THE U.S. NUCLEAR REGULATORY COMMISSION'S RESEARCH ON FUEL BEHAVIOUR UNDER ACCIDENT CONDITIONS

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ABSTRACT

The U.S Nuclear Regulatory Commission (NRC) research programs on nuclear fuel are focused on the behaviour of high-burnup fuel during postulated loss-of-coolant accidents (LOCAs) and reactivity-initiated accidents (RIAs). Some of the findings are presented here.

1. Introduction

In addition to participating in international programs, the NRC sponsors experimental work at Argonne National Laboratory, analytical work at Pacific Northwest National Laboratory and Brookhaven National Laboratory, and provides a grant to the Pennsylvania State University. In cooperation with France's Institute for Radiological Protection and Nuclear Safety, the NRC supports research at the Russian Research Centre – Kurchatov Institute. The NRC staff actively directs much of this work and performs technical evaluations and code calculations. In recent years, most of NRC's fuel research has been on high-burnup effects that would have an impact on loss-of-coolant accidents (LOCAs) and reactivity-initiated accidents (RIAs).

2. Loss-of-Coolant Accidents

Experimental results on fuel behaviour under LOCA conditions have shown strong alloy and burnup effects, or more precisely strong materials and corrosion effects [1,2]. Zirconium containing niobium oxidizes more slowly than zirconium containing tin under operating and transient conditions. Niobium does not go into complete solution in zirconium metal as tin does, affecting the alloy's microstructure. Niobium appears in the surface oxide as an aliovalent impurity making the oxide more susceptible to breakaway. Surface roughness of the cladding also makes the oxide more susceptible to breakaway. Corrosion, on the other hand, is accompanied by significant hydrogen absorption, and this always enhances oxygen embrittlement of the metal. Several mechanisms, which are related to these effects, are now known to affect cladding embrittlement under LOCA conditions.

2.1 Beta-layer Embrittlement by Oxygen

As temperature increases during a LOCA transient, the amount of oxygen that a zirconium alloy's beta phase can hold also increases. Above about 1473 K (1200˚C), oxygen solubility becomes high enough in these cladding alloys that, after cooling, the prior-beta region will be brittle. This mechanism was understood in 1973. Based on data available at that time, the 1477 K (1204˚C, 2200˚F) temperature limit in NRC's regulation was thought to preclude such embrittlement, but the data only covered unirradiated Zircaloy [3].

2.2 Beta-layer Thinning

With time, diffusion of oxygen into the metal will convert more and more of the beta phase to an oxygen-rich alpha phase – the alpha layer grows and the beta region shrinks. For long times at high temperature, the beta region becomes so thin that the macroscopic specimen exhibits brittle behaviour when it is cooled. This mechanism was also understood in 1973 and was thought to be accommodated by the 17% oxidation limit in NRC's regulation, but again the data only covered unirradiated Zircaloy [3].

2.3 Localized Hydrogen-induced Embrittlement in the Balloon

Steam that enters through a rupture in a balloon causes oxidation inside the cladding. Hydrogen that is freed during this reaction is not swept away as it is on the outside of the cladding, but is rapidly absorbed, resulting in enhanced embrittlement near the burst location. This effect, which is neither burnup nor alloy dependent, was discovered earlier [3] and has been confirmed in the present research program. The localized nature of this effect and the clean fractures that occur when stressed to failure do not suggest a safety issue.

2.4 Hydrogen-enhanced Beta-layer Embrittlement by Oxygen

Some hydrogen from the outside cladding corrosion process is also absorbed in the cladding metal during normal operation. When that cladding is exposed to high-temperature LOCA conditions, the elevated hydrogen levels increase the solubility of oxygen in the beta phase. Thus, even for LOCA temperatures below about 1473 K (1200˚C), embrittlement can occur for times corresponding to less than 17% oxidation, as seen in Fig. 1. This burnup effect was discovered in the current research program and can be accounted for by reducing the time permitted for oxidation at high temperatures in proportion to the amount of corrosion.

