td>
</tr>
<tr>
<td>Nb</td>
<td>2468</td>
<td>4927</td>
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<th>Stainless Steel</th>
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<td>Fe</td>
<td>1535</td>
<td>2862</td>
</tr>
<tr>
<td>Cr</td>
<td>1890</td>
<td>2672</td>
</tr>
<tr>
<td>Ni</td>
<td>1453</td>
<td>2914</td>
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<tr>
<td>Mn</td>
<td>1244</td>
<td>2062</td>
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<table>
<thead>
<tr>
<th>Fuel</th>
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<tbody>
<tr>
<td>UO₂</td>
<td>2840</td>
<td>3293</td>
</tr>
</tbody>
</table>

**Eutectic Melting**

Figure 28.34 shows a phase diagram for mixtures of Zr cladding and UO₂ fuel under accident conditions that expose the fuel rods to a lot of steam [28]. Because of the steam exposure, the Zr is saturated with oxygen and is in the oxygen-stabilized alpha phase. The UO₂ has also given up oxygen to the zirconium metal during the temperature escalation and the remaining mixture of U and Zr is in an oxide form that is hypostoichiometric (U,Zr)O₂₋ₓ. Notice that there is a relatively low-temperature liquid that forms just above 1900°C, below the melting point of oxygen-saturated Zr and well below the melting point of (U,Zr)O₂₋ₓ.
As indicated in this figure, oxygen-stabilized alpha zirconium will dissolve UO₂ if they are at temperatures in excess of about 1900°C. A eutectic melt is formed with a 15 percent mole fraction of UO₂, whereas a two-phase mixture of liquid with a dispersed (U,Zr)O₂ solid component exists at higher UO₂ compositions, analogous to a slush. A mixture of two liquids occurs at 2,400°C when the UO₂ mole composition is between 23 and 70 percent. Thus a large volume of UO₂ could be dissolved by molten cladding at temperatures in excess of 1,900°C, and core melting thus begins at this relatively low temperature.

**Fission Product Release**

Fission products are created inside the UO₂ fuel pellets, and their release to the atmosphere during an accident would present a health hazard. Significant release of most of the hazardous fission products occurs only at very high temperatures, and in general, the release takes place in three steps as illustrated in Fig.28.35. First, a fission product must move from the UO₂ interior to a surface, from which it can escape to the atmosphere. Migration within the oxide’s crystal lattice takes place by some diffusion mechanism. Second, upon reaching a free surface, a fission product must vaporize in order to get into the atmosphere. Third, the fission product as a vapor or a condensed aerosol must be swept through the atmosphere with other gases.
Fig.28.35. Fission product release from the point of creation in the fuel pellet to a point outside the core region.

Fission product release, like radioactive decay, is said to be first order when it depends to first order on the amount present. Although not mechanistic, a model based on this principle, agrees adequately with measured releases and the model has been widely used [27]

First order kinetics means that if \( M(t) \) is the unreleased mass of the species of interest at some time \( t \), then

\[
\frac{dM}{dt} = -k(T)M
\]  

(28.12)

where \( k(T) \) is the temperature dependent release-rate coefficient at temperature, which integrates to

\[
M = M_o \exp(-kt)
\]  

(28.13)

where \( M_0 \) is the initial mass.

This equation is often used in computer codes where time steps are finite and temperature is assumed to be constant during each time step. In that application, the amount released during time step \( \Delta t \) is \( \Delta M \), where

\[
\Delta M = M_o - M_o \exp(-k\Delta t)
\]  

(28.14)

or the fraction released in time \( \Delta t \) is
This is the form of the equation given in Ref.[29]. The release-rate coefficient \( k(T) \) is, in fact, a strong function of temperature and like the diffusion processes described above is found to have an Arrhenius form,

\[
k(T) = k_0 \exp\left(-\frac{Q}{RT}\right)
\]

(28.16)

where \( k_0 \) is a constant, \( Q \) is an activation energy, and \( R \) is the universal gas constant. Kelly, et al. demonstrated that many of the available fission product release data are adequately represented by this form [30]. By fitting the release data to curves of this form, activation energies could be determined for the limited data available. Meyer noticed that those activation energies were similar to the heats of vaporization, at least for the species with low volatility, and that vaporization was the likely rate-limiting step for those species. He thus used heats of vaporization for each fission product species along with a single empirically derived scaling factor to obtain the constants for the Arrhenius equation in the CORSOR model for all the species with low volatility [29].

For the more volatile species (Cs, I, Kr, Xe, Te), the rate-limiting step in the release involves migration of gas bubbles through the UO\(_2\) matrix. Those data were more prevalent such that the activation energy and pre-exponential factor could be determined directly from prototypical experiments. Thus, a simple fit of experimental data for the volatile species, without regard for their heats of vaporization, was the basis for the Arrhenius-type constants. Notice that migration of these gaseous species through the UO\(_2\) matrix is accomplished by the diffusion of U and O such that all are found to be released at the same rate.

Table 2 gives the pre-exponential factor and activation energy for all species in the CORSOR model.

