Fuel Reliability Assessment Through Radiochemistry and Poolside Examinations

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Ecolabelled printed matter, 441 799

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Contents

1 Abstract

Methods for detecting the occurrence of failure and assessing the condition of leaking fuel rods during and after operation in a nuclear power plant are presented in this report. The on-line methods are based primarily on measurements of the gamma activities of nuclides in the primary coolant and offgas systems. Evaluations of such data involve empirical indices based on activity levels and activity ratios. They can also involve phenomenological models structured to relate the observed nuclide activities with conditions in the leaking fuel. These methods provide information for the operation of a reactor with leaking fuel and for the treatment of failed fuel after discharge. The methods are reviewed in this report from the perspective of assessing fuel reliability and performance.

2 Introduction

Nuclear power plants are capable of operating safely with leaking fuel in their cores and have done so on many occasions. Fuel reliability is increasing so that fewer fuel rods develop leaks that release fission products to their primary coolant systems (defined as failure in this report).In addition, increasing numbers of nuclear power plants (NPPs) are now operating without leaking fuel. So, what is the reason for a report on assessing in-core fuel reliability by means of radiochemistry?

Fuel failures affect three principal areas: radiological safety, core and plant operation, and costs. With increased fuel reliability (reviewed in Section 5.1), the effects of leaking fuel on each these areas are heightened. Reactors are now operating with lower radiation exposure fields due to changes such as:

- The addition of zinc to the primary coolant to entrain gamma-emitting corrosion products in the surface scale (crud)on fuel rods,
- The elimination or minimization of fuel and control assembly materials that produce corrosion products that emit high-energy gamma radiation,
- Reduction in the frequency and number of failed fuel rods and by
- The progressive elimination of fuel particles that were dispersed into the primary systems by fuel failures during previous cycles of operating cycles.

The first two items were driven primarily by the desire to reduce radiation exposure to values that are as low as reasonably achievable (ALARA), though zinc injections also help reduce stress corrosion cracking of primary system components. The second two items also address ALARA concerns. That is, the fission products released from leaking fuel rods increase radiation exposure while the failed fuel is operating. Fuel material that is released from leaking rods, generally following post-failure or "secondary" degradation, has the potential for increasing radiation exposure levels for 10-12 years after the degraded fuel is discharged. While it is now almost unheard of for an NPP to reduce power or shut down because of exposure to site personnel or the surrounding public, increases in radiation exposure due to leaking fuel are more clearly evident and are subject to proactive management to prevent the resulting activity levels from compromising plant operation or adding significantly to operating costs.

The issue of secondary degradation is particularly severe because of the large increases in radiation exposure that such events can cause in the primary coolant and clean-up systems. As discussed in Section 5.2, secondary degradation involves a combination of factors, one of which is the physical loading imposed on fuel cladding by power changes in leaking fuel rods. The knowledge that fuel has failed during an operating cycle and, when possible, the location of the leaking fuel are important considerations in how an affected core is operated. In BWRs, where the risk of secondary degradation is typically greater than in PWRs or VVERs, material improvements combined with the insertion of control blades in and sometimes around cells containing leaking rods has enabled operation without secondary degradation during the 18-24 month operating cycles used by most U.S. plants; [Schneider et al., 2016]. On-line detection and assessment of leaking fuel using radiochemistry are important factors in managing failed fuel rods to avoid forced outages than can come from secondary degradation.

The cost of operating with leaking fuel is significant for both the affected operating plant and the fuel vendor. Costs vary with the nature of the fuel failure and are generally greatest when secondary degradation takes place. Some of the significant costs associated with operating an NPP with leaking fuel have been estimated by Buechel et al., [2014] and are listed in Table 2-1. This table identifies different categories of cost and assigns an estimated minimum and maximum range to the cost in U.S. dollars to the plant operator (utility) and the fuel vendor. The combined costs to the operating utility and fuel supplier range from $1.4M to $16.2M (1.2M to 13.1M €).

Table 2-1: Estimated cost of leaking core outage, [Buechel et al., 2014].

Cost category	PWR or BWR	Estimated cost ($K)			
		Utility		Vendor	
		Min	Max	Min	Max
Diagnosis, tracking, analysis during cycle	Both	5	10	10	20
Cost of sipping during offload	Both	60	100	110	250
Cost of Ultra Sonic Testing (UT) of leaking fuel	Both	40	60	150	250
Cost of repair of leaking fuel	Both	100	150	250	500
Cost due to higher dose rates during outage	Both	60	150	0	0
Cost due to additional maintenance of resin beds	Both	120	180	0	0
Cost due to extended outage	Both	0	3500	0	0
Cost due to unplanned shutdown	BWR	0	3500	0	0
Cost of isolating leaker and suppressing	BWR	40	80	0	0
Cost of redesign of core	Both	75	125	0	250
Value of lost energy in fuel	Both	0	150	0	0
Root cause analysis	Both	60	120	50	100
Corrective action implementation	Both	10	50	20	2500
Root cause field exams	Both	10	30	250	500
Hot cell exams (2)	Both	0	500	0	1500
					ANT International, 2015

The use of radiochemistry for the detection and tracking of leaking fuel will obviously not prevent failures from occurring, but can provide information for mitigating the radiation exposure, operational and economic consequences noted above. Unfortunately, the resulting information is frequently confounded by the presence of more than one leaking rod, by the presence of fissile material in the primary coolant system from leaking fuel rods in prior reactor cycles, during the current cycle or a combination of all of these conditions. As a result, the application of radiochemical data in assessments of fuel reliability has historically involved empirical observations supported by varying degrees of scientific rigor.

With the operational and financial consequences of fuel failure and secondary degradation, the ongoing trend has been to supplement empirical indices, such as changes activity levels and changes in the ratios of radionuclide activities, with methods based more on the physical processes through which fission products and fissile materials are released to the primary coolant system. These phenomenological methods relate activity measurements to terms such as release or escape coefficients. Although intended to better represent the release of radionuclides, the model terms effectively become new empiricisms due to the simplifying assumptions needed in their construction. However, the combination of empirical indices and phenomenological methods is useful in both detecting the occurrence and assessing condition of leaking fuel during reactor operation, particularly when combined with some of the on-line monitoring methods that are now being used.

The objective of this report is to identify methods for detecting failed fuel and assessing conditions associated with leaking rod(s) during operation in an NPP. The report provides background for understanding the capabilities and limitations of the tracking and assessment methods. A brief review of the reactor environments as related to coolant and offgas activities is given in Section 3. Information on the effects of operating conditions on fuel and cladding is presented in Section 4 as background for understanding the bases for the empirical and phenomenological methods. The causes and

characteristics of fuel failures and secondary degradation in modern NPPs are reviewed briefly in Section 5. Sections 6 and 7 review the methods used for detecting and assessing fuel failures during and after irradiation. Finally, the requirements and methods for assessing the hermeticity of spent fuel prior to long-term storage are reviewed in Section 8.

3 Reactor and fuel designs

3.1 General

Water cooled reactors operate with a wide range of core conditions. Typical values are summarized in Table 3-1.Core conditions are provided as background for fuel performance, fission product release and on-line monitoring of coolant activity. The focus of this report is on PWRs, VVERs and BWRs with occasional reference to CANDU reactors. Conditions in RBMK reactors are included for reference.

Table 3-1: Design parameters for water cooled reactors, after [Rudling et al., 2018].

Parameter	Western type PWR	VVER[4] (440/1000) MW	CANDU/ PHWR[1]	BWR	RBMK[2]
Coolant	Pressurized H_2O	Pressurized H_2O	Pressurized D_2O	Boiling H_2O	Boiling H_2O
Fuel Materials (Pressure tube materials)	Zry-4, Optimized ZIRLO[5], DUPLEX[6], M5, Inconel, SS[3]	E110, E635	Zry-4 (Zr2.5Nb)	Zry-2, Zry-4, Inconel, SS	Zr-alloy E110, (Zr2.5Nb)
Average power rating, (MW/m^3)	80–125	83/108	9–19	40–57	5
Fast Neutron Flux, Average, $n/m^2 \cdot s$ (E>1MeV)	6–9E17	5-7E17	1.6-4.3E17	4–7E17	1–2E17
Temperatures, °C					
Average Coolant inlet	279–294	267/290	249–257	200–235 (FW)	270
Average Coolant outlet	313–329	298/320	293–305	280–288	284
Max Cladding (outside surface)	320–350	335/352	330	285–305	290
Steam mass content, %				7–14	14
System pressure, MPa	15.5–15.8	12.5/16.5	10-11	7.0	6.7
Coolant Velocity, m/s	3–6*	3.5/6	3–5	2–5*	3.7
Coolant Chemistry**					
Oxygen, ppb	<5	<10	<10	200-300 (NWC[7])	<20
Hydrogen,					-
ppm	1.5–4	2.6–5.3	0.3–1[8]	FW (H_2) 1-2ppm (HWC without noble metals)	
STP cc/kg	17–50	30–60	3 to 10[8]	FW (H_2) 0.25-0.35ppm (HWC with noble metals)	
Boron (as H_3BO_3), ppm	0–2200	0–1500	–***	–	–
Li (as LiOH), ppm	0.2–6.0	0.05–0.5	0.35-1.4	–	–
K (as KOH), ppm	–	2–20	–	–	–
NH_3, ppm	–	5–25	–	–	–
NaOH, ppm	–	0.03–0.35	–	–	–

*Variation from lower to upper part of the core and from plant to plant.
** Zn in ppb quantities may be added to BWRs and PWRs; Pt in ppb quantities may be added to NMCA BWRs and ppt quantities to OLNC BWRs
*** Not in coolant but in moderator

1.Canadian Deuterium Uranium [CANDU 6 Program Team, 2005]; Pressurised Heavy Water Reactor (PHWR),
2.ReaktorBolshoiMozhnostiKanalov (RBMK),
3. Stainless Steel (SS),
4. VodaVodaEnergo Reactor (VVER),
5. Optimized Zirconium Low Oxidation (Optimized ZIRLO),
6. Cladding tube consisting of an outer soft layer and inner layer with high strength - normally high Sn Zry-4,
7. Normal Water Chemistry,
8. Deuterium

3.2 PWRs and VVERs

Pressurized water reactors have a core of UO_2 or $(U,Pu)O_2$ (MOX) pellets clad in zirconium alloy tubes, with the tubes arranged to form rectangular or hexagonal fuel assemblies. The assemblies are contained in a pressure vessel, through which light water, pressurized by an electrically heated source to 15.5-15.8 MPa, is circulated to an external steam generator. The fuel assemblies in PWRs and newer VVERs do not include external flow channels, so coolant can move laterally as well as axially.

The core entrance temperature is 279-294°C in PWRs and 267-290°C in VVERs. The fuel assembly outlet temperatures are slightly higher in PWRs than in VVERs and can range from 300°C to saturation temperature (~345°C). Initially, PWRs operated under conditions in which the coolant remained in the liquid phase while removing the core energy by forced, convective heat transfer. Currently, most PWRs operate with subcooled nucleate boiling. Some PWRs operate with boiling and modest steam quality in peak power locations. VVERs have historically operated at slightly lower powers; i.e., with liquid-phase heat transfer and little or no boiling.

The major elements of the PWR Reactor Coolant System (RCS) are shown in Figure 3-1and consist of (1) the Reactor Vessel containing an array of fuel rods, (2) two to four steam generators, (3) circulating pumps, and (4) a pressurizer. Water purification and treatment is performed in the PWR auxiliary system, that also contains the Volume Control Tank (VCT), and in case of the Siemens PWRs a degassing system. The major elements of the VVER coolant systems are similar, except that the steam generators are typically horizontal rather than vertical.

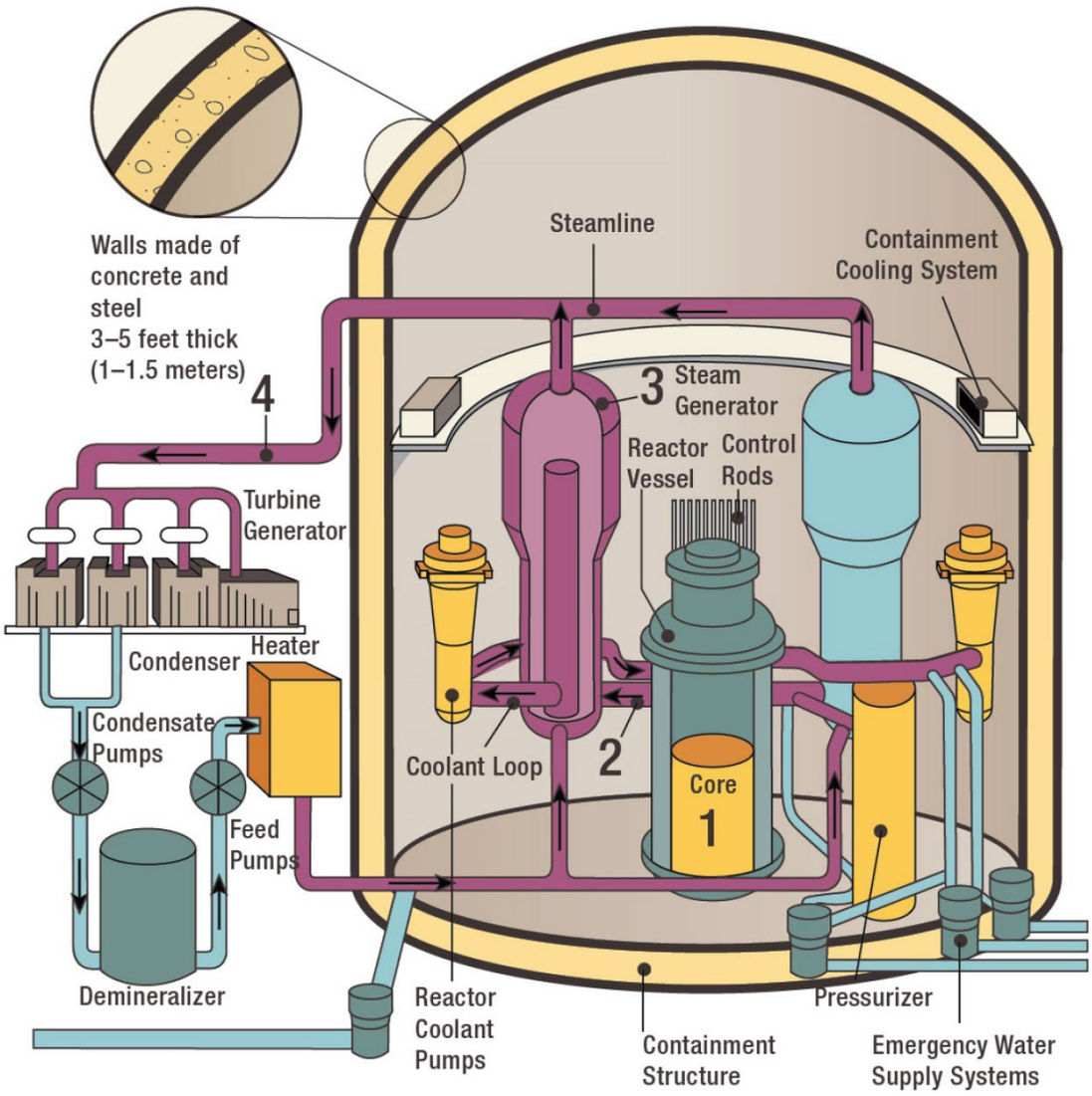


Figure 3-1: Major systems of a PWR nuclear power plant; [https://www.nrc.gov/reactors/pwrs.pdf].

The chemistry of PWR primary coolant systems is important to the behavior of fuel following failure and is reviewed briefly as background for material in subsequent sections.

The water in the primary coolant system is subjected to high radiation intensity in region of the reactor core. This radiation decomposes water to form a number of species including H^+, OH^-, H, OH, H_2, O_2, H_2O_2, e^-aq and $HO2^-$. The major, longer-lived products are H_2, H_2O_2 and O_2. During operation, the addition of hydrogen or NH_3 reduces $O_2$and H_2O_2 concentrations to undetectable levels. Hydrogen is typically kept in the coolant of the PWRs at 2.0-4.5 ppm (25-50 cc/kg). In this range, the corrosion potential at O_2concentrations up to 10 ppb should be between -0.5 to -0.8 V SHE versus Pt (out-of-pile). However, the oxygen potential in the core of a PWR is higher than in a hydrogenated, oxygen free, and radiation free environment even with >2 ppm hydrogen due to radiolysis.

In western PWRs, hydrogen is added and controlled via the cover gas composition of the VCT. Also, some $NH_3$is often seen in western PWRs if the cover gas in the VCT consists mostly of nitrogen or if excess hydrazine is used during start up. Once the ion resins have reached NH_3equilibrium, NH_3concentrations can reach values up to 2 ppm in the coolant. In modern PWRs where sub-cooled boiling occurs, some hydrogen can be stripped off from the water to the steam bubbles so that a local concentrations of oxidizing species are calculated to increase. In VVERs, $NH_3$is added and decomposes radiolytically to $N_2$and H_2. In VVERs hydrogen is kept between 2.6 and 5.3 ppm (30-60 cc/kg) and the NH_3content is typically >5 ppm.

PWRs contain boric acid for reactivity control; the concentration is reduced with time during a fuel cycle, usually being near zero at end of cycle. For a12 month cycle, the initial boron concentration is typically 1,500 ppm but is reduced rapidly over the first few days of the cycle to approximately 900 to 1,200 ppm while the concentrations of xenon and samarium poisons (neutron absorbers) increase to equilibrium levels in the fuel. Thereafter the boron concentration is reduced approximately linearly at ~ 3 ppm/d. For an 18 month cycle the initial boron concentration is typically 1,800 ppm dropping to 1,500 ppm after a few days. Some PWRs use boric acid enriched with the ^{10}B isotope. These plants operate with lower total B concentrations. Because acidic coolant conditions result in an increased attack of the circuit materials LiOH or KOH for VVERs is added to render the coolant slightly alkaline.

3.3 BWRs

In boiling water reactors, the core is contained in a pressure vessel through which water is supplied by a feed pump and is also circulated internally within the reactor pressure vessel by separate pumps, Figure 3-2. BWRs operate at a system pressure of about 70 bars with boiling that produces up to about 70% void fractions in the fuel assemblies. The coolant enters the core with a temperature of 272-278°C and leaves the core as steam water mixture with at the saturation temperature for the pressure at which the reactor operates; i.e., about 285°C. The maximum fuel rod surface temperature varies with heat flux, oxide and crud deposits, and coolant temperature and is 290-320°C for typical mid-to-late life conditions. Steam is separated from liquid water inside the pressure vessel and piped directly to a steam turbine. Liquid water from the steam separators remains in the reactor and circulates back through the core as part of the internal flow.

about 1% and 0.08% after a burnup of 35 MWd/kgU, respectively, and 1.3% and 0.016% after a burnup of 60 MWd/kgU, cf. [Bailly et al., 1999]. As a result, the self-shielding effect of these nuclides tends to overshadow the effect of the initial ^{235}U enrichment.

The fissioning isotopes are of interest because the inventory of fission products that develop with increasing exposure differs slightly between uranium and plutonium. This topic is discussed later in this section and in Section 6.

4.1.3 Fuel chemistry

The oxidation state and the related oxygen-to-uranium (O/U) or oxygen-to-metal (O/M) ratios are arguably the most important chemical properties of UO_2 fuel relative to its in-reactor behaviour. The oxygen potential of nuclear fuel, $\Delta G(O_2)$, affects the thermal conductivity of the fuel as well as diffusion-controlled processes such as grain growth, creep, fission product migration and fission gas release (FGR). It also affects the chemical state and behaviour of fission products, the crystalline structure of the fuel and the oxidation of the inner surface of zirconium-alloy cladding of sound (non-leaking) fuel rods.

As background regarding oxygen potential, it is important to note that the stability of an oxide fuel with respect to the exchange of oxygen increases as$\Delta G(O_2)$ becomes more negative. For example, the oxygen potential of UO_2 is approximately -750 kJ/mol at 350 °C, while the free energy of formation of ZrO_2 is about -980 kJ/mol at the same temperature. This indicates that oxygen from UO_2 is expected to react with Zr to form ZrO_2 if given sufficient time and a favourable diffusion path. Alternately, oxygen is expected to remain in the UO_2lattice rather than reacting with a fission product such as palladium, which has an oxygen potential of approximately -110 kJ/mol at 350 °C. That is, the oxygen potential of $UO_2$is thermodynamically expected to decrease in case of cladding oxidation and to increase with the liberation of fission oxygen in the presence of noble fission products such as palladium. This point is discussed further in the review of fission products and their chemical state later in this section.