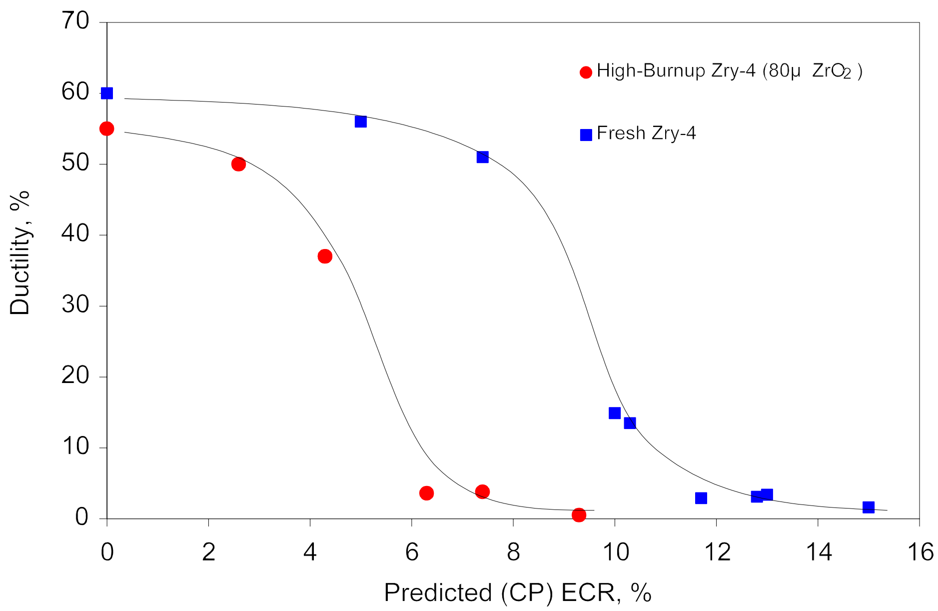

Fig 1. Ductility as a function of calculated oxidation at 1473 K (1200˚C) for irradiated and unirradiated Zircaloy-4 (rough surface), tested at 408 K (135˚C).

2.5 General Hydrogen-induced Embrittlement from Breakaway Oxidation

Zirconium dioxide can exist in several crystallographic forms (allotropes). The normal tetragonal oxide that develops under LOCA conditions is adherent and protective. There are, however, conditions that promote a monoclinic "breakaway" form that is not adherent and permits rapid hydrogen ingress, as seen in a specimen in Fig. 2. One of the conditions is cladding surface

Fig 2. Specimen of E110 cladding that exhibited severe breakaway oxidation after 1400 seconds at 1000˚C.

roughness (belt polishing is beneficial) and another is the impurity content of the zirconium starting material (sponge zirconium is beneficial). Hydrogen that enters in this manner during a LOCA transient has the same effect on embrittlement as hydrogen from the normal burnup process. Although breakaway oxidation was known in 1973, the connection to embrittlement and the factors affecting it were discovered only recently in NRC programs.

2.6 Oxygen Pickup from the Cladding Inside Diameter (ID)

At high burnup, there will be a corrosion layer on the outside diameter (OD) and there can be a bonding layer on the inside diameter (ID) between the UO_2 fuel and the cladding. The corrosion layer and the bonding layer are both largely ZrO_2, and they can provide oxygen for diffusion into the metal. This situation is like a multilayer diffusion couple as illustrated in Fig. 3. The actual thicknesses of the OD oxygen source and of the ID oxygen source are relatively unimportant because they generally contain much more oxygen than will diffuse into the metal (see Hofmann and Politis [4]).

