

Rethinking the Zircaloy Embrittlement Criteria and Its Impact on Safety Margin

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1. Introduction

During accidents, nuclear fuel rods are subject to dynamic temperature, pressure, and heat transfer mode transience, which remarkably influences cladding structural integrity. It has been considered that retaining adequate cladding ductility is a simple, yet effective, way of ensuring the structural integrity of fuel rods under unaccountably many accident scenarios. As a consequence, Zircaloy embrittlement criteria has become a basis for the current emergency core cooling system (ECCS) criteria, written in U.S. NRC's 10 CFR 50.46. The current ECCS criteria limit the peak cladding temperature to be 1204°C, and equivalent cladding reacted (ECR) to be 17%, in order to maintain adequate ductility.

Experimental investigations on the Zircaloy embrittlement were conducted in the early 1970s [1,2]. These experiments measured remaining ductility of oxidized and water quenched Zircaloy tubes by the ring compression methods. During 1973 Rule-Making Hearing (from Docket RM-50-1, April 16, 1973), a multi-step procedure was outlined to establish the current ECCS criteria [3]. In 1975, the American Physical Society published a report that outlined insufficient data for the established ECCS criteria [4,5]. As a consequence, a large number of experimental studies have been conducted in 1980s to validate the compliance of the Zircaloy embrittlement criteria with plausible fuel rod failure modes [3,6-10]. These fuel rod failure modes include integral thermal shock fracture, and impact tests. It is quite remarkable to see that the proposed Zircaloy embrittlement criteria attained from ring compression tests, in general, successfully assure structural integrity of fuel rods subject to relevant failure modes in accidents. This fact demonstrates that ductility of Zircaloy is the key metric to structural integrity of fuel rods.

However, the Zircaloy embrittlement criteria set in 1970s inevitably pose limitations that have become increasingly important for today's nuclear fuel and reactor operations. In particular, the criteria do not take into account the steady-state hydrogen embrittlement with burnup. This may be understandable considering the markedly lower discharge burnup in 1970s compared to that of today. The revision of the rule has been already conducted by the U.S NRC [11,12] to

account for high burnup effects on ECR while the temperature limit remains unchanged. The newly proposed rule of the U.S NRC stick to the similar ring compression tests conducted in the early 1970s [1,2]. The new draft-rule proposes a relation that decreases the ECR limit with respect to the pre-transient hydrogen content. The decrease in the allowable ECR value with respect to the hydrogen content is quite significant. The allowable ECR value linearly decreases to ~6% (calculated by the Cathcart-Pawel correlation) with the pre-transient hydrogen content of ~ 400 wppm. Considering the typical range of the pre-transient hydrogen contents [13,14], the newly proposed rule implies a significantly reduced safety margin.

Yet, there is an unsettled regulatory issue surrounding the allowable ECR with pre-transient hydrogen-induced embrittlement. The regulation of Japan is slightly different from that of the U.S. NRC. It limits the peak cladding temperature below 1200°C and the ECR below 15% (calculated by the Baker-Just correlation). This rule is set to assure structural integrity of ruptured fuel rods upon water-quenching. Despite the apparent similarity between the Japanese and the U.S. NRC rules, their difference in the underlying safety philosophy stands out in the high burnup consideration. Japanese Atomic Energy Agency (JAEA) conducted integral fuel rod thermal shock tests with high burnup claddings [14] which contain significant pre-transient hydrogen. Their ring compression tests with segmented specimens obtained near the rupture location showed a clear sign of significant embrittlement [15]. Nevertheless, these investigations confirmed that the original criteria (1200°C and ~15% ECR) still successfully guarantee fuel rod structural integrity upon thermal shock. Hence, the study concludes that the high-burnup effects are inconsiderable as the thermal-shock fracture boundary is not significantly reduced. It outlines that the Japanese ECCS criteria (15% ECR limit) can provide a sufficient margin for high burnup cladding up to ~80 MWd/kgU (~840ppm) [14].

Hence, there exists an obvious disagreement between the embrittlement-based criterion and the thermal shock fracture-based criterion. The regulatory judgement may be bound to a subjective outlook of nuclear safety. However, it important to consider their influence on the safety margin, reactor operation, and associated safety system design.

It is worth noting that the temperature limit (1204°C) of the zircaloy embrittlement criteria has not been altered since it was first determined in 1970s. In the monumental experimental investigation of Hobson and Rittenhouse in 1972 and 1973 [1,2], the experimental evidence for the current 1204°C was first addressed. The study found a reasonably accurate correlation between zero ductility temperature and the sum of alpha and oxide layer thickness for the specimens oxidized below 2200°F (1204°C). However, in spite of the similar oxidation degree, specimens oxidized at 2400°F (1315°C) were markedly more brittle than specimens oxidized at 2200°F (1204°C). The study explained this by the increase in solid-solution hardening due to a higher oxygen solubility at a higher temperature. Such a nice experimental correlation attained between the nil ductility temperature and the remaining beta layer thickness fraction below 1204°C has become a critical basis for the current temperature limit; at 1315°C- the correlation breaks down due to the remarkably increased oxygen solubility in the remaining beta layer. One can note that such a decision based on the two sparingly chosen temperature points (1204°C and 1315°C) leads to the maximum uncertainty of ~ 111°C (=1315 °C -1204°C). Since 1204°C was determined to be the zircaloy temperature limit, its validity has been successfully protected through the concerns on the runaway oxidation above the temperature.