The oxygen potential and the ratio of O/U or O/(U+Pu) undergo changes during irradiation due to the liberation of oxygen by fission, the generation of both fission products and conversion products and the reaction of oxygen with uranium, plutonium, fission products and the inner surface of Zr-alloy fuel cladding. During operation, the uranium ions in the as-built UO_2 lattice, primarily U^{4+} ions, are replaced with soluble fission products and with plutonium, which are typically at a valance of 3+[2]. These replacements require the introduction of oxygen vacancies in the fuel lattice, an increase in the oxidation state of some of the remaining U^{4+} ions or a combination of such changes. The net result is that the oxygen potential becomes less negative with increasing exposure relative to the initial value for typical UO_2fuel.

Historically, urania has been considered to shift from a nearly stoichiometric compound ($UO_{2.00}$) to a hyperstoichiometric compound (UO_{2+x}) with increasing exposure; e.g., Δ(O/M) ~ 0.0013 per atomic percent (at.%) burnup through approximately 5 at. %. Such increases are moderated by the oxidation of fission product molybdenum at local pellet temperatures below about 1300 °C; [Kleykamp, 1979, 1985]. As an example, increase in the oxygen potentials of UO_2, $(U,Gd)O_2$ and $(U,Pu)O_2$ are shown relative to burnup in Figure 4-8.

[2]Plutonium is predicted to change from Pu^{4+} from Pu^{3+} to maintain electrical neutrality with the introduction of soluble fission products at a valance of 3+ and the oxidation of some of the remaining U^{4+} to U^{5+} or U^{6+} [Olander, 1976].

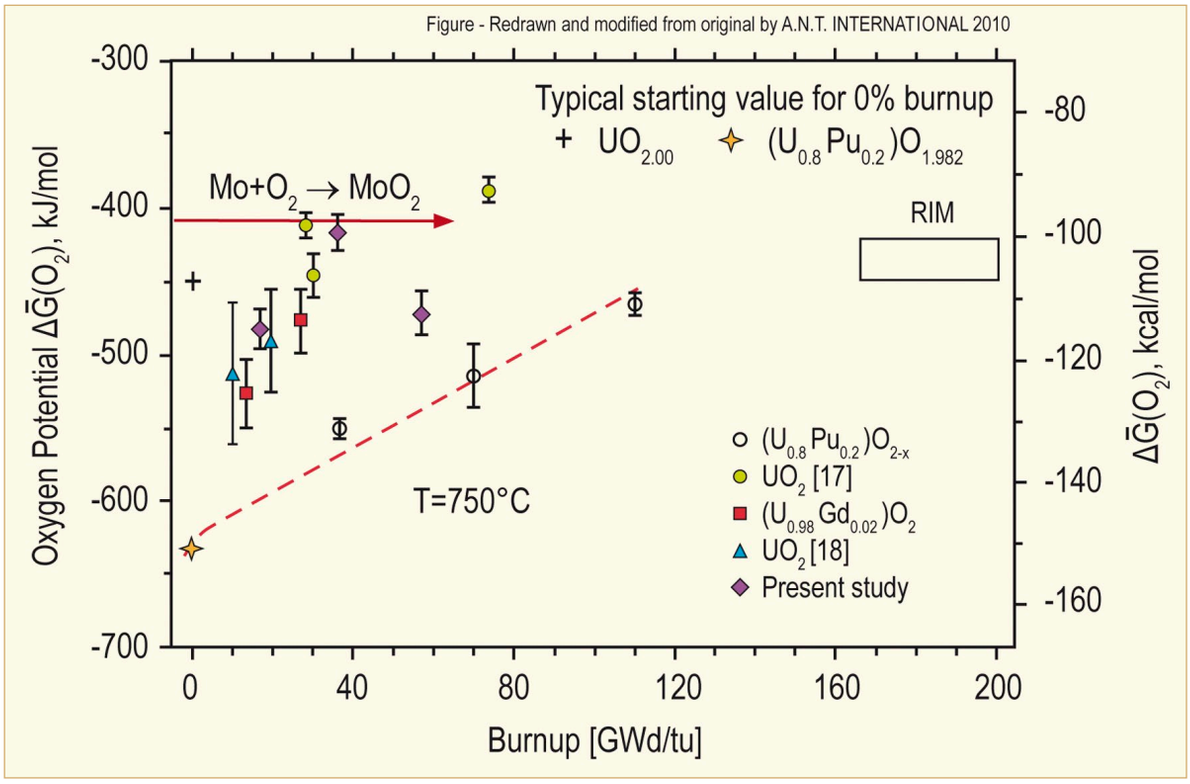


Figure 4-8: Change in oxygen potential at 750 °C relative to burnup for UO_2, $(U,Gd)O_2$ and $(U,Pu)O_2$ fuel, [Matzke, 1995].

Other investigations have shown the evolution of O/M ratio and fuel chemistry to differ from a uniform, linear relationship. As an example, measurements of high burnup fuel reported by Walker et al., [2005], which are summarized in Figure 4-9, show that oxygen potentials become larger (less negative) than the as-fabricated value, larger than the value for the Mo/MoO_2 reaction at high burnup and vary by a greater amount and in the opposite sense across the pellet radius than in the studies at lower exposure (Figure 4-8). The overall increase in oxygen potential in the work of Walker relative to those of Kleykamp and Matzke is attributed partially to the higher exposure of Walker's fuel; i.e., up to 102GWd/MTU pellet average burnup versus ≤ 70 GWd/MTU. Subsequent investigations suggest the evolution of oxygen potentials and the chemical states of fission product depends on the combined interaction of the fuel, fission products and cladding; see the reviews of recent thermochemical investigations given in Section 2.4 of [Adamson et al., 2016] and [Adamson et al., 2018].

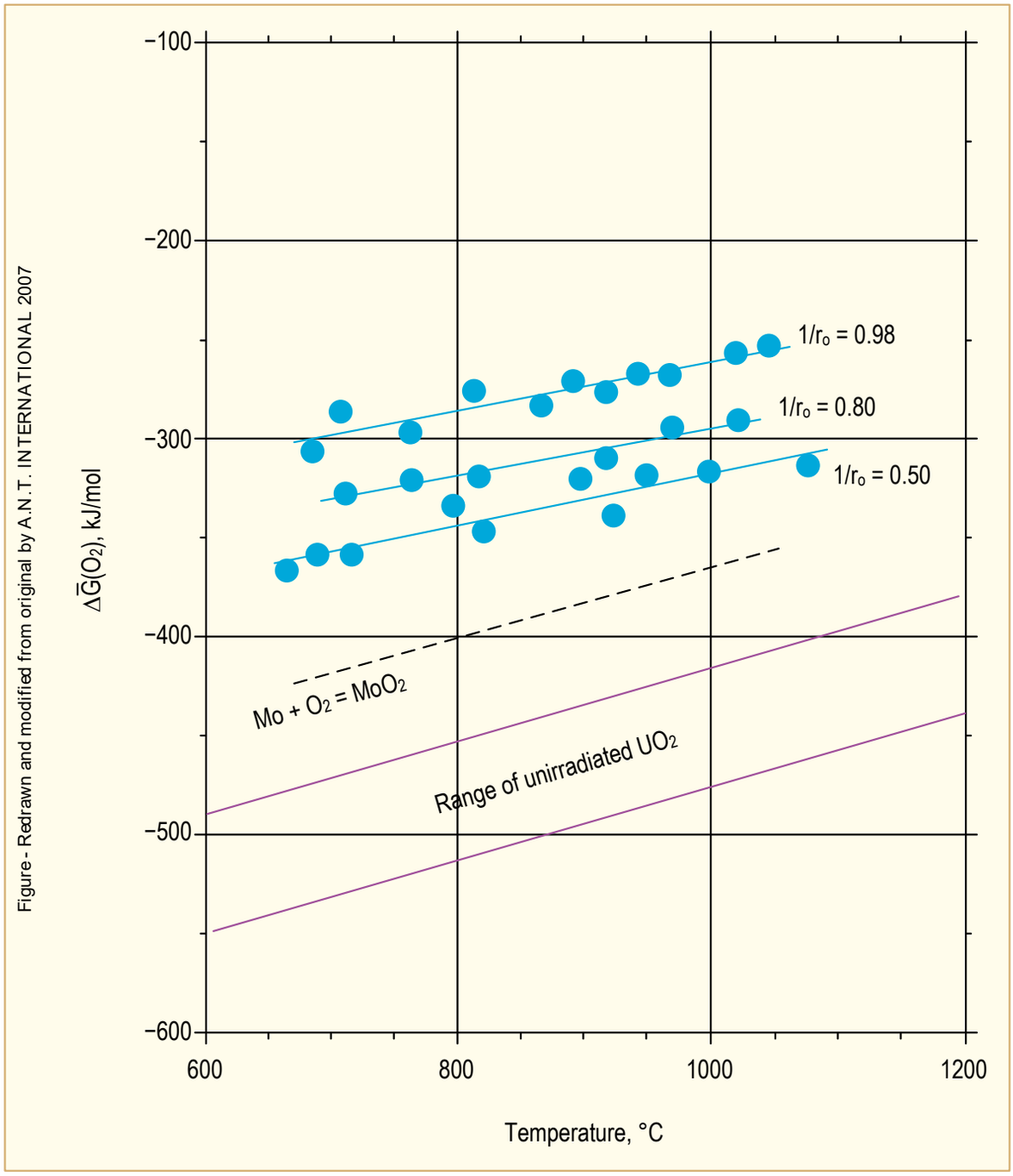


Figure 4-9: Oxygen potential of high burnup fuel relative to temperature, after [Walker et al., 2005].

For the purpose of this report, the noteworthy points are that the O/M ratio of fuel pellets evolve with exposure and, thereby, affects the chemical state of fission products and the structure of the fuel pellets. Additional information on the effects of oxygen potential on fuel chemistry and fuel behaviour is given by Olander, [1976], Patterson & Garzarolli, [2010] and in Section 2.4 of the ZIRAT21 and ZIRAT23 Annual Reports; [Adamson et al., 2016] and [Adamson et al., 2018], respectively.

It should be noted that the release of fission products from the fuel pellets to the free volume within a fuel rod (rod intraspace) is strongly influenced by the fuel oxygen potential in addition to factors such as local pellet temperature, confining (hydrostatic) stress and fission rate. All of these factors except the fission rate change with failure and the presence of water in the rod intraspace. These changes

adversely affect radiochemical models that attempt to relate the release of fission products from leaking fuel rods to the release in sound rods.

4.1.4 Fission products

As noted in the previous sections, the composition and structure of oxide fuel pellets change during irradiation due to the generation of fission products and the manner in which these products interact with the fuel matrix. That is, about 200 stable or long-lived fission product atoms are produced fuel per 100 fissions. The cumulative distribution or chain yields of these fission products are shown in Figure 4-10 relative atomic mass, with yields clustered in low and high mass distributions. As shown in this figure, yields vary slightly with the fissionable isotope (^{235}U or ^{239}Pu) and, to a lesser extent, with the energy of neutrons in water moderated reactors.

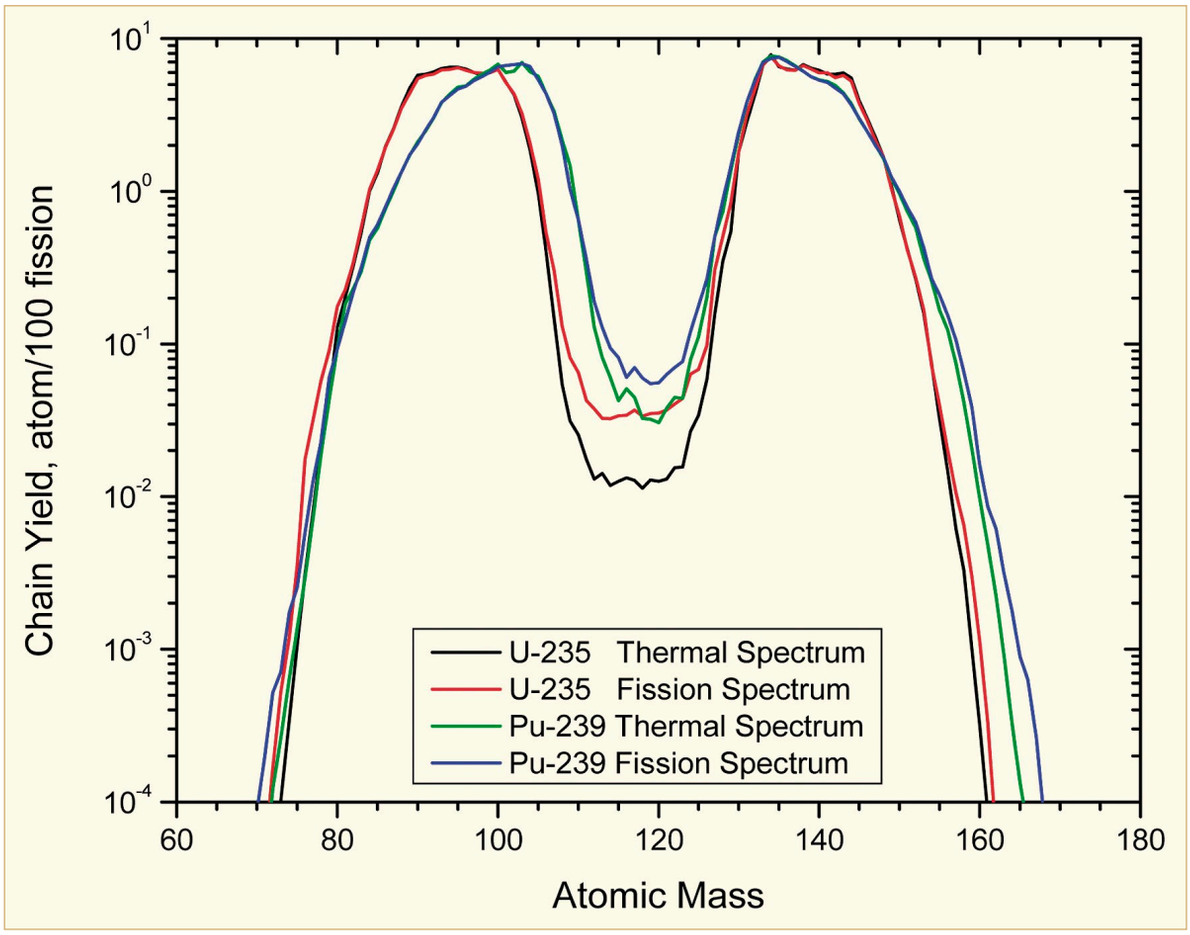


Figure 4-10: Fission yields by atomic mass, fissile isotope and neutron energy, [England & Rider, 1994].

An example of the fission product production in LWR fuel rods is given in Table 4-1. The production rates are the average, net generation rate of each nuclide during operation; i.e., µg of fission product per gram of the combined mass of actinides and fission products per 10 GWd/MTU. Data in this table are net inventories ≥1 ppm as calculated with the ORIGEN-2.1 computer program with operation at 18.5 kW/m core average power to 60 GWd/MTU, which is close to the time-average heat generation rate of typical BWR and PWR fuel. They represent the steady-state fission product inventories during in-reactor operation, without loss due to post shut-down decay. The cumulative rate of fission product production is approximately 0.1% per GWd/MTU.

Table 4-1: Fission generation in LWR fuel, based on ORIGEN-2.1, after [Olander, 1976] and [Kleykamp, 1985].

Fission product	Production rate wt. ppm per GWd/MTU	Notes	Probable valance
Noble gas			
Kr	10.9		0
Xe	157.5		0
Volatile			
Br	0.6	Elemental vapour or insoluble compound	0 or 1-
Rb	10.1	Elemental vapour or insoluble oxide	0 or 1+
Te	14.7	Elemental vapour or metallic inclusion	0
I	7.0	Elemental vapour of insoluble compound	0 or 1-
Cs	84.6	Elemental vapour or insoluble oxide	0 or 1+
Metal			
Mo	98.6	Metallic inclusion or insoluble oxide	0, 4+
Tc	21.4		0
Ru	74.7		0
Rh	9.1		0
Pd	45.4		0
Ag	2.1		0
Cd	4.3		0
Sn	2.8		0
Sb	0.9		0
Oxide inclusion			
Se	1.7		4+
Sr	25.9		2+
Zr	105.5	Oxide in fuel matrix; also in alkaline earth oxide phase	4+
Nb	0.6	Oxide in fuel matrix or soluble oxide	4+, 5+
Ba	45.6		2+
Oxide soluble in cation sub lattice			
Y	13.7		3+
La	36.6		3+
Ce	79.0	Soluble oxide; also in alkaline oxide phase	3+, 4+
Pr	32.7		3+
Nd	115.7		3+
Pm	2.1		3+
Sm	22.9		3+
Eu	6.0		3+
Gd	4.4		3+
Total	1037.2		

Helium is not included in Table 4-1 but is produced along with ^{3}H and other light nuclides by ternary fission. Helium is also produced during and after irradiation by (n, α) reactions on ^{16}O (minor production in water-moderated reactors) and by α-decay of certain actinides. The combined production rate of helium is 0.2 to 0.3% per fission of U and Pu, [Federici et al., 2007]. The production of helium and its release from fuel pellets to the rod intraspace during irradiation is small; e.g., <1% for typical UO_2 rods and 5-6% for MOX rods, [Lanning et al., 2005].Although helium has historically not been used in on-line analyses of fuel reliability, the release of the inventory of helium in the rod intraspace at the time of fuel rod failure is now being used to detect fuel failure is some BWRs. This topic is discussed in a later section.

As indicated in Table 4-1, the physical states and chemical forms of the fission products vary with temperature and oxygen potential and are typically divided into classes based on these forms; i.e.,

1) Noble gases: The noble gases Kr and Xe, which are essentially insoluble in the fuel matrix and can form either intragranular (within grain) voids or bubbles, intergranular (grain boundary) bubbles or be released from the fuel pellets to the free volume of the respective fuel rod.
2) Volatiles: Elements such as Br, Rb, Te, I and Cs, that exist as gases at high temperatures typical of the interior of an operating fuel pellet or as liquids or solids at the cooler exterior of a pellet.
3) Metals: Elements such as Mo, Ru, Pd, and Tc that form metallic precipitates and insoluble metallic alloys.
4) Insoluble oxides: Elements such as Zr, Ba and Sr that form oxides and are insoluble in the fluorite lattice comprising UO_2 and MOX fuels.
5) Soluble oxides: Elements such as Y, La and the rare earths that are soluble in the cation sublattice.

Volatile fission products can exist as gases at temperatures above their boiling points, as liquids or solids at lower temperatures. When the local pellet temperature exceeds the respective vaporization temperature of the volatile fission products, they behave as a gas and contribute to void formation and the gas release process similar to xenon and krypton. Under such conditions, the volatile fission products also diffuse from high temperature regions near the pellet centreline to lower temperature regions toward the outer pellet surface or escape from the hot interior and condense in the cooler pellet-cladding gap. It should be noted that at temperatures relevant to LWR fuel rods, iodine is a gas during operation (boiling point = 184 °C) while cesium is a gas at the interior of fuel pellets when operating at moderate-to-high power (671 °Cboiling point) or a liquid toward the outer pellet surface and pellet-cladding gap (28 °C melting point). The physical state of these fission products enable their escape from the fuel matrix, release to the rod intraspace and migration to the pellet-cladding interface.

For the purpose of assessing fuel reliability, the fission products of interest are primarily the noble gases, the water-soluble fission products in the rod intraspace at the time of failure and those that are released to the intraspace subsequent to failure. These fission products typically comprise the gaseous isotopes of xenon and krypton and the water-soluble fission products iodine and cesium. As discussed in Section 6, other soluble fission and decay products as well as helium are also used in some circumstances.

4.2 Evolution of fuel pellets due to effects of temperature and irradiation

Radiation affects the crystalline structure of UO_2 mainly through the interaction of charged, high energy emissions with the ions comprising the fluorite lattice in addition to the formation of fission products as noted in the previous section. This is particularly true for the fission fragments formed during operation which are massive on an atomic scale and highly energetic, i.e., atomic mass numbers in the range of 75 to 160 and initial kinetic energies of 70-100 MeV. This energy is dissipated by collisions with ions in the fuel matrix and with atoms dispersed among fuel particles over a cylindrical path that is 7-10 μm in length and 150-200 Å in diameter. Since the energy required to displace an atom from its normal lattice site is approximately 20-40 eV (see [Olander, 1976]), each fission

fragment displaces a large number of atoms through direct collisions with lattice ions (identified as primary knock-ons) and through collisions between the affected ions and other lattice ions (identified as higher order knock-ons). The result is that over 20 000 uranium ions can be affected in the collision-displacement cascade created by each fission fragment. Lattice ions can also be displaced by kinetic and electronic interactions with other high energy emissions from fission and decay. These displacements depend on the nature and energy of the emissions, relative masses and related factors such as displacement cross-sections and energy transfer functions. The net result is that the ions comprising UO_2 typically undergo many thousand displacements during their operating life in a water reactor.