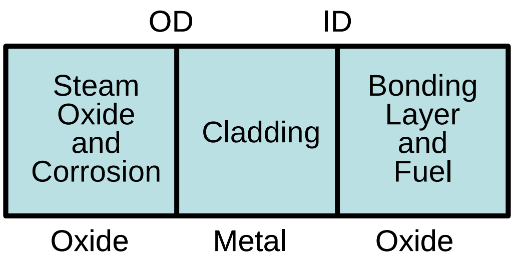

Fig 3. Diffusion couple character of oxygen sources and cladding metal.

2.7 Embrittlement Criteria

No change is foreseen in NRC's 1477 K (2200˚F) temperature limit. For temperatures below that limit, cladding embrittlement, including burnup and alloy effects, has been characterized with limits on time spent at high temperatures from relatively simple measurements. One time limit was obtained from ring-compression tests on unirradiated cladding, oxidized at about 1477 K (2200˚F), in combination with the known corrosion thickness for that type of cladding at the burnup of interest. The amount of oxidation at the onset of embrittlement in the tests was reduced by the corrosion thickness, and that reduced amount of oxidation was related to the time limit by the Cathcart-Pawel equation [5]. The other time limit was obtained from tests on unirradiated cladding specimens that were oxidized at various temperatures to determine the time at which rapid hydrogen absorption began as a consequence of breakaway oxidation. That time was used as the limit when it was less than the one mentioned above. These oxidation-related time limits are similar in form to the current oxidation limit in 10 CFR 50.46. Although NRC's research has found these criteria to be effective, they have not yet been subjected to public comment or adopted in regulations.

3. Reactivity-Initiated Accidents

Data from international programs show that hydrogen absorption related to corrosion is also more important than burnup in determining cladding failure under RIA conditions. Several conclusions were reached that affected the NRC's adjustment (scaling) of test results to account for non-typical test conditions: (a) thermal expansion models in many codes are not suitable for application to RIAs with edge-peaked power distributions, (b) the failure level for mixed-oxide fuel is the same as for uranium-oxide fuel, (c) the failure level for cladding with oxide spallation is the same as for non-spalled cladding with the same average hydrogen concentration, and (d) effective cold gap widths for rapid transients are not always the same as those for steady-state irradiations.

3.1 Thermal Expansion Models

In fuel rod computer codes, most thermal expansion models are based on an implicit assumption that the fuel is hotter in the centre than at the outside surface. In that case, the total expansion is given by the sum of the expansion of each of the rings in the nodal scheme. Although not usually stated, it is understood that ceramic rings have poor tensile strength such that the outer (hence cooler) of two adjacent rings will simply crack without diminishing its thickness when its inner neighbour expands more than it does. This model does not give the correct answer for a pellet that is hotter near the surface than at the centre because of an edge-peaked power distribution. In that case, the outer rings are hotter, and the displacement of the outer surface will not be affected by the inner part of the pellet that is cooler. The FRAPTRAN code was modified to accommodate this effect with a subsequent improvement in its predictions of cladding strain during a pellet-cladding mechanical interaction (PCMI).

3.2 Mixed-Oxide Fuel

The database contains a number of mixed uranium-plutonium oxide (MOX) fuel rods. It has been suggested that inhomogenieties in MOX fuel might affect the mechanical loading on the cladding, thus increasing the strength of the PCMI [6]. The dynamic-gas-expansion hypothesis that would produce this increase does not appear plausible for two reasons. First, it would require rapid swelling of pores in UO_2, and this diffusion-controlled process is probably not fast enough for the 10-to-40 millisecond pulse periods of interest. Second, many of the plutonium-rich islands that are postulated to contribute to this effect are deep within the pellet where their expansion is overshadowed by expansion of the outer rim in these edge-peaked power excursions. Therefore, the PCMI loading, which is driven largely by thermal expansion, should be the same in MOX and UO_2 fuel because thermal properties of MOX and UO_2 are quite similar [7].