<table>
<thead>
<tr>
<th>Species</th>
<th>( k_0 ) (min(^{-1}))</th>
<th>( Q ) (kcal/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs, I, Kr, Xe, Te</td>
<td>( 2.0 \times 10^3 )</td>
<td>63.8</td>
</tr>
<tr>
<td>Ag</td>
<td>( 7.9 \times 10^3 )</td>
<td>61.4</td>
</tr>
<tr>
<td>BaO</td>
<td>( 2.95 \times 10^3 )</td>
<td>100.2</td>
</tr>
<tr>
<td>Ru</td>
<td>( 1.62 \times 10^4 )</td>
<td>152.8</td>
</tr>
<tr>
<td>ZrO(_2)</td>
<td>( 2.67 \times 10^8 )</td>
<td>188.2</td>
</tr>
<tr>
<td>Fe</td>
<td>( 2.94 \times 10^4 )</td>
<td>87.0</td>
</tr>
<tr>
<td>Mo</td>
<td>( 0.0 )</td>
<td>--</td>
</tr>
<tr>
<td>SrO</td>
<td>( 4.40 \times 10^7 )</td>
<td>117.0</td>
</tr>
<tr>
<td>Cr</td>
<td>( 4.62 \times 10^4 )</td>
<td>84.5</td>
</tr>
<tr>
<td>Ni</td>
<td>( 5.36 \times 10^4 )</td>
<td>92.2</td>
</tr>
<tr>
<td>Mn</td>
<td>( 5.04 \times 10^7 )</td>
<td>56.8</td>
</tr>
</tbody>
</table>
La group | 0.0\(^a\) | --  
\(^a\)Very low vapor pressure

Most of these species (Kr and Xe being notable exceptions) do not exist in the vapor phase at room temperatures. Therefore, very shortly after evaporating from a hot fuel surface, the fission products condense into liquid or solid particles that are very small and are classified as aerosols (i.e., smaller than 5 microns). These condensed particles have such a large surface-to-volume ratio that they are easily entrained in hot air and other gases in the atmosphere (like smoke). Movement through the atmosphere is then governed by aerosol physics and involves particle collisions, agglomeration, and deposition on surfaces.

**Steam Explosions**

During a severe accident, steam explosions can occur if molten core material comes into contact with water. Steam or vapor explosions take place when two liquids at different temperatures come into physical contact in such a way that the internal energy, transferred from the hot liquid to the cold liquid, causes explosively rapid boiling. The following four stages have been identified for an energetic explosion resulting from a pouring mode of contact [31, 32].

1. At some depth into the water, the melt and coolant form a coarse mixture. Given the very high melt temperature, the melt is surrounded by a vapor blanket so that heat transfer rates are relatively low. During this stage the system is relatively quiescent.

2. There is some initiating event, for example, melt contact at the vessel bottom, that causes a local collapse of the vapor blanket. Local fragmentation of the melt occurs causing high heat transfer rates and pressurization in that region of the mixture.

3. A propagation stage follows during which the local pressurization creates a shock wave that propagates through the mixture causing fragmentation and rapid heat transfer from the melt to the coolant throughout the mixture.

4. An expansion stage follows during which the high-pressure coolant expands, doing work on its surroundings. Damage may result from either the high pressure produced or from the slug impact of materials flowing out of the interaction zone.

After an initiating event, the propagation stage becomes an autocatalytic mixing shock wave followed by an expansion stage. For the melt and coolant in a stratified mode of contact (that is, where the molten core material forms a pool on the lower vessel head with an overlying pool of water), a coarse mixture stage does not exist.

In addition to the mechanical energy that is released, hydrogen production can be significant during a steam explosion because of the large-scale intimate contact between water and core debris, which may contain large amounts of unoxidized zirconium and steel. Debris dispersal and fission product releases can also accompany steam explosions. Powers examined debris dispersal and fission product releases resulting from steam explosions [33]. Figure 28.36 shows the size distribution of particles recovered from steam explosion tests. Less than 1 percent of the
debris mass is in particulate form in the aerosol range. The large particulates that are preferentially formed would fall out of the atmosphere very quickly and would not contribute significantly to the fission product aerosol inventory. However, refractory oxides of fission products like barium, lanthanum, and plutonium, which typically are not released in significant quantities, might have their release enhanced significantly.

Core-Concrete Interactions
Molten core debris that penetrates the lower head of the reactor vessel would fall onto the concrete floor or into a concrete cavity underneath the reactor vessel. The core debris will be very hot and will continue to generate heat because of its fission product inventory. The core debris will also contain amounts of unoxidized metals, mainly zirconium and steel, which can oxidize during the core-concrete interaction releasing more heat. Fission products can also be released during this interaction. Siliceous concrete (silica aggregate, SiO₂) and limestone concrete (limestone aggregate, CaCO₃) have both been studied. Aerosol production with siliceous concrete is very low, and this concrete does not produce as much decomposition gas (H₂, CO, CO₂, and H₂O) as limestone concrete, which is used in some U.S. reactor construction.
Tests at Sandia National Laboratories have investigated the effects of overlying water pools since in some cases the reactor cavity may be flooded and the core debris may be submerged [34]. Experiments with stainless steel melts in water-cooled cavities showed that melt-concrete interactions are little affected by an overlying water pool. Figure 28.37 shows measurements from which this conclusion was reached. An overlying water pool will, of course, have a big effect on retention of aerosols.

Fig. 28.37. Erosion of concrete by molten stainless steel with and without an overlying water pool. (from Ref. [34], see also fig.6.15 of Ref. [27])
References