The displacements due to fissioning are important in radiochemical assessments of fuel reliability because they drive fission products located within about 10 μm of free surfaces from the fuel matrix to the rod intraspace or directly into the coolant in the case of fuel that is exposed to or distributed in the coolant. This process, identified as recoil or knockout release, varies with fission rate and is independent of temperature.

Fuel in commercial power reactors is used in the form of circular cylinders; i.e., fuel pellets. These pellets are fabricated by sintering compacted powders of UO_2, urania with burnable nuclear absorbers such as gadolinia or erbia, or mixtures of urania and plutonia to densities of 94-98% of the respective theoretical values; e.g., 10.96 gm/cc for stoichiometric UO_2.Most pelletizing and sintering operations now produce pellets that are free of the small pores that contribute to pellet densification during irradiation and are also essentially free of pores or collections of pores that are connected to pellet surfaces; i.e., open porosity.

Even with the high densities of modern fuel pellets, the thermal conductivity of the oxide is low so that fissioning and heat generation creates high temperatures at the center of the fuel pellets and steep gradients across the pellet radius. Typical conditions for LWR fuel at the beginning of life are shown in Figure 4-11.The lower power curve in this figure (20 kW/m) corresponds to the approximate average linear heat generation rates over the life of modern PWR and BWR fuel. The upper power curve (40 kW/m) is close to the peak operating power-temperature distribution in current BWR fuel. Higher powers and fuel temperatures are encountered during some of the "anticipated operating occurrences" (AOOs) to which PWR and BWR fuel can be subjected. The peak, centerline pellet temperature is bounded in almost all fuel designs by the value for fuel melting; i.e., ~ 2800°C for $UO_{2.00}$ at the beginning of life.

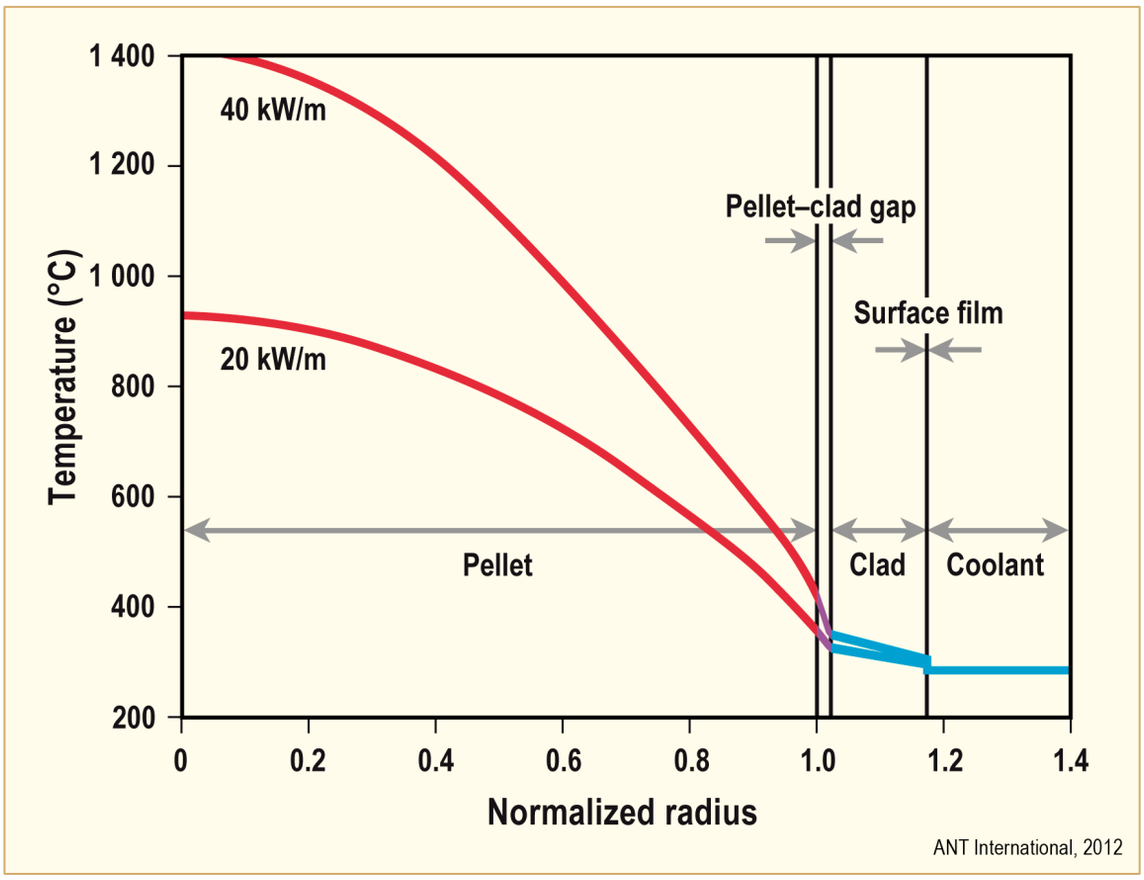


Figure 4-11: Temperature relative to radial position and linear heat generation rate of a Zircaloy-clad UO_2 fuel rod at the beginning of irradiation, after MATPRO relationships [INEL, 1993], [Siefken et al., 2001].

Fuel rod temperatures vary strongly with heat generation rates and also with conditions that depend on burnup and burnup history. Temperature calculations are performed as part of the design process by means of computer programs that address the thermal and mechanical behaviour of fuel and cladding materials and of integral fuel rods. Such calculations are also performed during irradiation by some reactor operators to evaluate the expected effects of operating maneuvers on fuel performance and reliability. Details of such thermal-mechanical programs are beyond the scope of this report and can be found in the documentation of publicly available programs such as the FRAPCON code of the U.S. NRC; see, for example,[Geelhood & Luscher, 2015; Geelhood et al., 2015] and [Luscher et al., 2015]. Aspects of operational behaviour that are relevant to the assessment of fuel reliability by means or radiochemistry involve the production and release of fission products and are discussed below.

The release of fission gas from inside fuel grains, where the gas is generated, to the free volume inside a fuel rod is one of the more complex in-reactor processes. Gas release depends strongly on fuel temperature and burnup. It also depends on fuel characteristics such as O/M ratio, the ratio of open pellet surface area to pellet volume, grain size, microstructural changes due to radiation damage and temperature and on pellet restraint (hydrostatic pressure) due to pellet-cladding mechanical interaction. The processes and interactions that affect fission gas release are shown in Figure 4-12, with those related to the release of fission products shown in red.

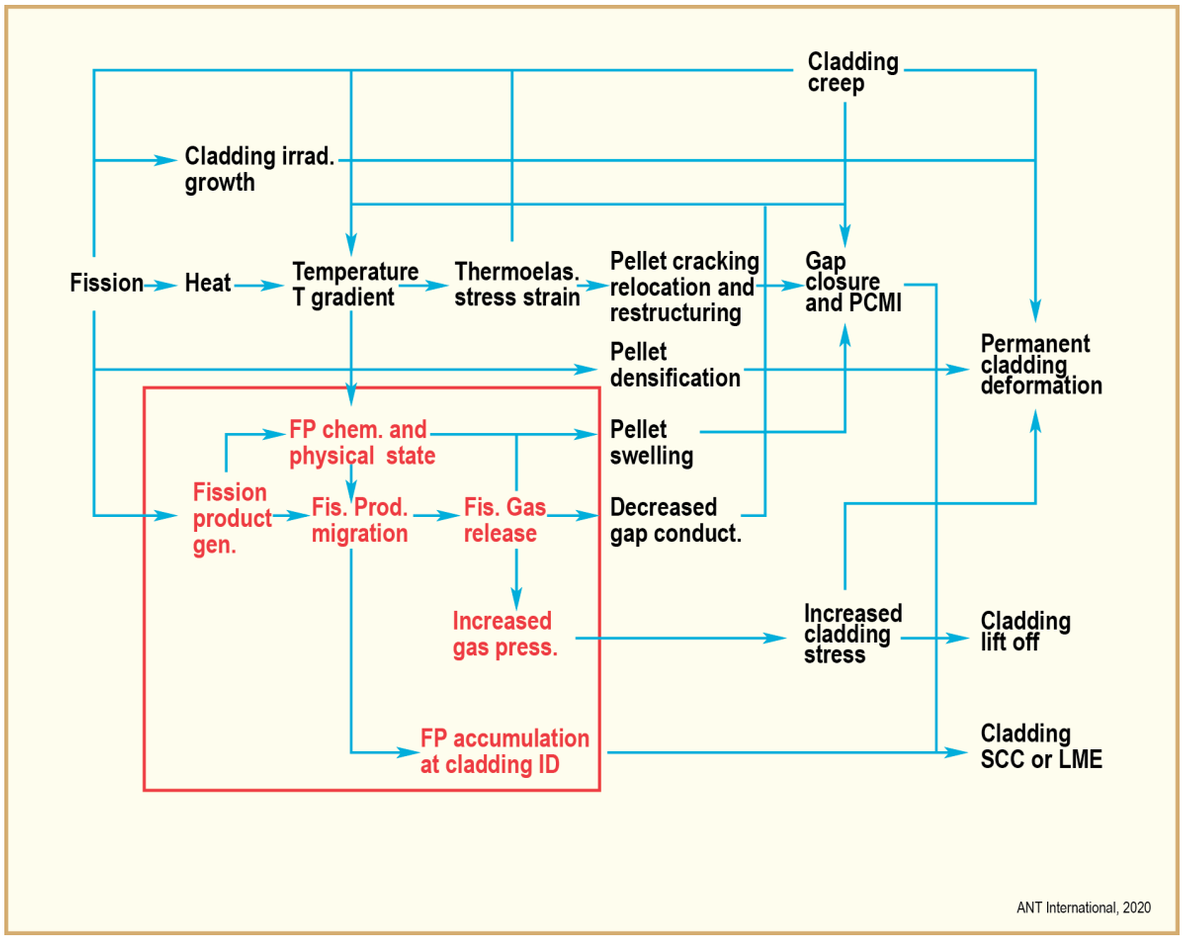


Figure 4-12: Schematic description of processes and interactions during fuel irradiation with factors relevant to radiochemical assessments of fuel reliability shown in red.

From data in Table 4-1, xenon and krypton are produced at a combined rate of approximately 168 wt. ppm per GWd/MTU, which corresponds to 29.2 l/GWd at Standard Temperature and Pressure (STP). This gas contributes to fission product swelling of the fuel pellets, degradation of thermal conductivity of the pellets, degradation of the thermal conductance of the pellet-cladding gap and to increased pressure inside of fuel rods. The production, transmutation and decay of ^{135}Xe also affects the nuclear behaviour of the fuel and the reactor core because of its large production rate (6.54 atoms per 100 fissions of ^{235}U) and large neutron absorption cross-section (σ_a = 2.65E6 barns for thermal neutrons) and moderate half-life (9.10 hr). Overall, fission products are produced at a rate of ~0.1% per GWd/MTU.

The heavy atoms of xenon and krypton released to the free volume decrease the thermal conductivity of the helium fill gas in the rod intraspace. When the pellet-cladding gap is open, the lower thermal conductivity of the mixture of helium, xenon and krypton increases the temperature of pellets at a given rate of heat generation. Such temperature increases lead to more gas release from fuel that operates at temperatures high enough to activate the thermal diffusion of fission gas; i.e., the centre regions of fuel pellets at moderate-to-high power (Figure 4-11).

In addition to the thermal effects, the larger amounts of xenon and krypton in the free rod volume that come with higher burnup increase the pressure of the gas mixture in the rod intraspace. Depending on the design of a fuel rod and its operating history, fission gas release can lead to internal gas pressures that exceed the coolant pressure and alter the release of fission products into the primary coolant system at the time a leakage path develops through the cladding.

Data on fission gas release of LWR fuel rods under steady state operation are shown relative to Linear Heat Generation (LHGR) in Figure 4-13and relative to burnup in Figure 4-14. The data of

5 Fuel failure causes and characteristics

As noted earlier, fuel reliability in this report refers to the maintenance of fuel rod hermeticity during and after in-reactor operation. That is, reliability is defined as the absence of a breach of the cladding wall, end plugs or end plug welds that releases fission products or fuel material from within an irradiated fuel rod. The broader sense, reliability also involves other measurable design and safety factors such as strain, corrosion and dimensional stability and other calculated criteria such as fuel temperature, margins to heat transfer limits and transient response to postulated (design basis) accidents. However, the focus in this report is on fuel rod breaches that release radionuclides from an affected fuel rod. The review that follows id divided into discussions of the causes and characteristics of primary failures and of post-failure (secondary) degradation.

5.1 Primary failure

5.1.1 Observed fuel failure mechanisms and rates

The causes of fuel failures in water-cooled reactors vary slightly among reactor types but, with the exception of plant or reload-related upsets, tend to be similar for a type of NPP within a country due to the commonality of fuel designs, materials and operating conditions. The mechanisms by which fuel rods have failed in the past and are continuing to fail are summarized in Table 5-1 for PWRs and Table 5-2 for BWRs. These tables are based on 5-year intervals because the number of fuel failures has become small and leads to large fluctuations in the calculated rates when considered on an annual basis. The information available on fuel failures in VVER and PHWR/CANDU reactors does not permit the construction of similar tabular summaries, but is reviewed below.

Note that the mechanism percentages in Table 5-1 and Table 5-2are based on the failures for which the root cause has been determined. The sum of the failure proportions for the known mechanisms is 100% in each time interval. The proportions in the “Unknown/undetermined” category are based on the total number of failures in each time interval, which causes the total for each time interval to exceed 100%; i.e., 100% for the known causes plus the percentage of unknown causes. This treatment differs from typical failure tabulations that are based on the total number of failures, but provides estimates of the failure fractions due to each cause that are unbiased by the unknown category. Normalizing the known failure proportions for each mechanism to the column total will give the customary proportions relative to the total number of failures in the respective time interval; e.g., normalizing the 57.9% fraction of observed PWR failures due to grid-to-rod fretting in the 2011-2015 interval to the column total, 140.2%, gives 41.3% due to this cause relative to the total number of failures.

Table 5-1: Summary of world-wide PWR fuel rod failure mechanisms for the interval 1987-2015; [IAEA, 2019].

Failure mechanism	Proportion of fuel failures for each mechanism (%)						
	1987-1990	1991-1994	1995-1998	1999-2002	2003-2006	2007-2010	2011-2015
Grid-to-rod fretting	16.6	42.7	73.0	87.6	78.0	58.4	57.9
Debris fretting	55.6	46.7	14.5	7.1	13.9	19.5	33.7
Fabrication issue	20.8	6.7	9.5	3.4	7.2	16.4	8.4
Crud or corrosion	0.0	0.0	2.2	1.5	0.0	0.0	0.0
PCI/SCC	0.0	0.0	0.0	0.0	0.9	5.7	0.0
Handling	2.8	3.8	0.8	0.4	0.0	0.0	0.0
Baffle jetting	4.2	0.0	0.0	0.0	0.0	0.0	0.0
Unknown/undetermined	50.0	48.0	26.7	14.6	33.2	28.4	40.2

Table 5-2: Summary of world-wide BWR fuel rod failure mechanisms for the interval 1987-2015; [IAEA, 2019].

Failure mechanism	Proportion of fuel failures for each mechanism (%)						
	1987-1990	1991-1994	1995-1998	1999-2002	2003-2006	2007-2010	2011-2015
Debris fretting	17.5	50.5	39.6	53.4	32.2	58.6	66
Crud or corrosion	42.3	4.4	46.8	23.1	52.9	3.9	13.2
PCI/SCC	27.7	34.1	9.9	11.5	14.2	36.2	18.9
Fabrication issue	10.1	11	3.7	11.5	0.7	1.3	1.9
Handling	2.4	0	0	0	0	0	0
Unknown/undetermined	1.62	32.5	24	27.8	13.6	29.2	26.4

For PWR fuel rods, failures during the past 10 years have been the result of grid-to-rod fretting, debris-induced fretting, manufacturing defects and a limited number of pellet-cladding interaction (PCI) events. The leading causes of PWR fuel failures continue to be grid-to-rod fretting and debris fretting. Failures due to crud, corrosion, baffle-jet effects and handling have not been observed. The fraction of unknown causes is in the range of 30-40% during this interval.

For BWR fuel rods, failures during the past 10 years have resulted from debris fretting, crud, corrosion, PCI and manufacturing defects. The leading causes of BWR fuel failures are debris fretting, PCI, and crud or corrosion. The fraction of failures due to unknown causes is in the range of 10-30% for the past 10 years.

The information on fuel failures in VVER fuel is sparse. From tabulations by the IAEA, the leading, recurrent cause of failure is debris fretting; [IAEA, 2010], [Inozemtsev &Onufriev,2013] and [IAEA, 2019].Instances of failure due to grid-to-rod fretting, crud or corrosion and manufacturing defects, including primary hydriding are also reported.

Failure data on PHWR/CANDU fuel indicate the leading mechanisms are debris fretting and fabrication defects (primarily weld issues). Isolated failures are reported to have been caused by pellet chips (PCI due to chipped pellets), micro cracks in fuel cladding and handling damage.

The fuel rod failure rates are shown by year and country for each reactor type in Figure 5-1 through Figure 5-4. These failure rates are based on the number of fuel assemblies found to be leaking after discharge by sipping and/or post-irradiation inspection combined with the average number of leaking rods observed historically in each type of fuel relative to total number of discharged assemblies and the number of fuel rods in each assembly. The numbers of failed rods in each leaking fuel assembly assumed in the IAEA, [2019] assessment are 1.6 for PWRs and VVER-1000 reactors and 1.1 for VVER-440s, CANDUs and BWRs.

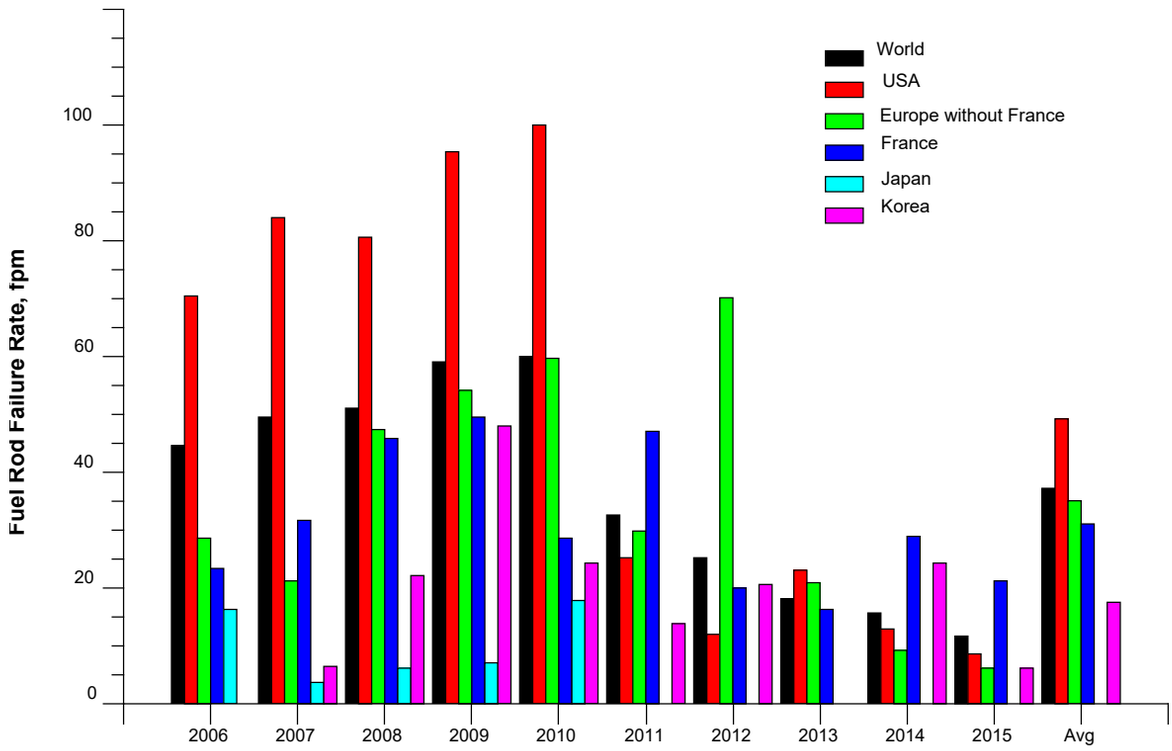


Figure 5-1: PWR fuel rod failure rates relative to year of fuel discharge and country, [IAEA, 2019].