3.3 Oxide Spallation

Spallation can lead to the formation of hydride blisters, which in principle might act as defects that could initiate cracks at lower fuel enthalpies. However, the blisters that were found in test rods were relatively small in relation to the cladding thickness, and metallography does not show a significant relation between the blisters and the cracks. This observation is also consistent with recent work by Glendening et al., which concludes that thin blisters are not more effective at initiating cracks than uniform hydride rims [8]. Therefore in the range of 80-110 microns of oxide thickness, spalling does not appear to affect the fracture process. It is also relevant that the absence of spallation could not be guaranteed in commercial fuel rods with corrosion in the range of 80-110 microns.

3.4 Cold Gap Widths

Measured plastic hoop strains for tests with non-failed cladding in Cabri and NSRR are shown in Fig. 4. The zero-strain intercepts of the various data sets correspond to the enthalpy required to close the

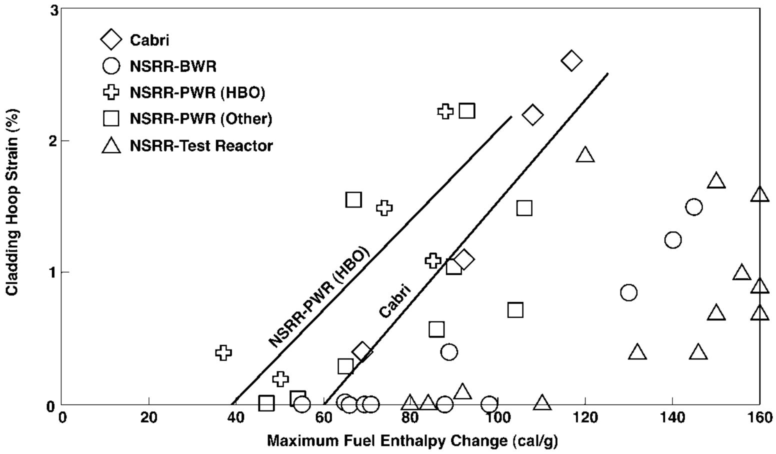

Fig 4. Plastic strain measured from non-failed cladding as a function of maximum fuel enthalpy change for tests in Cabri and NSRR.

gap and to expand the cladding through the elastic range. Even when careful attention was given to the different fabrication and pre-irradiation conditions, the large spread in zero-strain intercept values was not predicted well by our codes. It was, therefore, necessary to override calculated pellet-to-cladding gap sizes to get transient strain predictions that were realistic for these data sets. This behaviour is consistent with large variations in preconditioning of the pellet-to-cladding gap in the different test series.

3.5 Analysis of Data

With modelling assumptions based on these conclusions, an analysis was made that resulted in estimates of the effects of non-typical test conditions on test results [9]. The adjusted failure data are shown in Fig. 5, and the adjusted data are not much different from the raw test data (-80 to +113 J/g, -19 to +27 cal/g).

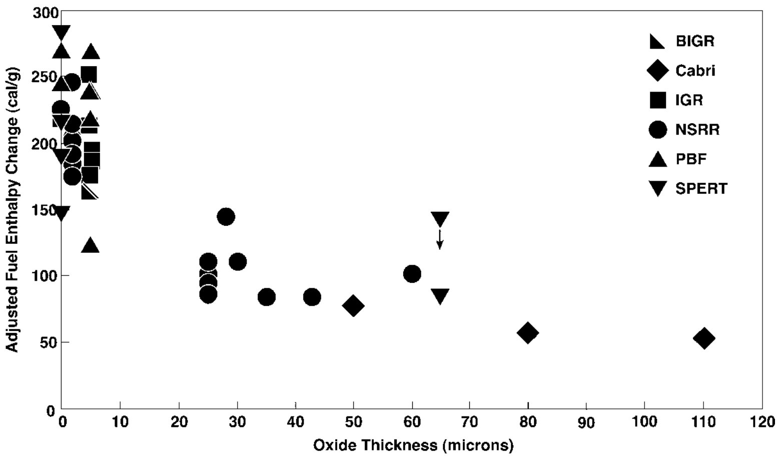

Fig 5. Cladding failure data with adjustments from a scaling analysis to correspond to PWR hot-zero-power conditions.