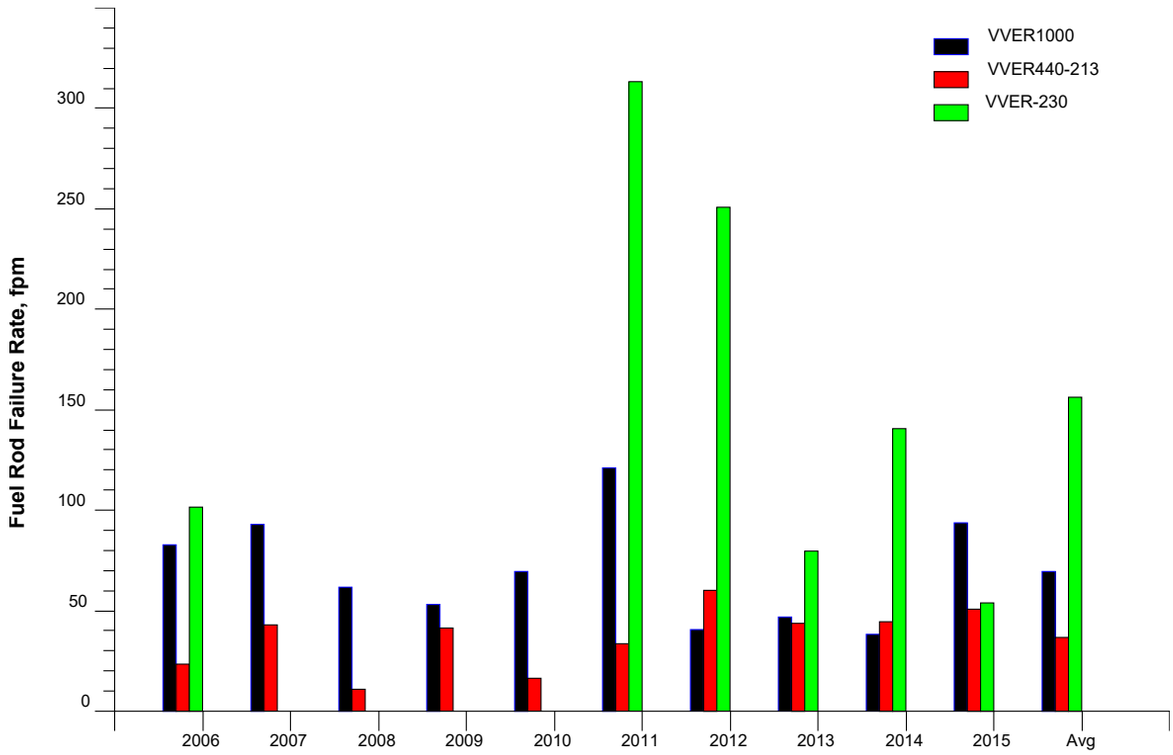


Figure 5-2: VVER fuel rod failure rates relative to year of fuel discharge for all countries, [IAEA, 2019].

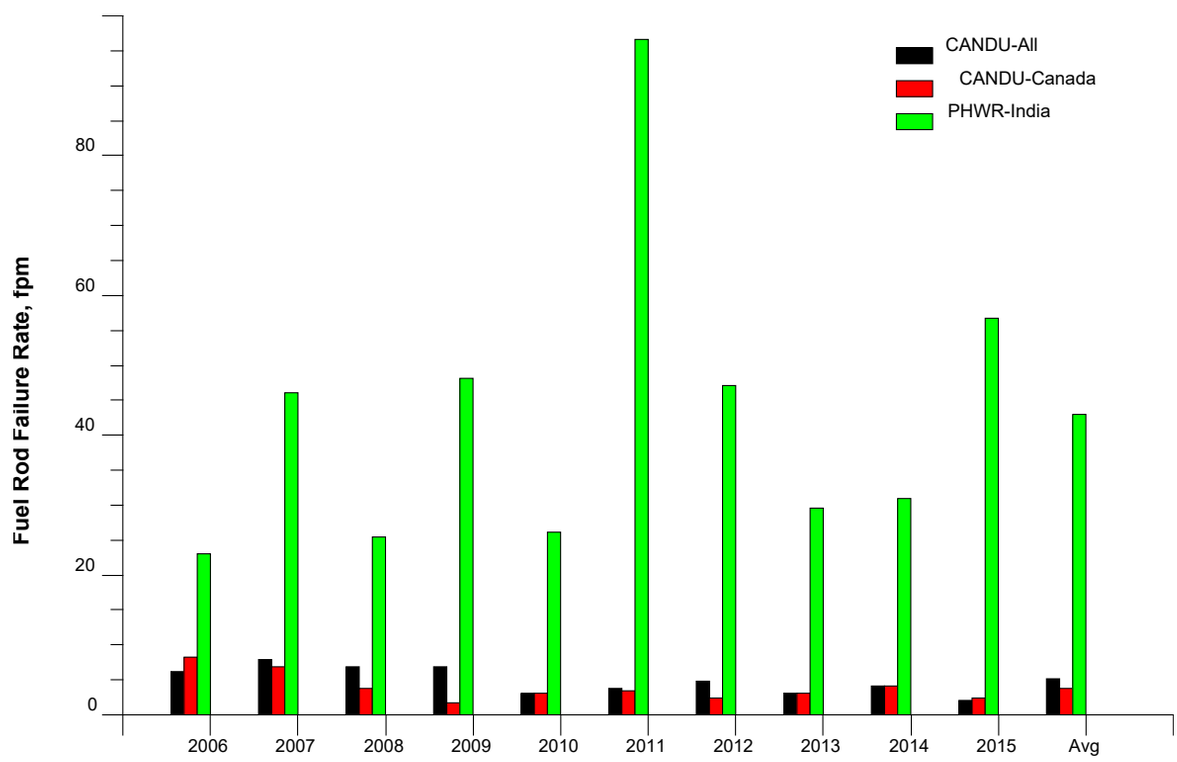


Figure 5-3: CNADU and PHWR Fuel rod failure rates relative to the year of fuel discharge, [IAEA, 2019].

interval to remove failed fuel assemblies with the objective of eliminating the risk of large residual contamination from tramp uranium. More recently, the risk of degradation and residual contamination has been reduced by the use of corrosion resistant liners in BWR fuel and by the use of monitoring and leaker management programs. As a result, forced and voluntary outages are now less common.

An extensive review of secondary degradation is given in Appendix A of "The Effects of Hydrogen on Zirconium Alloy Performance, Volume II", [Strasser et al., 2008b]. This source should be consulted for additional information.

5.2.1 Secondary degradation of failed BWR fuel

Armijo, [1994] and Hüttmann et al., [1997] have shown that axial splits in a failed Zr-lined fuel rod occur only in conjunction with a power ramp of intermediate to high burnup rods. Thus, if a failed rod is not subjected to a power ramp, no axial split will form. It should be pointed out however, that power ramps must be performed in the reactor for other reasons and consequently, it is typically impossible to run a plant without any power ramps – the range of power control via feedwater heating and flow control tends to be insufficient with most BWR core loadings to allow moving control blades only at low power.

On the other hand, the formation of transversal breaks is not correlated to power ramping and can result during operation of a failed rod during constant power;[Sihver et al., 1997]. Sometimes, it seems that lowering of the reactor power to such an extent that the lower part of the rod may be filled with water, *water logging*, such as during a cold shut-down, may enhance the risk of transversal breakage upon return to full power. The transversal break tendency is similar for 8x8 and 10x10 fuel designs. It is also noteworthy that the transversal break tendency decreases with burnup level.

Obviously, axial cracks can occur along the whole length of the rod while transversal breaks mostly form in the bottom part of the failed rod at some minimum distance from the primary defect, Figure 5-48 and Figure 5-49. However, there are cases when a transversal break has formed at the bottom and top part of a rod with a primary failure in the middle part of the rod.

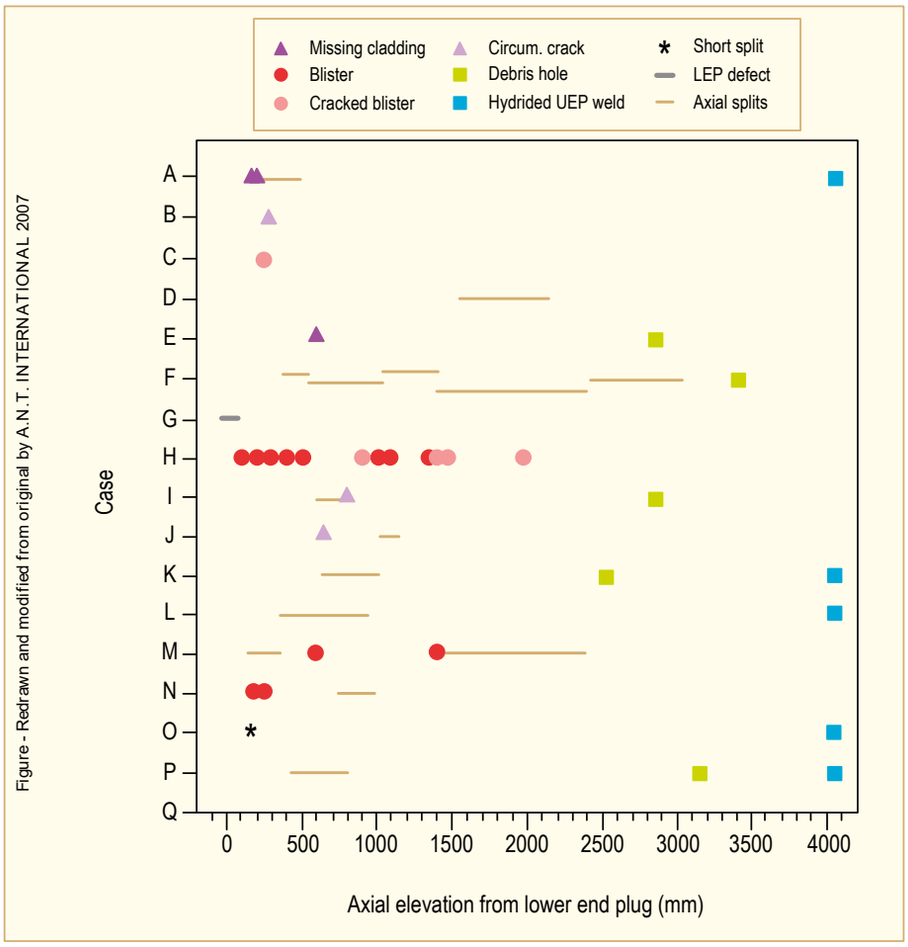


Figure 5-48: Location and dimension of secondary damage in failed *GE* (now Global Nuclear Fuel (*GNF*)) and ANF (now AREVA) rods. *UEP* = Upper End Plug, *LEP* = Lower End Plug,[Harbottle et al., 1994].

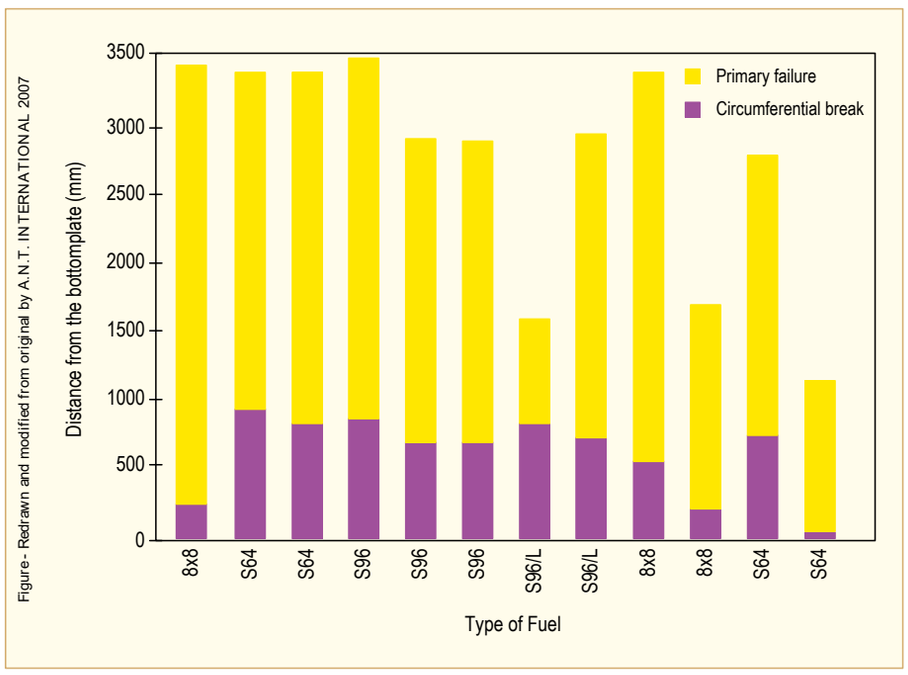


Figure 5-49: Locations of primary failures and transversal breaks, on the fuel rods, for all cases where the exact location of the primary failure has been identified, [Sihver et al., 1997].

5.2.1.1 Transvers break formation

If a failed rod has low to intermediate burnup, one may expect first, that the overall pellet-cladding gap is large and second, that this gap is much smaller at the lower part of the rod due to the downshift in power profile for these rods in PWRs or upshift in BWRs. Now provided that, at a specific axial rod elevation, the ratio of hydrogen to steam partial pressure is large enough and the protecting clad inner surface oxide is thin enough, massive hydrogen ingress into the cladding may occur at this location.

When the hydrogen solid solution solubility has been exceeded, precipitation of hydrides can form *hydride blisters* that can subsequently grow into massive hydrides throughout the cladding thickness along its whole circumference. Since zirconium hydrides are very brittle, the cladding zone that is completely transformed into zirconium hydride will be very brittle and may easily fracture even during operation at constant power.

When the steam penetrates the failed rod through the primary defect, hydrogen is being produced in the pellet cladding gap by the steam oxidation process of the Zr alloy material forming an oxide at the clad inner surface. Zirconium oxide is an excellent barrier towards hydrogen ingress into the cladding. The released hydrogen through clad inner surface oxidation will result in a hydrogen partial pressure, p_{H2}, that will increase with distance from the primary defect. Simultaneously with this increase in hydrogen partial pressure, a corresponding decrease in steam partial pressure, p_{H2O}, occurs with increasing distance from the primary defect. Thus, there exist a ratio between the hydrogen and steam partial pressures at rod each elevation.

At (p_{H2}/p_{H2O}) ratios below the critical ratio, $(p_{H2}/p_{H2O})_{critical}$, the cladding inner surface oxide thickness does not impact the secondary hydriding tendency as long as the oxide remains protective and consequently no massive hydriding will occur, irrespective of the oxide thickness.

A prefilm thickness to about 1-2 μm is postulated to significantly increased the incubation time for transverse fracture and that a prefilm thickness between 2 to 3 microns is the optimum to get the longest incubation times. However, too large prefilm thickness offers poor hydriding resistance.

As mentioned earlier, a prerequisite for a transversal break is that the fuel cladding is locally embrittled around the whole circumference of the cladding at a specific elevation of the cladding. Such an embrittlement is a result of massive local hydrogen pickup at the cladding inner surface resulting in zirconium-hydride, $ZrH_{1.66}$ formation. The zirconium-hydride phase is very brittle and is easily fractured at low stress levels in the hydrided zone. It seems that scram, shutdown and cold shutdown may increase the risk of getting transversal break of a failed rod.

The oxidation of the cladding inner surface results in some homogeneous hydriding of the cladding with a hydrogen pickup fraction[5]ranging from 0.2 to 0.4; [Olander & Yagnik, 1997]. The events leading to massive hydriding, Figure 5-50, where the hydrogen pickup fraction may be locally more than 1, results from the (p_{H2}/p_{H2O}) exceeding the critical value as shown schematically in Figure 5-51.

[5]This fraction describes the fraction of the hydrogen released through the corrosion reaction between water/steam and zirconium alloy that is picked up by the zirconium alloy material.

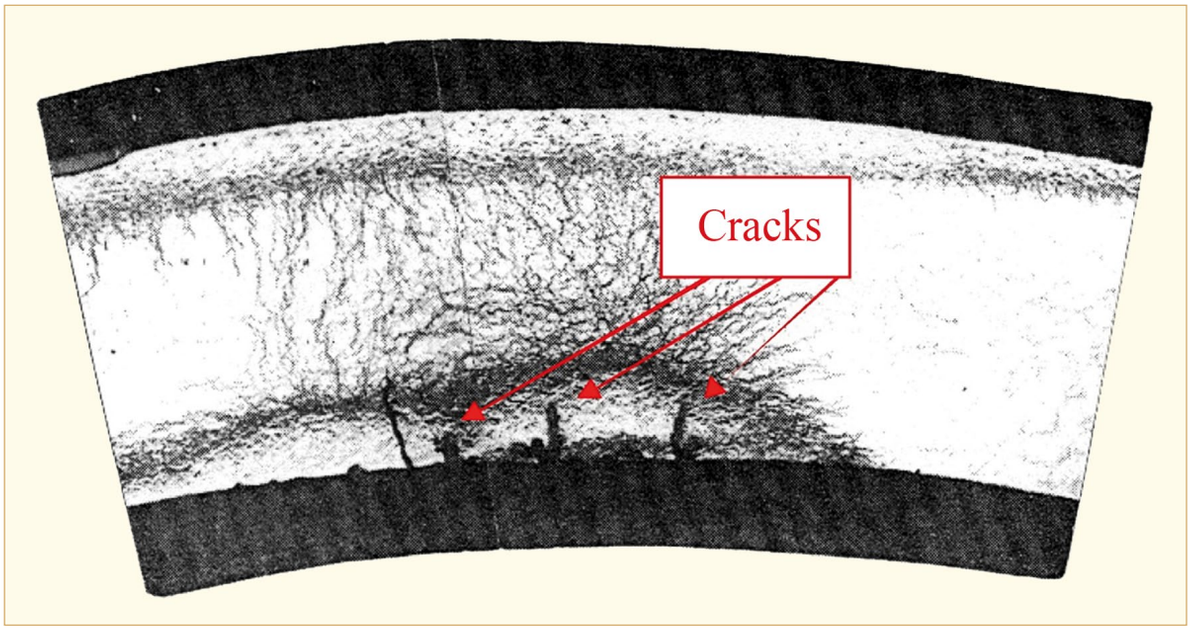


Figure 5-50: Metallographic cross-section at a sunburst. Cracks can be observed in the centre of the hydride blister, [Garzarolli et al., 1979].

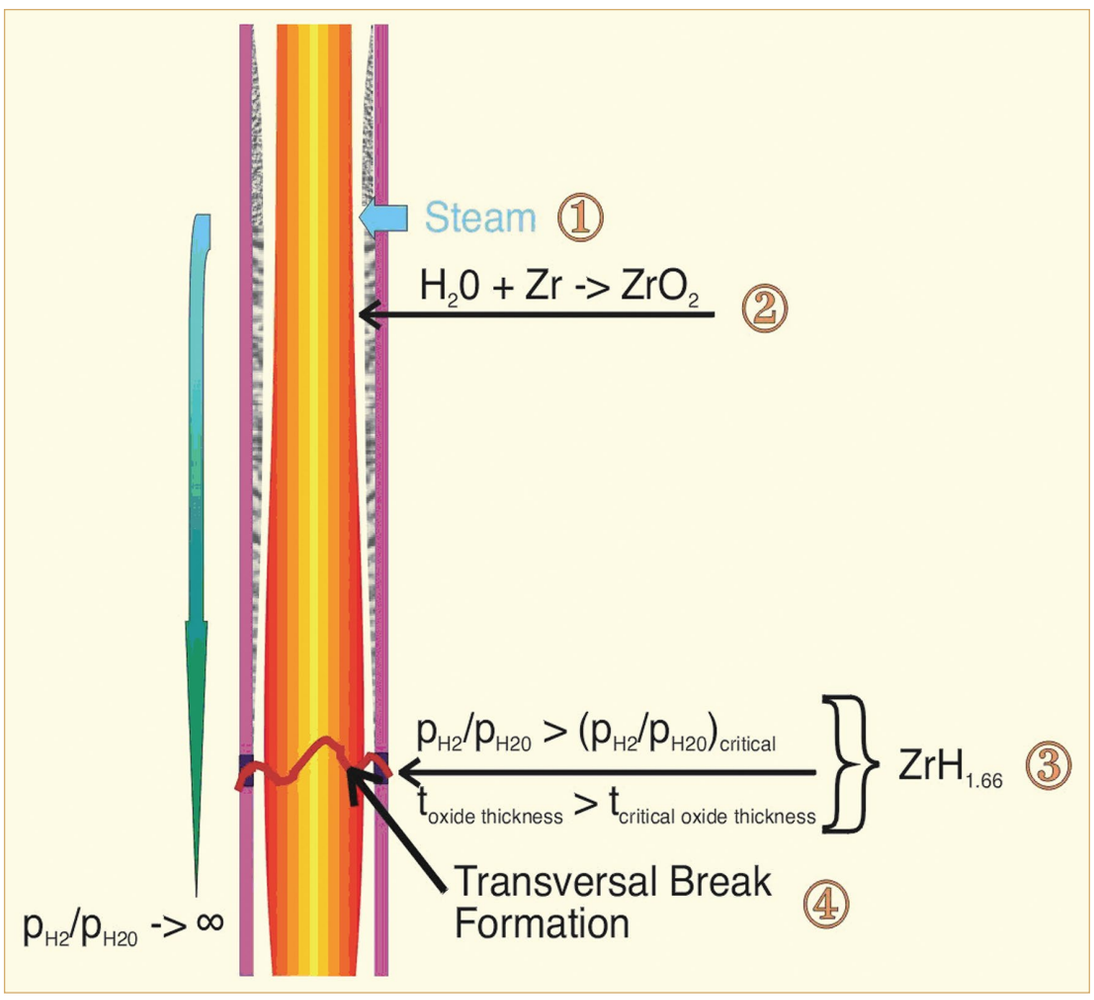


Figure 5-51: Schematics showing the events resulting in transversal break formation. The numbers in the figure relate to the sequence of the different events that may lead to a transversal break.

6 Online assessment of fuel reliability using radiochemistry

6.1 Overview of section

On-line monitoring of fuel reliability is performed at different levels of detail. As occurrence of a failure during a cycle corresponds to a sudden presence (or an increase) of fission product concentrations in the primary circuit, measurement and assessment of primary activity levels relative to radiological limits is a high-level form of monitoring. The "fuel reliability indicators" (FRI) of industry organizations such as the Institute of Nuclear Power Operations (INPO) and the World Association of Nuclear Operators (WANO) are another form of high-level monitoring. These indices are based on the activity of iodine isotopes in the primary coolant and offgas activity for BWRs, with corrections for the effects of core power, coolant cleanup rate and background activity (e.g. distributed sources coming either from previous failures, or from contamination by tramp uranium still present on primary circuit walls or rod cladding; discussed below). Such industry indices are designed to indicate the presence of leaking fuel in operating reactors, for comparisons with license limits or internal specifications, and comparisons among utilities and reactors. Other, broader-scope monitoring methods also indicate the presence of leaking fuel and can provide varying degrees of additional insight as to the number of leaking rods, their average linear heat generation rate, burn-up, the condition of such fuel and its expected evolution during operation. The focus of this section is on the later class of monitoring methods.

6.2 Effects of failure on in-core behaviour

6.2.1 Primary failure

Primary failure results from the formation of one or more leakage paths through the cladding, end plugs or welds of an affected fuel rod. A brief review of the types of primary failures relevant to modern water reactor fuel is given in Section 5.1.The initial leakage paths due to such failures are typically small enough that only the fission products that are in the gaseous state or that are soluble in reactor coolant are released to the primary coolant system; fuel material is typically retained in leaking rods. As a result, activity release rates generally increase at the time of failure, as the stored inventory of long-lived gaseous and soluble fission products (mainly Xe, Kr, I, Cs) are released, and then continue at rates that depend on the local power in the leaking rod and of the effective size of the leakage path. In the absence of factors that increase the size of leakage paths, like cladding corrosion or local hydriding, the release rates from primary failures vary with the power of the leaking fuel rod(s) in many cases[6] and can be limited by reducing the local power in the leaking fuel, preferably at or near the location of the leak. Such action is generally limited to BWRs as it requires local variations in power by means of control blade movements with activity measurements either before and after each blade move or continuously during the moves to identify the leaking fuel. Although the magnitude of activity release rates, or the total number of leaking rods (when a generic cause affects a lot of rods) can pose issues with respect to exposure, radiation limits for workers and safety authorizations, the significant characteristic of primary failures is that the activity release returns to pre-failure values when the leaking fuel is removed from the core.

6.2.2 Secondary degradation

Secondary degradation, reviewed in Section 5.2, involves changes to a fuel rod subsequent to its initial failure that enlarge the effective size of the leakage path or produce a second (and often larger) path and thereby increase activity release rates. Although the fission products released directly from a rod

[6] Small cladding breaches have been observed to be sufficiently restrictive that only long-lived fission gas is released to the primary system. In such cases, the amount of release and its composition are essentially independent of power. Failed fuel rods with this behaviour have been identified as "weak leakers" and are discussed later in this section.

can be large enough to pose activity issues, the most significant factor associated with secondary degradation is the dispersal of fuel into the primary system.

As noted earlier, fuel particles that are released from a damaged rod and transported by the coolant, add to the activity of the primary coolant system while the source rod is in-core. When fuel particles are released to the primary coolant and circulate through the core at each coolant rotation, a supplementary release of fission products occurs (mainly very short half-life isotopes). Moreover, the fuel particles from the failed rod, fuel washout, is quickly deposited on tubes and walls in contact with the coolant. Part is deposited under neutron flux, mainly on the cladding of neighbouring rods. Part also deposits on the steam generator tubes or PWRs and VVERs (they offer the largest contact surface) or on the steam separators of BWRs. So, the deposited fuel particles subjected to a neutron flux can continue to release fission products after the leaking / damaged rod is removed from the core. Large increases in the activity release rates during and after secondary degradation can adversely affect plant operation and almost always add to the cost of running a nuclear plant as noted in Section 2.

Although practices have changed over time, emphasis on operating nuclear plants well within their allowable limits have caused utilities to assess the risk of degradation and sometimes undertake mid-cycle outages to remove leaking fuel if large increases in activity could occur. Such outages are more frequent in BWRs because of their open primary systems and because the oxidizing nature of BWR coolant is conducive to the post-failure corrosion and hydriding that leads to the degradation. By comparison, both the likelihood of secondary degradation and its consequences are smaller in PWRs because of the closed primary system and the reducing nature of the coolant. Adverse effects arise in PWRs, but tend not to be as severe as in BWRs and are not generally severe enough to force mid-cycle outages.

An important aspect to successful NPP operation is detecting fuel failures as soon as they occur, assessing the condition of the leaking fuel and estimate the activity release evolution. It is specifically important to minimise fuel washout since the resulting tramp uranium will increase the background radiation level in the core for up to 10 years. These topics are reviewed in the sections that follow.

6.3 On-line monitoring

6.3.1 Summary of physical processes activated in a leaking fuel rod

The composition and concentration levels of fission products in the primary coolant and offgas streams changes with the occurrence of a leak in a fuel rod and varies with both the condition of the leak and its operating conditions. Failure by any of the mechanisms discussed in Section 5 produces a leakage path between the interior of the affected rod and the primary coolant system. At the time of perforation, coolant enters into the rod if the internal gas pressure, due to manufacturing filling with helium and the fission gases released out of the fuel material, is under the primary coolant pressure. Moreover, in some countries, a safety specification on the rod design imposes that its inner gas pressure shall be systematically under the primary pressure, even at the end-of-life for the most powerful rod. In these cases, the pressure discrepancy provokes ingress of primary water in the inner free volume of the rod (upper or lower plenum, fuel-clad gap). In other countries, the pressure of gas in the rod intraspace is allowed to exceed the coolant pressure and gas can be released to the coolant at the time of failure. Depending on the local temperature at the defect level during rod operation, different situations can occur with intraspace pressures less than the coolant pressure:

- If the local temperature is lower than the saturation temperature at the primary pressure, the entered water remains in a liquid phase and fills progressively the accessible volume. This is the case for failures occurring at rod extremities (e.g. plug welding, failure due to a debris blocked under the bottom grid…) which operate at lower linear power (or without power). The water quantity can be large, especially if it fills a plenum.
- If the local temperature on every surface is higher than the saturation temperature at the primary pressure, the entered water is transformed into steam. This is the general case along the fuel stack length (except a few tens centimeters at upper and lower extremities of PWR fuel or the lower extremities of BWR fuel). Then steam propagates axially through the free volumes of the rod and can condense at rod extremities, for the same reason as above.

- When the radial temperature gradient authorizes at the same elevation both a liquid phase (e.g. on the inner surface of the clad) and steam (e.g. on the fuel outer surface), situation is more complex because a vaporization-condensation mechanism can be activated and maintained, as long as the water still enters. This mechanism, called "two phase regime", can be agitated and can provoke successive sudden ejections of vapor out of the rod. It concerns a PWR rod with a moderate local linear power (e.g. at high burn-up) and is often the common case when the reactor is operated at partial power.

After a while depending on the leak size, pressure equilibrium is reached between the inner free volume of the rod and the primary coolant pressure. This can last several days (and even few weeks) if the leakage path is small or if the fuel-clad gap is closed (as for high burnup rods). Due to the fuel cladding creep down and fuel rod swelling the pellet-cladding gap decreases with burnup and at about 20MWd/kgU there is fuel – cladding contact. This equilibrium can be quickly reached for end-of-life failures, i.e. when the inner pressure is already near the external pressure. After this step, gas or liquid starts to escape into the primary system because the rod inner pressure becomes higher than the external pressure. At constant power level and for a primary failure, the fission product release rate is continuous and presents an approximately constant level. It is due mainly to three main processes:

- steam continues to be formed as long as liquid is present,
- water and steam are radiolyzed by neutrons, gammas and recoil fission products, and finally
- fission gases continue to escape from the fuel materials and contribute also to the pressure increase.

The generation and release of fission products from the fuel pellets proceeds somewhat as described in Section 4 except that temperatures, oxygen potentials, physical states and chemical forms change due to the entry of water into the rod inner free volume.

The escaping gases include a mixture of xenon and krypton, with a routing gas which is probably hydrogen. Fission gases have an isotopic composition that depends on their source, rate of production and release, half-lives, decay chain and time. If the leakage path is large, or if the local linear power is moderate (i.e. water can remain in a liquid phase), coolant can enter into the rod intraspace and dissolve soluble fission products. In this case, gases and soluble fission products are released to the coolant by a combination of diffusion and mass transfer. This progression is shown schematically in Figure 6-1.As the size of the leakage path increases, the rate at which coolant flows into and out of a leaking rod also increases. Similarly, the time needed for radionuclides to escape to the coolant decreases and the concentrations of soluble nuclides such as iodine and cesium in the primary coolant increase. The ratio of short half-life isotopes in the total escaped inventory increases also. Concurrent with the release of radionuclides from the rod intraspace to the primary system, additional fission products are being generated in the fuel and released to the rod intraspace.

Except for the "two phase regime" presented above, which can present high / very high release rates at constant power, the release rate of fission products is moderate at constant power for the most of the cases, because there is no pressure difference upstream and downstream the defected rod. However, in the long term, the water present in the rod should disappear if there are no power changes: radiolysis is probably the most effective mean for that and formed hydrogen provides the inner pressure increase for fluid release. Oxygen is then consumed, but at a lower rate, primarily by forming an oxide on the cladding surface and to a lesser extent, with the fuel (formation of oxide phases). Finally, without water ingress, the rod void should become dry.

However, under transient conditions that involve changes in power or coolant pressure, previous pressure equilibrium is completely changed. Fission products in the rod void can be released in a spike-like manner, by vapor ejection. Such release, frequently provide the first indication of failure in reactors that rely primarily on the activity of soluble radionuclides such as iodine isotopes in the primary coolant to indicate failure.

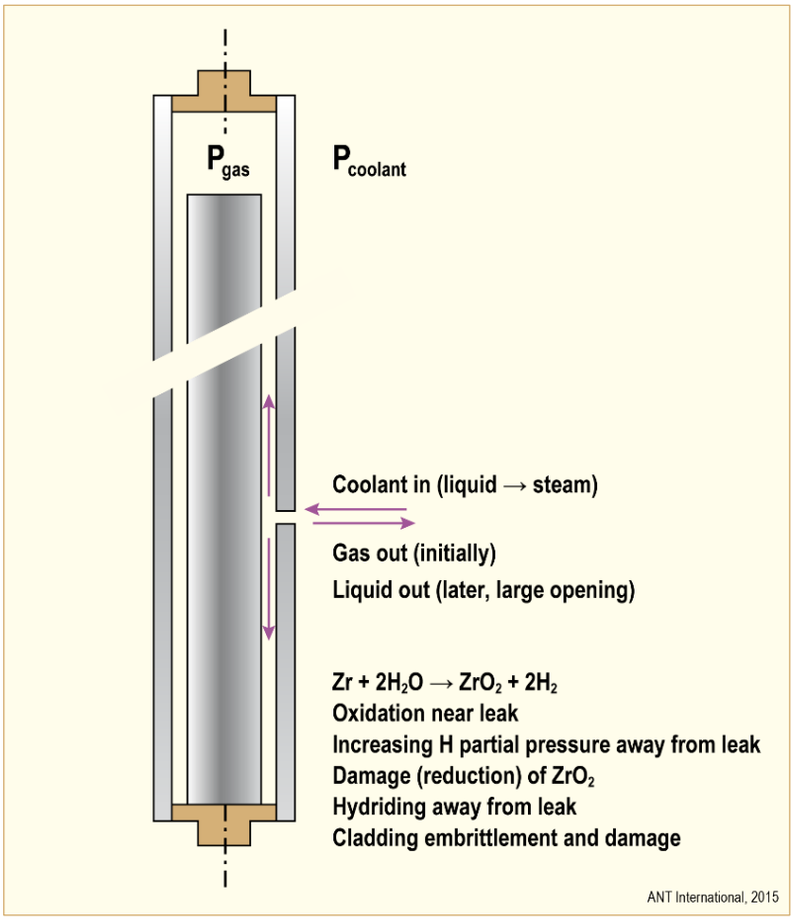


Figure 6-1: Schematic view of the exchange of primary coolant and radionuclides in a leaking fuel rod.

6.3.2 On-line monitoring of the primary coolant activity

Monitoring fuel reliability typically involves sequential, radiochemical measurements of the primary coolant and gas extracted from the primary coolant or the steam line (in BWRs). Measurements of the gross (total) beta or gamma activity of the primary coolant or offgas by means of sensors built into the reactor system provide early indications of fuel failures and secondary degradation, but are generally insufficient for monitoring fuel reliability. In specific cases (as when fissile material is suspected of being released), alpha spectrometry can be carried out on a coolant sampling to detect presence of transuranic isotopes.

Detailed assessments are usually based on spectrometric measurements of coolant gamma activity to determine the composition and concentrations of radionuclides (fission products, but also activation products), with respect to their magnitudes and trends relative to time, burnup and plant maneuvers. In most reactors, the sources of such data are discrete samples of the primary coolant and offgas (BWR), or coolant and extracted-gas samples (PWR) that are processed in a gamma activity measurement laboratory (counting room). In some cases, and more recently, on-line measurements are being performed continuously with flowing samples routed through detectors, such as a γ-spectrometer, that are located near or connected to the coolant or offgas streams; e.g., [Parrat et al., 1991], [Sihver et al., 1999] and [Olsson et al., 2017]. Both monitoring methods are complementary: the on-line measurement deals with short half-life isotopes (approximately from a few tens of minutes to one day), because they dominate the gamma spectrum in terms of number of rays and background level. On the contrary, sampling is counted after different decay times (from few hours to few days) and middle- and long half-life isotopes are successively detected, as short half-life contributors progressively disappear from the gamma spectrum by radioactive decay. The radionuclides of interest include xenon and krypton isotopes in the coolant and gas samples, iodine and cesium isotopes in the

primary coolant, and other nuclides that appear generally in the coolant with a large leak or after secondary degradation; e.g. Te, Ba, La, Rb, Sr, Ce, Np, etc.

An example of monitoring fuel reliability in a BWR is shown in Figure 6-2. This figure is a time-based plot of reactor power and the γ-activities of a long-lived fission gas, ^{133}Xe (5.25 d) and the sum of six isotopes of xenon and krypton with relatively long half-lives (called "Sum-of-Six" and discussed later). In this case, the reactor was operating at full power when spikes in the ^{133}Xe and Sum-of-Six activities indicated the failure of a fuel rod. These activity data came from gamma spectrometry measurements of periodic samples. The initial spikes are due to the release of fission gases stored in the void volume of the leaking rod prior to failure.

In Figure 6-2, variations in the relative magnitudes of ^{133}Xe and the Sum-of-Six activities represent changes in the state of the leaking fuel rod subsequent to failure. The large initial increase in ^{133}Xe activity was due to its longer half-life and the larger inventory of this isotope in the rod inner free volume. The decrease of ^{133}Xe activity relative to the Sum-of-Six activity reflects depletion of the initial inventory of the long-lived nuclide and the progression toward a mixture more representative of the production rates of the six gaseous nuclides; i.e., greater concentrations of the shorter-lived isotopes. The subsequent increases in activity while at constant or decreasing power resulted from what was ultimately found to be the formation of a long axial crack in the fuel cladding and the transport (washout) of fissile material through the crack, see Figure 6-3.

Power suppression tests were conducted to locate the fuel assemblies with leaking rods. The leaking assemblies were confirmed by sipping and removed during a refueling outage at the normal end of the operating cycle. The plant began the next cycle with elevated coolant and offgas activity due to fuel material that had been transported from the leaking rod and subsequently deposited on core internals and fuel rods that were in the core when washout occurred and then continued operation in the next reactor cycle.

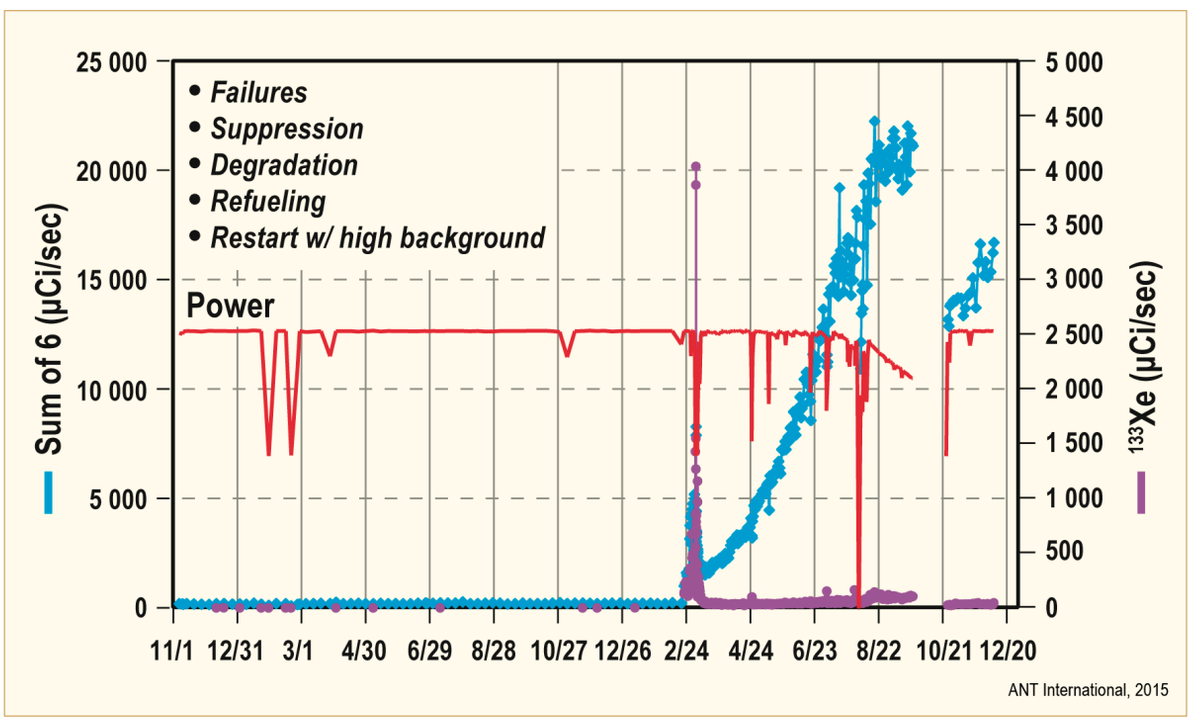


Figure 6-2: Example of a BWR fuel failure occurrence and resulting changes in offgas activity, after [Yeager & Schneider, 2005].