4. Analytical Capabilities

The NRC sponsors the development of computer codes and uses them to analyse the thermal and mechanical behaviour of fuel rods and to calculate the power level generated in the fuel during transients and accidents.

4.1 Thermal and Mechanical Analysis

The FRAPCON code is used for steady-state and mild transient analysis [10]. A variety of thermal and mechanical phenomenon are considered, including fission gas release. The code has been validated for burnups over 65 GWd/t. Another code, FRAPTRAN, is used for transient and design-basis accident analysis [11]. The NRC staff use these codes to analyse events such as the reactivity-initiated accident and loss-of-coolant accident. To calculate the behaviour of fuel rods with prior burnup, the FRAPTRAN code can be started using a FRAPCON output file.

Accuracy of the codes was assessed by comparing the code predictions of fuel temperatures, fission gas release, rod internal void volume, fuel swelling, and cladding creep, growth, corrosion, and hydriding with data from integral irradiation experiments and post-irradiation examination programs. Predictions from both codes compare reasonably well with the experimental data.

Neither the FRAPCON nor the FRAPTRAN code is intentionally conservative (i.e., both codes have been developed to provide a best estimate of experimental data). Conservative model options are available in the codes, but these are neither recommended nor used by the NRC staff. These options are provided to maintain consistency with previous regulatory assumptions.

4.2 Neutronic Analysis

The PARCS three-dimensional neutron kinetics code is used to calculate the power level generated during a transient or accident [12]. During a postulated rod-ejection accident, for example, the linear power may be a thousand times higher than during normal operation and the shape of the power pulse is determined by the reactivity inserted, Doppler feedback, and other factors. We have explored this relation and found that pulse width varies as a function of the energy deposited during a reactivity transient. High energy pulses are narrow and low energy pulses are broad, as shown in Fig 6.

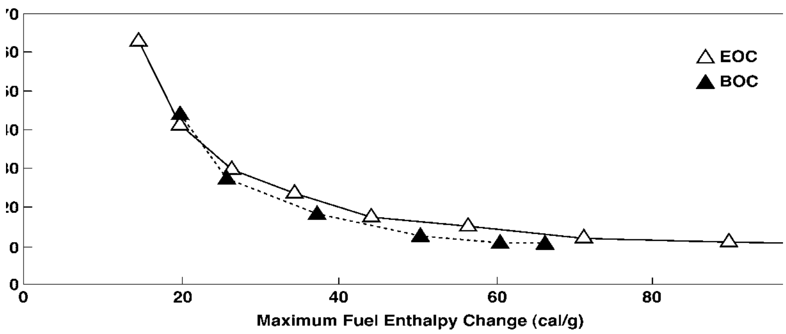

Fig. 6. Dependence of pulse width on energy (fuel enthalpy change) for beginning-of-cycle (BOC) and end-of-cycle (EOC) conditions for a representative PWR core.

The main consequence of this relation is that the energy deposited during a narrow pulse results in a more adiabatic heat-up of the fuel and cladding. Thus testing and analysis should be done with pulse widths that are consistent with the deposited energies.

To complete our assessment of postulated reactivity accidents, plant analyses were performed to examine the conditions necessary to fail the cladding. The results showed that the enthalpy change is only 167 J/g (40 cal/g) for rod worths of $1.5, and it is unlikely that rod worths would exceed this value in commercial power plants.

5. Summary

Based on results of this research, revisions to NRC's regulatory criteria for LOCA and RIA are underway. Although current criteria can be non-conservative under certain conditions, the criteria have been applied in an overall conservative way such that U.S. plants operate with adequate safety margins.

6. References

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