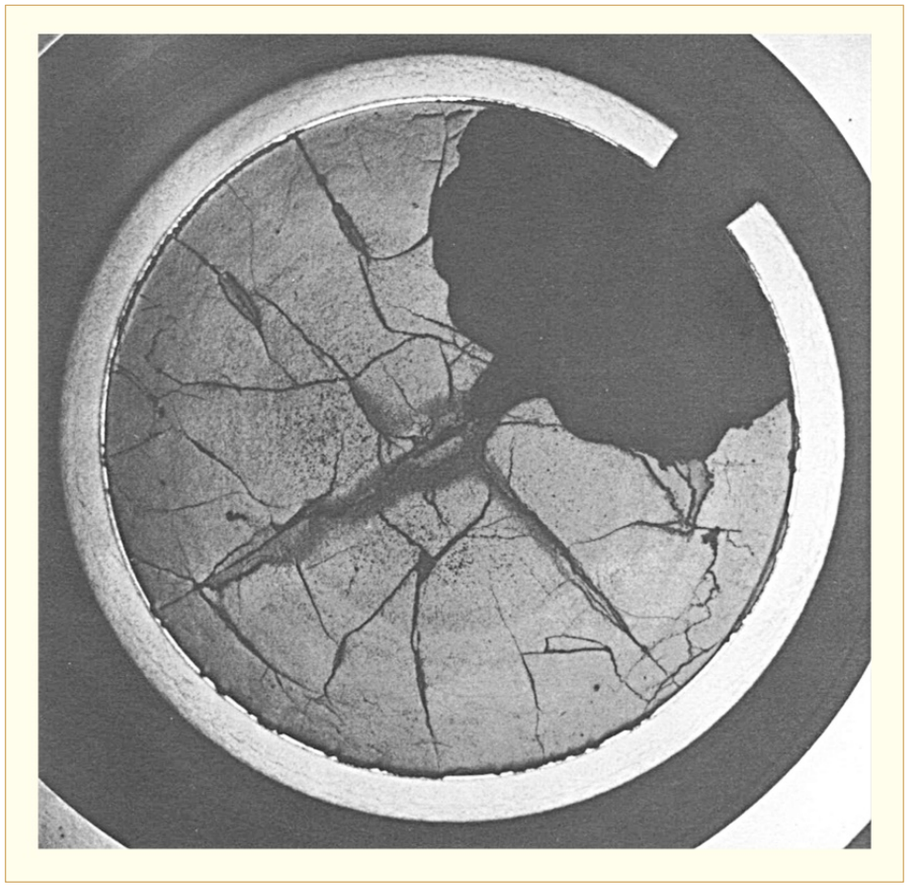


Figure 6-3: Macrography of a BWR rod, showing a secondary degradation of the Zr-lined, with a long axial cladding crack, [Yeager & Schneider, 2005].

6.3.3 Radionuclides used in reliability monitoring

The fission products commonly used to monitor fuel performance are identified in Table 6-1. This table lists fission gas and iodine isotopes in the order of their decay half-lives; i.e., shortest half-life to longest-half-life. It also includes their production rates and approximate steady-state inventories. In addition, the fission products ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{91}Sr and ^{92}Sr, and the transuranics listed below are frequently used to monitor fuel performance during operation; i.e.,

- Neptunium ^{239}Np (PWRs and BWRs with normal water chemistry (no hydrogen));
- Plutonium ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu; (specifically if mixed oxide fuel is used in some assemblies)
- Americium ^{241}Am;
- Curium ^{242}Cm,^{243}Cm, ^{244}Cm.

The gamma activities of fission gases generally provide the first sustained indication of a new failure because they can escape through small leakage paths and do not deposit on surfaces. The fission gas isotopes ^{138}Xe (14.1 min) through ^{133}Xe (5.25 d) decay slowly enough to be collected and measured in counting rooms via "grab samples". The sum of the activities of these isotopes, together with ^{135}Xe (9.10 h), ^{85m}Kr (4.48 h), ^{88}Kr (2.84 h) and ^{87}Kr (76 min) constitute the "Sum-of-Six" mentioned above. The activities of shorter-lived isotopes, ^{89}Kr (3.15 min) and ^{137}Xe (3.82 min), are sometimes measured in addition to those of the longer-lived nuclides by means of γ-spectrometers connected to flowing sample lines. They typically decay too rapidly for assessment by means of discrete samples since firstly the transport times from a leaking fuel rod to normal sample points are in the range of 3–6 minutes, and secondly the sample has to be transferred to the counting system, which also takes time.

Measurements of the activities of soluble fission products such as the isotopes of iodine listed in Table 6-1 are used alone and in conjunction with measurements of fission gas to identify and track the state of leaking fuel rods. The presence of iodine, cesium and other soluble nuclides (^{88}Rb, ^{89}Rb, ^{91}Sr, ^{92}Sr, ^{132}Te, …) during steady-state operation indicates mass transfer of coolant into and out of one or more leaking fuel rods. However, when the inner free volume of the leaking rod becomes empty of water), the release of these isotopes can be reduced dramatically compare to the gases release level, because they deposit on the inner surface of the clad; i.e., they are "trapped": see an example of Figure 6-4. Other soluble fission products or those released as metallic or oxide particles (^{91}Y, ^{92}Y, ^{99}Mo, ^{103}Ru, ^{106}Ru, ^{140}Ba, ^{140}La, ^{144}Ce…) are generally observed with larger leakage paths or following secondary degradation. The exception is the occurrence of spikes in the activities of soluble nuclides such as iodine, which can occur during shutdown, startup or other transients involving changes in pressure that promote the flow of coolant (in liquid phase or after intense vaporization) into and out of a leaking fuel rod.

When observed in measurements of coolant activity, the range of decay rates of the five iodine isotopes starting with ^{134}I (52.6 min) through ^{131}I (8.04 d) provide a means for assessing the state of leaking fuel that is similar to the gaseous radionuclides. That is, the long-lived isotope ^{131}I is observed in greater relative proportion to the shorter-lived isotopes in a new failure or in the case of a restrictive leakage path. The ratio of long-to-short lived isotopes decrease with depletion of the inventory of iodine in the rod intraspace and with increasing rates of release to the coolant. In some plants, a specification on the primary activity monitoring uses the ^{134}I isotope as a "signature" of fissile material release, and a quantitative assessment (relationship between the concentration of ^{134}I in coolant and the quantity of released fuel material) can be set up with some hypotheses on the type of fuel concerned and its burn-up.

Table 6-1: Tabulation of fission product isotopes useful in on-line monitoring with their respective decay constants, production rates and typical inventories, after [Rudling & Patterson, 2009].

Nuclide	Half-life	Decay constant 1/sec	Production rate			Production ratio			Inventory	Inventory ratio
			^{235}U	^{239}Pu	^{241}Pu	^{235}U	^{239}Pu	^{241}Pu		
Fission gas isotopes			MBq/(gm sec)			$^{133}Xe/x$			MBq/rod	$^{133}Xe/x$
^{89}Kr	3.15 m	3.66E-03	1.31E+02	3.99E+01	3.29E+01	6.10E-04	2.05E-03	2.40E-03	3.84E+07	2.33
^{137}Xe	3.82 m	3.02E-03	1.43E+02	1.37E+02	1.53E+02	5.59E-04	5.96E-04	5.16E-04	7.68E+07	1.17
^{138}Xe	14.1 m	8.19E-04	3.91E+01	3.05E+01	3.77E+01	2.04E-03	2.69E-03	2.09E-03	7.68E+07	1.17
^{87}Kr	76 m	1.52E-04	2.96E+00	1.08E+00	8.90E-01	2.69E-02	7.59E-02	8.88E-02	2.30E+07	3.89
^{88}Kr	2.84 h	6.78E-05	1.88E+00	6.94E-01	5.12E-01	4.25E-02	1.18E-01	1.54E-01	3.20E+07	2.8
^{85m}Kr	4.48 h	4.30E-05	4.34E-01	1.88E-01	1.31E-01	1.84E-01	4.36E-01	6.05E-01	1.28E+07	7
^{135}Xe	9.10 h	2.12E-05	1.07E+00	1.22E+00	1.17E+00	7.42E-02	6.73E-02	6.77E-02	2.62E+07	3.41
^{133}Xe	5.25 d	1.53E-06	7.97E-02	8.19E-02	7.90E-02	1	1	1	8.96E+07	1
Iodine isotopes						$^{131}I/x$				$^{131}I/x$
^{134}I	52.6 m	2.20E-04	1.30E+01	1.22E+01	1.37E+01	1.70E-03	2.36E-03	1.80E-03	9.60E+07	0.43
^{132}I	2.28 h	8.45E-05	2.80E+00	3.43E+00	3.06E+00	7.87E-03	8.39E-03	8.03E-03	5.95E+07	0.69
^{135}I	6.61 h	2.91E-05	1.41E+00	1.42E+00	1.55E+00	1.56E+00	2.03E-02	1.58E-02	8.32E+07	0.49
^{133}I	20.9 h	9.21E-06	4.80E-01	4.91E-01	4.77E-01	4.59E-02	5.87E-02	5.15E-02	8.96E+07	0.46
^{131}I	8.04 d	9.98E-07	2.21E-02	2.88E-02	2.46E-02	1	1	1	4.10E+07	1

Notes:
10 × 10 rod
2% fissile material (^{235}U or ^{239}Pu)
24 W/(g HM)

ANT International, 2015

Note that measurements of fission product activity via coolant samples can be confounded both during and soon after the end of operation with significant fuel leakage due to the effects of fission products in the cleanup systems or on the surfaces of fuel rods. Such confounding occurs also in the sampled fluid itself, when it is counted after several days of decay. An example is the decay of tellurium to iodine, xenon and then cesium. Issues can arise when measurements are made after power or flow transients or soon after shutdown, as in sipping during fuel shuffling. On-line measurements tend not to be affected by such precursor effects when performed during steady-state operation – a common practice.

In addition to fission products, fuel can be transported from a leaking rod when fuel pellets are exposed to flowing coolant and its radiolytic decomposition products. In such conditions, $UO_2$and MOX fuel pellets oxidize first along their grain boundaries and then into the affected grains. The oxidation of UO_2 to U_4O_9 produces a slight volumetric contraction; i.e., $\Delta V/V \sim -2\%$. Continued oxidation leads to the formation of U_3O_8 with a large volumetric expansion, +36%, which causes stress on the clad material and can directly affect the leak size. Further exposure to coolant can produce small, hydrated crystals similar to the mineral schoepite, $(UO_2)_4O(OH)_6 \bullet 6(H_2O)$. The oxidation process and volumetric strains weaken the pellet matrix and form grain-size and smaller particles that can be transported from a leaking fuel rod by flowing coolant as shown in Figure 6-3. This degradation is even very efficient when submicronic grains are already present at the pellet surface, coming from the "high burn-up structure", HBS (it is a porous peripheral layer formed at intermediate and high burn-up and at low temperature).Fuel particles which are distributed throughout the primary system are called tramp uranium or, more generally, a tramp source as noted earlier. Tramp sources in the active core produce fission products as does the fuel in a leaking rod.

7 Poolside assessments

If a core is suspected or known to contain leaking fuel based on the methods discussed in the previous sections, the fuel is almost universally checked for leaks during the subsequent refueling (or sometimes forced) outage. In many cases, all fuel assemblies of the core are checked for leaks using failure detection systems that either do not affect or have only a minor impact on outage time. In a few cases, when failure diagnosis made during the cycle gives unambiguous information on the failed rod(s) (e.g. a specific set of assemblies based on a narrow range of burn-up or power suppression testing), it is possible that only a part of the fuel assemblies is checked.

As LWR fuel assemblies are now dismountable and "reparable"[8],identifying individually the leaking rod(s)enables continued irradiation of the affected assemblies after the leaker(s) have been replaced with suitable, non-leaking rods. This approach, based on technical and economic considerations in relation with plant operation specifications, is clearly valuable when the assembly has to be irradiated during at least two more cycles. When only one more cycle remains, other information is typically considered to make a decision on its reutilization; e.g., length of the next cycle, number of detected failed fuel assemblies[9], burn-up already reached, availability of equivalent fresh rods etc. Moreover, as an assembly repair campaign is expensive, time consuming and requires space in a fuel handling pool, repairs are often carried out not during the concerned refuelling outage, but later, when the plant is operating. Repair operations are sometimes performed when several leaking fuel assemblies are present on the plant site and are gathered in the same pool to take advantage of a "serial effect". However, the timing of such repairs is affected by constraints such as the need to resume plant operation with suitable fuel that is available on site and the ability to store the leaking fuel for extended intervals (one to a few years). Sometimes these situations are not easily manageable by the plant operator.

Independent of of the resource and scheduling issues, poolside assessments involve identifying the leaking fuel assembies and then identifying the leaking fuel rods in the affected assemblies if they are to be repaired for continued irradiation or if a detailed failure investigation is needed. The methods for identifying the leaking fuel are reviewed in te sections which follow.

7.1 Identification of failed fuel assemblies by "sipping test" methods

7.1.1 Physical principles applied in a sipping test

Various techniques are used to identify leaking fuel assemblies and are collectively referred to as "sipping" methods. Although the used equipment can vary considerably, the underlying method is basically the same: it consists of placing the checked assembly in a closed, or partially closed, environment to create physical local conditions and provoking a release of radionuclides from out of the failed rod to this environment. Radioactive isotopes of fission products are more are used as a "signature" of the leak presence. These conditions correspond the most often to a relative pressure discrepancy between the inner free volume of the rod and the external pressure, at the defect level. This relative discrepancy can be made:

- Either by increasing the rod internal gas pressure (and maintaining the external pressure constant by other means). This change is generally achieved by increasing its temperature by a few tens of °C. Temperature increase can be due to the decay heat of the fuel assembly and/or to a supplementary external heating. There are two contributions to this pressure increase (see Figure 7-1):

[8] Reparable in this case means that each fuel rod can be extracted from the assembly and replaced with another fuel or dummy rod (e.g., solid Zr-alloy rod) for continued operation.

[9]The term « failed/leaking fuel assembly » is often used: it means that one or several untight fuel rod(s) is/are present inside the assembly

- as the inner free volume is filled by non-condensable gaseous products (mainly ^4He coming from rod pressurization at manufacturing, xenon and krypton isotopes coming either directly from fission, or after radioactive decay of a father "isotope", iodine or bromine respectively, and H_2 coming from water radiolysis), the Boyle-Mariotte law PV=nRT is active and is used in practice with its differential form : $\Delta P/P = \Delta T/T$.
- as liquid water is often present in a failed rod (see previous chapters), temperature increase provokes vaporization of a part of this liquid and formation of a vapor pressure following the saturation law. When the local temperature is higher than about 65°C, this effect becomes more active than the first one.

- Or by decreasing the external pressure. This can be achieved:
  - by a vertical ascent of the fuel assembly in the reactor pool or in the storage pool of the plant. The external pressure corresponds to the hydrostatic pressure at the defect elevation (i.e. the depth in the pool), so it decreases,
  - by applying vacuum to an upper gas blanket when the checked assembly is enclosed in a "sipping cell" (see details below).

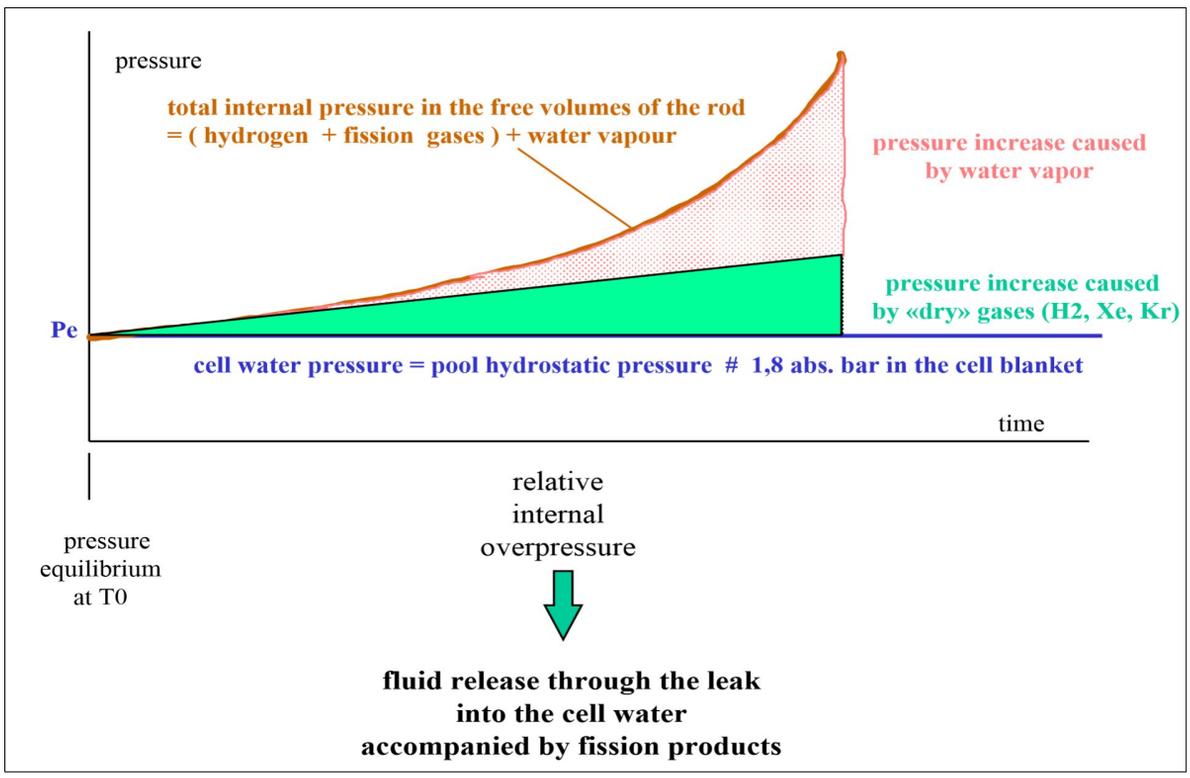


Figure 7-1: Effect of the heating of the free volumes of a failed fuel rod on the release of fission products (temperature increase is considered as proportional to the time).

For both physical phenomena, the achieved pressure difference with standard sipping methods is about 0.8 to 1 bar. The fluid transiting through the failure can be either gas or liquid water, depending on the composition of the inner free volume in the vicinity of the failure. Fission products are transported by the released fluid. If xenon and krypton isotopes are routed either by gas or by liquid in solubilized mono atomic form, a liquid phase is able to carry solubilized (or under the form of small particles) chemical compounds formed with other fission product elements: iodine, caesium, molybdenum, ruthenium, barium, lanthanum.

7.1.2 Sipping test assessment and analysis of the activity signal

The detection of a failure relies on changes in the concentration of fission products that are released from the fuel assembly or rod being tested; i.e., changes in gamma activity, beta activity, isotopic composition of fission products or combinations of such measurements. These changes can be monitored either by on-line measurement (e.g. by total beta or gamma activity, or by on-line gamma spectrometry), or by sampling with delayed analysis. In both cases, the fluid in contact with the fuel rods shall be routed to the measurement system or to a sampling (collection) point. The technique used to identify a leaking assembly can vary depending on the size of the leak, the background activity from tramp uranium and on the time of sipping relative to reactor shutdown. Changes in the gross (total) gamma activity of water or noble gas samples representative of the fuel being tested are sometimes used to identify a leaker, particularly in cases of low background activity. In general, however, changes in the gamma or beta activity of nuclides with moderate-to-long half-lives are typically used to minimize the effects of background activity and decay time. Isotopes such as ^{133}Xe (5.25 d), ^{131}I (8.04 d),^{85}Kr (10.72 y), ^{134}Cs (2.06 y) and ^{137}Cs (30.2 y) are the most detected. Detection of less volatile isotopes, such as ^{99}Mo (66.0 h), ^{140}Ba (12.75 d), ^{140}La (1.68 d) etc. indicate generally presence of a relatively large defect. Finally, detection of non-volatile isotopes, such as ^{103}Ru (39.3 d), ^{106}Ru (373 d), ^{144}Ce (284.9 d) or heavy isotopes (^{239}Np, 2.36 d) indicates a probable release of fissile material through a large defect. In addition, many of the current sipping methods also involve the collection and measurement of noble gases to enable the detection of leaks that are too small, or when the inner free volume of the rod is filled only with gas, to allow the release of soluble fission products in quantities clearly detectable in the sipping process.

7.1.3 Main categories of sipping test methods

Historically, sipping methods have been classified as wet, dry or vacuum based on the manner in which fission products are collected for measurement [Lin, 2013]. In practice, however, sipping techniques can better be classified as ”open” or ”closed” methods based on the manner in which the fuel being tested is isolated from other assemblies or from its environment during the sipping process. This section gives an overview of each method. Additional information is provided on the methods in the sections that follow., which are described individually more in details in the following paragraphs: Sipping methods comprise:

- The wet, in-reactor building methods represented by the “TELESCOPE sipping”, “IN-MAST sipping”, “ON-LINE sipping system OLSS” and various hood systems are open methods, which take benefit either from the pressure differential produced by the positive altitude variation when the assembly is unloaded from the core or from temperature increases They have become the primary means of leak detection because of the small amount of supplementary time needed to inspect a BWR, PWR or VVER core; e.g. about 16 hours for a large BWR with an in-core sipping hood, versus close to a week for vacuum sipping [Knecht et al., 2001]. They are simple and passive systems that quickly provide information about the presence of absence of fuel rod leaks during the refueling operations. Although the open methods have become the standard for sipping, a disadvantage is that their resolution can be insufficient to detect small leaks, particularly when large leaks have occurred and background activity is high.

- Wet sipping is also used in storage pools to test individual assemblies or fuel rods. It makes use of a cannister (called “sipping cell”), anchored permanently on the bottom of the spent fuel storage pool or installed temporarily for a specific sipping campaign, to isolate the fuel being tested. Release of fission products is provoked by heating of the cell water. It is a closed test method similar to the vacuum sipping method described below which provides good resolution but is slow relative to the open methods.

- The dry sipping method uses also a cannister but is not used today in power reactors because of issues related to handling and test time, decay heat and cladding temperature.

- The vacuum sipping method also makes use of a cannister, located in the storage pool, to isolate individual fuel assemblies. As for the wet in-cell sipping, vacuum systems include a gas circuit to facilitate the fission product measurement or collection. It is frequently used to

supplement the in-reactor methods, because of its higher resolution and detection capabilities. Moreover, a detectable signature is obtained in favourable cases after a few minutes, and it is not necessary, compare to wet in-cell sipping, to wait for the effect of the heating, which starts to be detectable after about 10 or 15 minutes. This method utilizes decay heat and does not require an external heat source, which is an advantage regarding the technology of the circuits, the management of the steam in the gas circuit and the risk of associated contamination.

Note also that the results of power suppression tests are sometimes used in conjunction with the wet, in-reactor methods to decrease the number of bundles that must be sipped and to decrease the chance of missing a leaking assembly. This is the case in some BWRs with an operation called flux tilting or power suppression testing as described earlier.

7.1.4 Wet in-core sipping systems

The configuration of wet, in-core sipping systems varies according to reactor type. In BWRs, where each fuel bundle is surrounded by a flow channel, sipping can be performed with a sampling hood at the top of a fuel assembly or a group of assemblies. An example of a multi-assembly sipping hood and measurement system is shown in Figure 7-2. In this system, the hood is open at the bottom and divided into four quadrants which match the core cells. The hood is placed on the upper core support grid over a 4 x 4 array of fuel assemblies by means of the refueling mast. Air is injected into the top of the hood to displace water and expose the upper ends of the fuel channels, which extend slightly above the support grid. The air bubbles isolate the assemblies covered by the hood and interrupt the convective flow of cooling water through the bundles. As a result, the temperature of the fuel increases and fission products are driven out of any leaking rods that might be present. The fuel channels prevent cross mixing of released fission products among the 16 assemblies. Suction probes, which also include thermocouples to monitor water temperature, extend down into the fuel channels. After fuel heatup, water is drawn from the fuel channels and passed through gas separation and scanning systems. Leaking fuel is identified by continuous measurement of the extracted gas to detect changes in the activity of ^{133}Xe or ^{85}Kr.

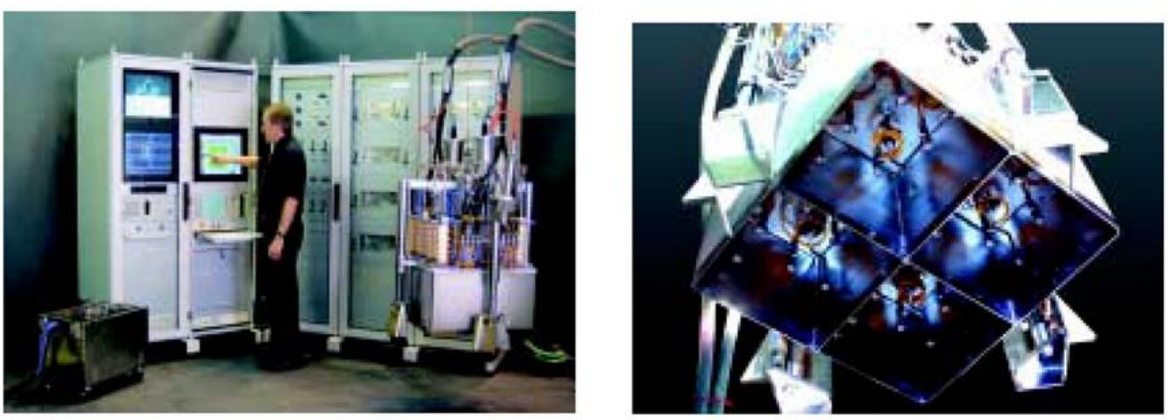


Figure 7-2: Wet in-core sipping system with control and data acquisition modules, detector and housing and 16 bundle sampling hood, [Knecht et al., 2001].

This concept has also been applied to individual fuel rods by replacing the in-core hood with a tubular sleeve. The single-rod technique is used in fuel handling or storage pools, where rods can be extracted from fuel assemblies. As shown in Figure 7-3, a fuel rod is removed from its bundle and drawn vertically into the sleeve. The sleeve isolates the rod from the pool and interrupts the convective flow of cooling water, so the fuel temperature increases due to decay heat. Fission products diffuse or flow out of a leaking rod at a rapid rate because of the increase in temperature inside the fuel rod and because of the decrease in ambient pressure caused by raising the rod into the sleeve. The concentration of released fission products in the water within the sleeve is enhanced by the small annular volume between the sleeve and fuel rod. Samples are drawn from the top of the sleeve and routed through separation and measurement systems in the same manner as the in-reactor, hood or

mast sipping systems. Water samples can also be analyzed for the presence of soluble fission products such as ^{131}I or ^{137}Cs.

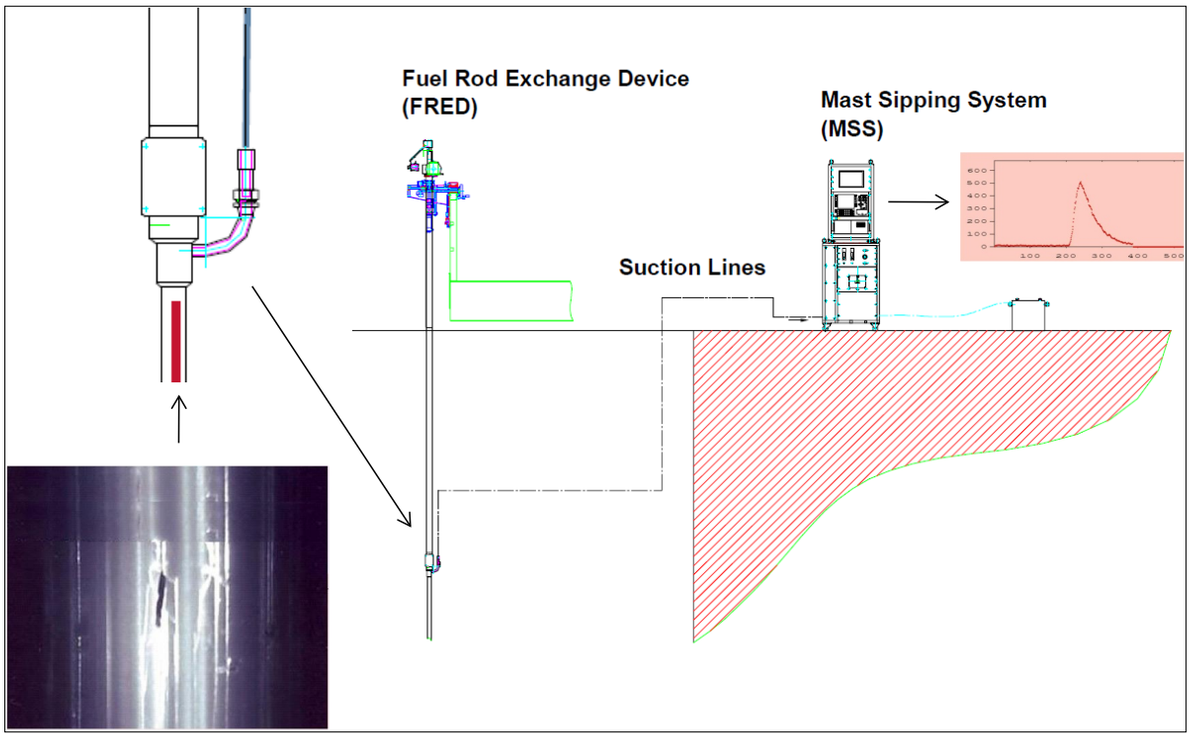


Figure 7-3: Wet, in-pool system for sipping individual fuel rods, after [Dust et al., 2008].

In-core wet systems for sipping PWR fuel differ from those used with BWR fuel because of the absence of a flow channel around PWR fuel bundles. In a PWR, sipping is performed by lifting a bundle into a tube that surrounds the refueling grapple. The tube isolates the fission products released from the bundle being tested from the balance of the core. The release of fission products from a leaking rod is enhanced by the change in elevation as the bundle is lifted into the tube (see Figure 7-4, "phase 1"). As shown in Figure 7-4 "phase 2"', air is also sparged upward through the tube and fuel assembly to enhance the transport of gaseous fission products from the fuel to the collection port located at a higher elevation in the tube. The collection port is connected to a suction line which routes the noble gases to an on-line gamma activity analyser, e.g. with a window centred on the 81 keV gamma ray of the ^{133}Xe (see Figure 7-4, "phase 3"). Figure 7-5 details the air circuit used for sweeping the fission gas dissolved in water or present in the form of small bubbles. Leaking fuel is identified by changes in the gross or ^{133}Xe gamma or beta activity as shown in Figure 7-6.

Testing is typically performed during refueling outages when fuel is moved among core positions. Detection is at best when the fuel assembly has reached the upper position in the mast of the handling machine and when this machine is stationary. The process is reported to add only a few minutes to each move so that the incremental time to test an entire core is less than a day; e.g., incremental time of 2 minutes per move and about 5 hours for a 900 MW core, [Beuneche et al., 1988].

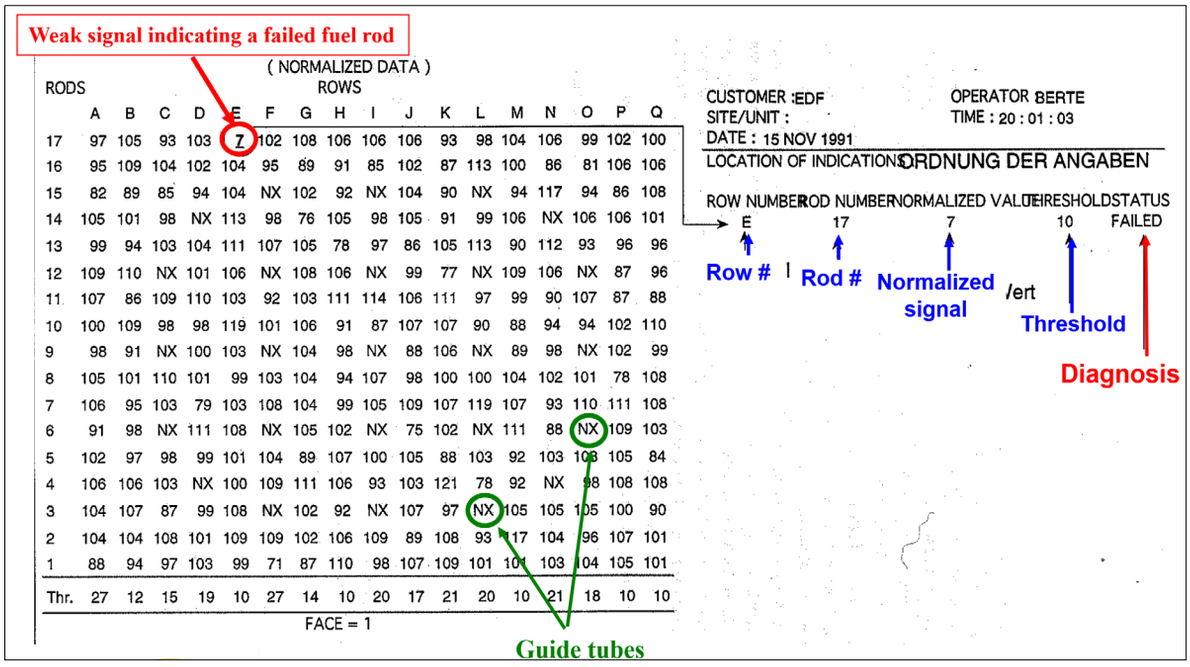


Figure 7-18: Example of ultrasonic checking results for a PWR 17*17 fuel assembly (Framatome campaign obtained with the ECHO 330 system).

Experimental feedback shows this technology is effective when the fuel-clad gap at pool temperature is open (as for low burn-up assemblies) and/or when a large quantity of liquid water is present in a failed rod. As the extremities of a fuel rod have been irradiated at lower linear power during operation, they present a lower temperature during outage so that water condenses preferably at these locations. For the same reason, radiolysis of liquid water by gamma rays emitted by the fuel material occurs at a lower rate, so water disappears more slowly at the ends of a leaking rod. In particular, the bottom part of a rod plays the role of a "water reservoir" and is generally the preferred zone for ultrasonic inspection.

Note, however, that the risk of "overcall" (i.e. to attribute a failure when it doesn't exist) increases when the fuel-clad gap is closed or when crud is present at the clad outer surface. Note also that there is a risk of "under call" (i.e. to miss a real failure) when there is no more water inside the rod due to conditions such as low ingress, previous vaporization or disappearance by radiolysis and chemical reactions with the inner cladding surface.

7.3 Identification of failed fuel rods in a fuel assembly by visual inspection

Even if it can be considered as obvious, it has to be mentioned here that an underwater visual inspection of the four external faces of a fuel assembly can localize failed rods in favourable cases with a direct view of the failure/rod aspect or of gas bubbles escaping from a leaking fuel rod. Visual inspection of the assembly faces can be valuable when there is a known risk of damage on the peripheral rods: hydraulic turbulences or vibrations, friction between two neighbouring assemblies etc. Chapter 5 gives valuable examples on the interest of this checking (see for example Figure 5-54 and Figure 5-55).

Moreover, visual inspection can also be oriented to inspect the water channels between two rows of rods. Such inspections require adequate lighting, typically combined with lighting through the assembly from the side away from the camera or periscope lens. Peripheral examinations are can detect rod deformation, channel blockage by debris etc.

The rod upper end plugs can also be visually checked for the presence of unusually large or small fuel rod growth.

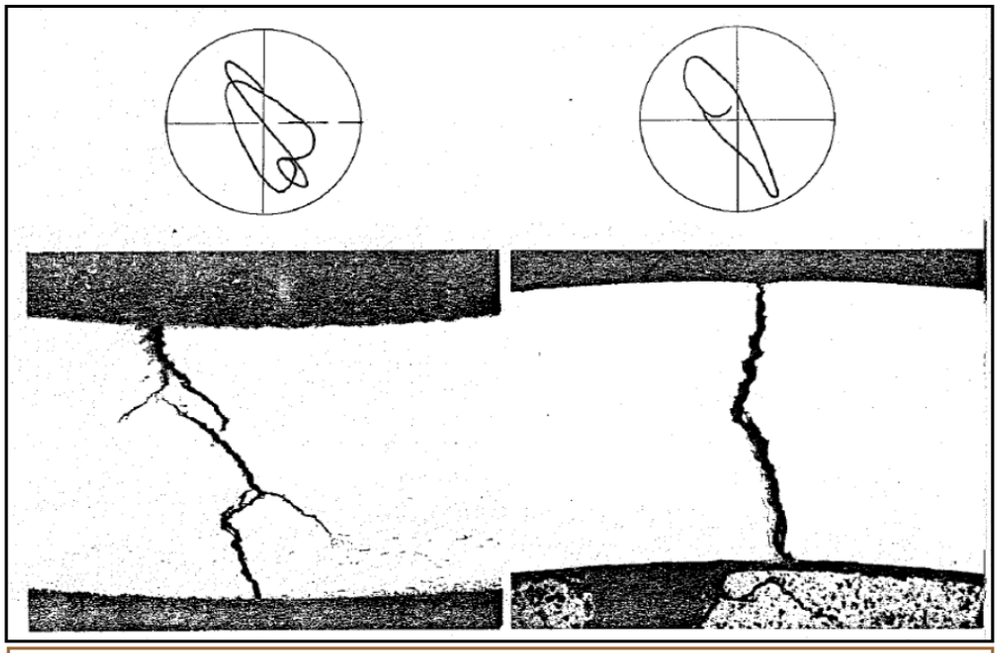
7.4 Identification of a failed fuel rod after removal from the fuel assembly

When the condition of a leaking fuel rod allows it to be removed from its assembly, two examination methods other than the sipping and ultrasonic techniques already mentioned are commonly used to locate and characterize the failure; i.e., eddy current (EC) and visual inspections. Both methods usually allow determining the failure location and identifying the presence of multiple leakage paths; e.g., primary and a secondary defects. The results of visual examinations are typically the starting point for identifying the cause the failure.

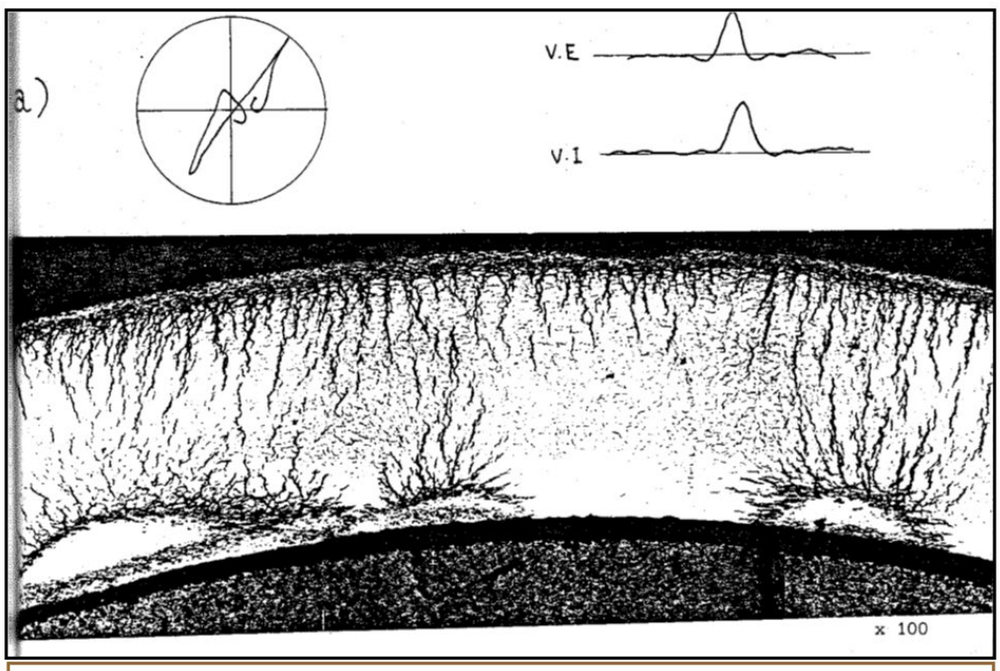
7.4.1 Identification of a failed fuel rod by Eddy Current (EC) technique

Eddy current inspections are intended to detect partial or through-wall defects in fuel cladding and identify their location. The most common form in poolside inspections utilizes an encircling coil, which is moved along the full length of the fuel rod. In its basic form the coil is made of a copper wire, which is excited with an alternating electrical current. This wire produces a magnetic field around itself, which oscillates at the same frequency as the current running through the coil. When the coil approaches a conductive material, currents opposed to those in the coil are induced in the material: they are called “Eddy Currents”. A defect in the conductive material disturbs the path of Eddy Currents, creating a local magnetic field that changes the balance of the system. This can be detected by measuring the changes in the impedance in the coil, which is in turn a sign of the presence of defects inside the material being examined.

The amplitude and phase of the coil current are monitored simultaneously when the coil is moved along the rod or when the rod is moved while the coil is held at a fixed position. The amplitude and phase-angle of the resulting signal are compared to those obtained by calibrating the EC system to reference standards containing defects of known size and shape. Figure 7-19 gives examples of signals obtained with real defects observed on a PWR irradiated zircalloy-based cladding tube.



Signals due to through-wall cracks
CEA/DEN



Signal due to pure hydriding
CEA/DEN

Figure 7-19: Examples of electromagnetic signals observed with defects on a PWR irradiated cladding tube.

Experience with the encirciling-coil EC technique with irradiated LWR fuel rods shows that it is a relatively reliable method for identifying defects, even if there is no evidence from outside (e.g. a defect under a coating). In particular, it doesn't need to detect fluids or products present in a fuel rod (e.g. liquid water as for UT technique), or released by it (e.g. fission products, for sipping methods). So it is well adapted to detect many types of defects, even small through-wall penetrations.

However, examinations with encircling-coil EC systems require the extraction of the rods to be examined from their respective fuel assembly skeleton. Such handling can be a risky operation if the rod is fragile due to the primary failure or damaged by secondary hydriding. Moreover, signal assessment is often complex, particularly for high burn-up fuel rods with a strong pellet-cladding contact and/or with incipient cracks starting from the inner surface of the clad. Hydriding of the base metal is also a source of signal deformation. Finally, the rod to be checked must be already chosen as a "leaker" or "doubtful" by another method.

7.4.2 Identification and characterization of a failed fuel rod by visual inspection

In many cases, a rod is already known as a "leaker" based on the results of techniques such sipping or ultrasonic inspection. In other cases, visual examinations must be used to both identify the failed rod and characterize the conditions of failure. Visual inspections are intended to identify the location and the external aspect of the failure and, thereby, to contribute to the determination of the most likely cause of failure. They a powerful mean to understand the level and the evolution of the fission product concentrations in the primary circuit during the previous cycle.

Visual examinations in a storage or handling pool needs a vertical examination bench which allows vertical motion and rotation of the fuel assembly or rod. For fuel assemblies, a fuel preparation machine (elevator) with a support system that enables the rotation of the assembly about its longitudinal axis is typically. For individual fuel rods, a system that moves the rod axially and azimuthally relative to the camera is used. A high magnification underwater camera monitors the surface of the cladding. As huge progresses have been made thanks to high-sensitivity, radiation-resistant CCD cameras, a lot of information on the overall status of the rod can be gained thanks to this examination (external corrosion, cruds, wear, fretting…).

8 Spent fuel storage

The primary objective of this report has been to provide guidance for improving fuel reliability. The first step is to determine whether the core is defect-free or not using radiochemistry online assessment during reactor operation (Section6.3). When defected rods are detected or suspected, failed rods can be identified primarily during poolside examinations (Chapter 7) possibly supplemented by examinations in hot cells, if necessary. An obvious reason for doing so is to assess failure root cause(s) to ensure that any identified failure mode does not occur again. A second reason is to satisfy fuel-integrity-related requirements that are applicable to operations to be conducted during the back-end of the fuel cycle.

8.1 Introduction

Spent fuel is generated continually by operating nuclear reactors. It is stored in the reactor fuel storage pool for a period of time for cooling and then may be transferred to a designated wet or dry spent fuel storage facility, where it will await reprocessing or final disposal.

The discussion in this section will be mostly limited to dry storage and transportation of spent LWR fuel.Storage at the reactor sites were initially intended to serve for very limited periods of time (as short as 90 days) prior to shipment of the spent fuel from the reactor sites to a reprocessing or disposal facility. However, because of delays in developing disposal facilities and limited market appeal for reprocessing in most countries, storage periods have been progressively extended, in many cases, beyond the design lifetime of the reactor facility.

Operation of spent fuel storage facilities in accordance with the requirements of the safety case, the license conditions and the applicable regulations is the responsibility of the operating organization [IAEA, 2012, page 11].

8.2 Spent fuel Storage

Wet and dry storage of spent LWR fuel have been safely implemented for over 50 and 35 years, respectively. IAEA Safety Standards Series No. SSG-15, Storage of Spent Nuclear Fuel, provides generic guidance for ensuring the safety of spent fuel storage. As stated in para. 1.3 of SSG-15, “The safety of a spent fuel storage facility, and the spent fuel stored within it, is ensured by: appropriate containment of the radionuclides involved, criticality safety, heat removal, radiation shielding and retrievability.”

8.2.1 Containment

Containment prevents the release of radioactive material into the environment and is provided by the spent fuel cladding and the storage system (e.g., welded or bolted cask or welded canister). Therefore, maintaining spent fuel cladding integrity during dry storage is an important component of regulations.

8.2.2 Criticality Safety

Criticality safety precludes an unplanned criticality event. Ensuring subcriticality often relies on geometry control. However, maintaining the geometry of spent fuel especially during hypothetical accident conditions may be challenging. Criticality control functions are often enhanced by other structures, systems and components (SSCs). These may include controlling fissile content, inclusion of neutron absorbers and preclusion of moderators.

8.2.3 Decay heat removal

Effective removal of decay heat is important because degradation phenomena that could affect spent fuel integrity and some SSCs important to safety, such as polymers, are thermally activated.

8.2.4 Radiation Shielding

Shielding ensures that radiation exposure remains within safe limits and must be provided by the storage and transportation system.

8.2.5 Retrievability

Retrievability is important to the extent that it may minimize the complexity, and thereby maximize the radiological safety of future spent fuel management operations, such as transportation, or spent fuel management options, such as repackaging for final disposal.

8.3 Discharged Fuel Assemblies

Clear documentation of the state of all spent fuel assemblies or bundles is required for subsequent operations associated with the back-end of the fuel cycle. Table 8-1 shows examples of operational limits and conditions for dry storage, which require pre- and post-irradiation information related to the assemblies or bundles that are considered for dry storage in approved systems.

8.3.1 Constraints on Loading Leakers – Damaged Fuel

The concept of damaged fuel is first rooted in reactor operations. In-reactor damage is based on the ability of the fuel to perform as desired. This same claim can also be made for defining fuel as damaged for the back-end operations of the fuel cycle, but instead of irradiation performance, pool or system contamination, handling capabilities, pressurization of containers, criticality and other issues are the main driving concerns. Leaking fuel rods and fuel assembly hardware defects need to be both considered. The need to obtain good photographic records and inspection data on the damaged fuel cannot be overemphasized.

- Detection of specific leaking rods within an assembly is covered in Sections 7.2 through 7.4 of this report. The variety of methods available includes visual, eddy current, ultrasonic and gamma scanning among others. Each has its pros and cons.

- The primary methods for determining assembly hardware defects are operation records and visual examination.

Currently, there is no internationally accepted definition of ‘damaged fuel’ for back-end operations. Historically, cladding breaches were the primary cause for classifying a fuel as damaged. However, in the United Kingdom, leaking fuel rods are not considered as damaged fuel unless there is gross damage to the rods. In Ukraine, the distinction is based on whether the rod can leak gas and whether water can contact the fuel. In Germany, rods with any sort of cladding breach could not, until recently, be put in dry storage. Damaged fuel is handled differently in various countries depending on regulatory requirements, available technologies and the stage of the fuel cycle, i.e., wet storage, dry storage, transport, or disposition in a repository or reprocessing. A summary of how damaged fuel is identified and managed in the USA and Germany is presented in the next sections.

Table 8-1: Examples of Operational Limits and Conditions for Dry Storage of Spent Fuel. Adapted from Table I (page 15) of [IAEA, 2012].

Subjects	Operational limits and conditions
Confinement	Constraints on loading leakers or assembly with some level of damage
Subcriticality	Maximum allowable fresh fuel enrichment or Pu content Minimum allowable concentration of neutron poisons in fixed absorbers, if applicable Restricted movement and restrictions on storage configurations of spent fuel Restricted use of moderator Specified minimum spent fuel burnup, if applicable Spent fuel assembly characteristics
Decay Heat Removal	Specified availability of cooling systems with specified maximum and minimum system temperatures Minimum cooling period after discharge of the spent fuel from the reactor and maximum burnup of the spent fuel Maximum temperature of concrete and of the cask surface Minimum tightness of spent fuel cask
Radiation Shielding	Maximum allowable burnup of spent fuel Minimum allowable water level in storage pool Requirements for radiation monitors, alarms and interlocks Minimum cooling period after discharge of the spent fuel from the reactor Maximum radionuclide concentrations in pool water Maximum radiation dose rates on cask surfaces and a specified distance (e.g. 1-2m) from the cask Minimum tightness of spent fuel cask
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'Damaged' fuel is handled differently in various countries depending on regulatory requirements, available technologies and the stage of the fuel cycle, i.e., wet storage, dry storage, transport, or disposition in a repository or reprocessing. The United Kingdom would not consider leaking fuel rods as damaged fuel unless there was gross damage to the rods. In Ukraine, the distinction is based on whether the rod can leak gas and whether water can contact the fuel. In Germany, rods with any sort of cladding breach would not, until recently, be put in dry storage.

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8.3.1.1 USA (Main Sources: [IAEA, 2018] and [NRC, 2007])

In 1983, in the absence of design information regarding the repository concept, damaged fuel was defined in US Federal regulations [US Department of Energy, 1983] as:

- Visually detectable fuel that cannot be handled normally;
- Radioactive leakage;
- Assemblies that had to be encapsulated for handling purposes (containment).

In 1984, the United States Nuclear Regulatory Commission (US NRC) issued a Director's Decision defining rods with breaches greater than pinholes or hairline cracks as damaged [Nuclear Regulatory Commission, 1984]. Although pinholes and hairline cracks were never explicitly defined, this did

specify an identifiable characteristic of the fuel that could be used as a threshold beyond which the "damaged" classification would apply.

In October 2002, the US NRC issued Interim Staff Guidance-1 (ISG-1)[10] Rev. 1 [Nuclear Regulatory Commission, 2002] providing a definition of damaged spent fuel based on the functions in storage regulations [Nuclear Regulatory Commission, 10 CFR 72] and transport regulations [Nuclear Regulatory Commission, 10 CFR 71] by specifying certain characteristics of spent fuel that determined whether or not the fuel or assembly was damaged.

In 2005, the American National Standards Institute (ANSI) issued a standard for defining damaged fuel for storage and transport [ANSI, 2005].

Finally, the US NRC issued Revision 2 of ISG-1 "Classifying the Condition of Spent Nuclear Fuel for Interim Storage and Transportation Based on Function" in May 2007. This revision provides guidance to the NRC review staff by classifying spent nuclear fuel based on their ability to perform fuel-specific and system-related functions that depends on (1) whether the fuel is being stored or transported, and (2) the design of the storage or transportation system.

8.3.1.1.1 Definitions

Definitions introduced in ISG-1, Rev. 2 are:

- Damaged SNF - Any fuel rod or fuel assembly that cannot fulfill its fuel-specific or system-related functions.
- Undamaged SNF - SNF that can meet all fuel-specific and system-related functions. Undamaged fuel may be breached. Fuel assembly classified as undamaged SNF may have "assembly defects."
- Breached spent fuel rod - Spent fuel rod with cladding defects that permit the release of gas from the interior of the fuel rod. A breached spent fuel rod may also have cladding defects sufficient to permit the release of fuel particulates. A breach may be limited to a pinhole leak or hairline crack, or may be a gross breach.
    - Pinhole leaks or hairline cracks - Minor cladding defects that will not permit significant release of particulate matter from the spent fuel rod, and therefore present a minimal as low-as-is-reasonably-achievable concern, during fuel handling and retrieval operations.
    - Grossly breached spent fuel rod (a subset of breached rods) - A breach in spent fuel cladding that is larger than a pinhole leak or a hairline crack. An acceptable examination for a gross breach is a visual examination that has the capability to determine whether the fuel pellet surface may be seen through the breached portion of the cladding. Alternatively, review of reactor operating records may provide evidence of the presence of heavy metal isotopes indicating that a fuel rod is grossly breached.
- Intact SNF - Any fuel that can fulfill all fuel-specific and system-related functions, and that is not breached. Note that all intact SNF is undamaged, but not all undamaged fuel is intact, since under most situations, breached spent fuel rods that are not grossly breached will be considered undamaged.
- Can for Damaged Fuel - A metal enclosure that is sized to confine one damaged spent fuel assembly. A fuel can for damaged spent fuel with damaged spent-fuel assembly contents must satisfy fuel-specific and system-related functions for undamaged SNF required by the applicable regulations.
- Assembly Defect - Any change in the physical as-built condition of the assembly with the exception of normal in-reactor changes such as elongation from irradiation growth or assembly bow. Examples of assembly defects are: (a) missing rods; (b) broken or missing grids

[10] An Interim Staff Guidance (ISG) is a NRC document issued to clarify or to address issues not discussed in a Standard Review Plan (SRP)

9 Summary

Fuel reliability has improved in all types of water-cooled reactors so that fewer fuel rods fail by developing leaks during operation, failures are being caused by fewer damage mechanisms and failures are taking place in fewer reactor operating cycles. Nevertheless, incentives exist to detect the occurrence of fuel failure and to mitigate both the near and long-term effects of fuel failure. On-line measurements of the radioactivity of nuclides and fissile materials released from leaking fuel rods to the reactor coolant and offgas system are used for both purposes. These data are supplemented by poolside examinations and, when necessary, by hot cell examinations.

The detection of a new failure is typically indicated by spike increases in a long-lived gaseous fission product such as ^{133}Xe (5.25 d) in plants that monitor gas activities or in spike increases of a long-lived soluble fission product such as ^{131}I (8.04 d) in plants that monitor only coolant activity. Success in detecting fuel failure is also reported with the use of an on-line spectrometer set to detect the presence of helium from the rod intraspace in the offgas stream. Changes in the ratio of long and short-lived nuclides are also useful in the detection of failure and in assessments of the progression of failure, particularly of small leaks; e.g., the ratio of ^{133}Xe/^{138}Xe or ^{131}I/^{134}I activities. The magnitude of ratios such as ^{133}Xe/^{138}Xe relative to the activity of the long-lived term (^{138}Xe) is used in some analyses to define the presence or absence of leaking fuel.

Fission gases are typically released first after fuel failure, with the release of soluble fission products following after enlargement of the leakage path. However, the presence of a failed rod with a small leakage path is sometimes indicated by spike increases in the activity of ^{133}Xe or ^{131}I during or after a reactor reactor transient involving changes in power or coolant pressure.

The onset of failure, the condition of the leaking rods and, to a limited extent, the number of leaking rods can also be inferred from changes in the regression coefficients of phenomenological models. In one type of model, fission product release is evaluated in terms of the relative activity release rates attributed to the assumed, underlying mechanisms; i.e., recoil, equilibrium or diffusion. In a second type, the release of fission products is characterized by means of empirical coefficients attributed to release from the fuel material to the rod intraspace, to escape from rod intraspace to the reactor coolant and to tramp uranium. The resulting values and applications tend to be specific to the respective models.

The consequences of cladding degradation after the initial failure (secondary degradation) are usually more severe than the failure itself because of the dispersal of fuel material into the primary coolant system due to the corrosion and erosion of fuel pellets exposed directly to the coolant. Radioactivity increases in the primary coolant system due to fuel failure and, to a greater extent, due to enlargement of the leakage path as a result of secondary degradation, which allows dispersal and deposition of fuel material in the core and related systems. The dispersed material contributes to system activity while the leaking fuel is operating and after the fuel has been discharged due to the residual material (tramp uranium) that remains in the core or that returns to the core from components of the primary system; e.g., steam generators in PWRs.

When leaking fuel is detected in BWRs, power suppression (or increase) testing is frequently performed to identify the location of the failed fuel. This testing involves the sequential movement of control blades to vary power in the respective control cells while measuring offgas or offgas and coolant activities. Increases in activity following a blade move indicates the leaking fuel is in a region where power is affected by the control blade. In many instances, the specific leaking fuel assembly can be determined by activity gradients around a control cell. When the leaking cell or fuel assembly is located, one or more control blades can be inserted to protect the leaking fuel from secondary degradation due to power changes during subsequent operation. Note that the need for such action has been reduced with the introduction of corrosion-resistant (lightly alloyed) Zr-liners in BWR fuel cladding. Note also that the risk of secondary degradation is lower in a PWR due to differences in coolant chemistry and power control, so the proactive management of leaking fuel practiced in BWRs is not required in PWRs or VVERs.

A common issue in the analyses of coolant or offgas activity is the effect of tramp uranium. Activity released by distributed fissile material can be a problem with respect to radiation exposure but is also a problem because it obscures activity released directly from failed rods. Methods have been developed

to identify the relative contributions of tramp sources and leaking fuel rods and are summarized in Table 6-3.

Additional information such as the burnup of a leaking fuel rod can be approximated by the ratio of short and long-lived isotopes of cesium; e.g., ^{134}Cs(2.07 Y)/^{137}Cs(30.17 y). An example is shown in Figure 6-8. However, measurement uncertainty combined with decreasing changes in this ratio with increasing burnup frequently restrict the use of such ratios to the identification of fuel in its first or second cycle (i.e., at low-to-moderate burnup) versus its third or later cycle (high burnup).

Fuel reliability specialists tend to have preferred radiochemical methods for detecting and assessing fuel performance. However, no single method gives unambiguous results on a consistent basis. Uncertainty associated with the interpretation of measurement data is generally reduced by applying a combination of empirical and phenomenological methods.

If one or more leaks are indicated by on-line measurements, then sipping is generally used at end of cycle to identify the failed fuel assemblies and to verify that only non-leaking fuel is loaded back into the core. The sipping process involves the sampling and analysis of coolant activity of samples that can be associated with specific fuel assemblies. The methods used for sipping vary among utilities and reactors, but include sampling hoods placed over groups of bundles in a reactor core, sampling probes on fuel-handling masts and sipping chambers both in a reactor core and in fuel handling pools. Similar techniques can also be used to assess the hermeticity of spent fuel prior to placement in dry storage containers.

The identification of leaking fuel rods in an assembly that has been identified as failed by in-core monitoring and post-operation sipping requires a number of poolside inspection techniques. The attenuation of ultrasonic waves in fuel rod cladding by water in the rod intraspace is a common and effective method for identifying leaking fuel rods. Such ultrasonic tests can be performed while the fuel rods are in their respective assemblies. Sipping of individual fuel rods or groups of rods is also used to identify failed fuel rods. The location of leakage paths through fuel cladding and other damage (secondary hydriding) is identified by eddy current inspections and visual examinations. Visual examinations with image capture by radiation-resistant, high-resolution closed circuit video cameras is typically required to characterize cladding perforations and either establish the most likely cause of failure or contribute to decisions regarding hot cell examinations.

References

Adamson R. et al., *Corrosion of Zirconium Alloys*, *ZIRAT 7* Advanced Nuclear Technology International, Uppsala, Sweden, 2002.

Adamson R. et al., *High Burnup Fuel Issues, ZIRAT-8 Special Topics Report*, Advanced Nuclear Technology International, Surahammar, Sweden, 2003.

Adamson R. and Cox B., *Impact of Irradiation on Material Performance,* Advanced Nuclear Technology International, Surahammar, Sweden, 2005.

Adamson R. et al., *Pellet-Cladding Interaction (PCI and PCMI), Vol ZIRAT-11 Special Topic Report*, Advanced Nuclear Technology International, Skultuna, Sweden, 2006.

Adamson R. et al., *Corrosion Mechanism in Zirconium Alloys, ZIRAT12*, Advanced Nuclear Technology International, Skultuna, Sweden, 2007.

Adamson R. et al., *ZIRAT21 Annual Report*, ANT International, Mölnlycke, Sweden, 2016.

Adamson R. et al., *ZIRAT23 Annual Report*, ANT International, Tollered, Sweden, 2018.

American National Standards Institute, ANSI Standard N14.33-2005, Storage and Transport of Damaged Spent Nuclear Fuel, 2005.

Armijo J.S., *Performance of Failed BWR Fuel*, Proceeding of the 1994 International Topical Meeting on Light Water Reactor Fuel Performance, West Palm Beach, Florida, USA, American Nuclear Society, pp 410-422, 1994.

Armijo J.S. and Abdullahi Z., *Pellet Cladding Interaction Fuel Failures during Anticipated Operational Occurrences in Pressurized Water Reactors*, US NRC Advisory Committee on Reactor Safeguards, Rockville, Maryland, USA, 2014.

Baily W.E., Harding L.P., Potts G.A., Proebstle R.A., *Recent GE BWR Fuel Experience,* International Topical Meeting on LWR Fuel Performance, AVIGNON, France, April 21-24, 1991.

Bailly H., Menessis D. and Prunier C., *The Nuclear Fuel of Pressurized Water Reactors and Fast Neutron Reactors*. CEA, France, Lavoisier, 1999.

Barner J., Cunningham M., Freshley M., Lanning D., *Relationship between Microstructure and Fission Gas Release in High Burnup UO2 Fuel with Emphasis on the Rim Region*, ENS International Topical Meeting on LWR Fuel Performance, "Fuel for the 90's", Avignon, France, European Nuclear Society, 1991.

Baurens B. et al., 3D *Thermo-Chemical–Mechanical Simulation of Power Ramps with ALCYONE Fuel Code*. Journal of Nuclear Materials, 452, pp 578-594, 2014.

Bechtel S. et al., *The German Quiver Project*, VGB Power Tec, 2019.

Beuneche D. et al., *Overview of Fuel Sipping in French Power Plants, International Technical Meeting on LWR Fuel Performance*, Williamsberg, Virginia, USA, American Nuclear Society, 1988.

Booth A., *A Method of Calculating Fission Gas Diffusion from UO2 Fuel and Its Application to the X-2-f Loop Test*, AECL, Chalk River, Canada, 1957.

Bouineau V. et al., *A New Model to Predict the Oxidation Kinetics of Zirconium Alloys in a Pressurized Water Reactor*, Zirconium in the Nuclear Industry: 15th International Symposium,Kammenzind, B., Limback, M. (Eds.), Sunriver, Oregon, USA., ASTM International, vol ASTM STP-1505, 2009.

Buechel R. A. R., Bradfute J. And Norell J., *Fuel Reliability: How it Affects the Industry, and One Fuel Vendor's Journey to Flawless Fuel Performance*: Nuclear Power International. Tulsa, Oklahoma, USA, Penn Wall Corp., vol 7, pp 22-24, 2014.