As a consequence, efforts on revisiting the ECCS criteria have been predominantly centered on the ECR level. The runaway oxidation above 1204°C is worth considering. However, such a qualitative concern alone should not limit further discussions on the potential increase in the temperature limit. The concerns on the runaway oxidation above 1204°C requires a further quantitative justification under design based accident scenarios. Indeed, there is ample experimental evidence that supports the possibility that Zircaloy ductility can be maintained beyond 1204°C if it is oxidized for a limited time period. The increase in the temperature margin can lead to a reactor power uprate, or allow an increased accident coping time.

This paper aims to achieve a comprehensive understanding of an engineering compatibility between nuclear fuel behaviors, safety system, and safety criteria, in the pursuit of increasing safety margin of LWRs. Specifically, this study presents a Revision the current temperature limit (1204°C) with adjustable ECR levels through an extensive literature review. Discussion on the impact of safety system capacity increase on the safety margin, with the newly proposed temperature limit in this study is followed.

The impact of this temperature limit on the actual DBA accident margin is discussed with results of the transient thermal-hydraulic analysis simulation code MARS.

2. Review on the current ECCS Criteria with highlight the potential temperature limit increase

Fig. 1 show the summary of experimental failure/non-failure data for both unruptured claddings (Fig.1a) and ruptured claddings (Fig.1b). Fig.1a represent the failure/success map for unruptured fuel rods. This is compatible with the U.S. NRC's approach. Fig. 1b shows the failure/success map based on the Zircaloy rod integral tests that accompanied fuel rod rupture. It is worth noting that the current U.S NRC's criteria set from the ring compression tests can still well assure cladding structural integrity upon the thermal shock of ruptured fuels and their anticipated impacts (0.3J), as shown in Fig.1b.

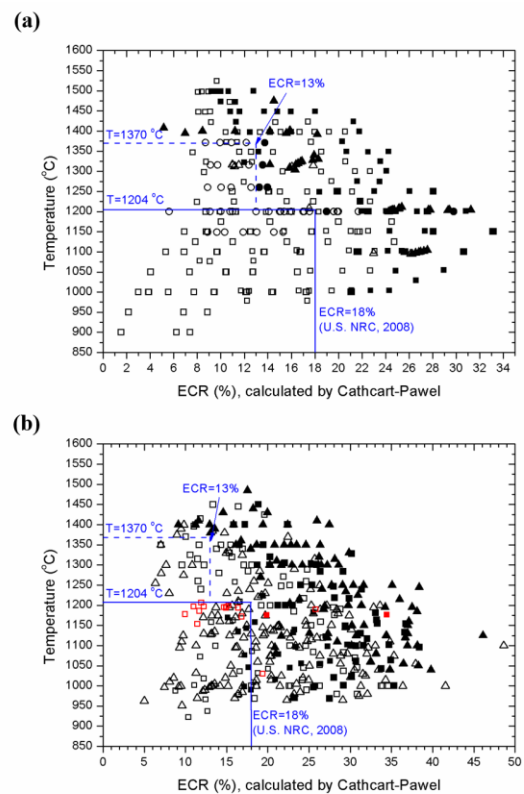


Fig.1 Summary of experimental data for (a) the Zircaloy embrittlement criteria based on the unruptured specimens, (b) Zircaloy fracture based failure map with ruptured rod integral tests (red symbols represent irradiated claddings with partial axial constraints). [Filled: failed, Open: survived, triangle: 0.3J impact test, rectangle: thermal shock fracture tests, circle: ring compression tests at 135°C]

An unhighlighted acceptable ECCS criteria region is newly identified in Fig.1 - 1370°C and 13% ECR. As can be noted in Fig.1, there is ample experimental evidence that supports the possibility that Zircaloy ductility can be maintained up to 1370°C, as long as the oxidation is below 13% ECR by the Cathcart-Pawel

correlation. This new window of acceptable ECCS criteria is also shown to be successfully applicable for the rupture fuel rod integrity.

3. Potential Gain in the Safety Margin

KINS-Realistic Estimate Method (KINS-REM) is the best estimate method with uncertainty treatment developed by KINS. This method adopts Wilk's formula for non-parametric statistic which enables the fixed number of code calculations for 95% probability level regardless of the number of input variables. According to Wilk's third-order formula, it is indicated that the representative value with 95% probability and 95% confidence level is same to 3rd largest value from the calculations of 124 cases. Therefore, 124 input cases determined by simple random sampling with 22 uncertainty parameters were calculated to find the case which had PCT95/95 by using MARS-KS code (1.3 version). The fundamental code input was based on UCN 3&4(OPR-1000) in Korea.

In this paper, the power, one of the uncertainty parameter, was fixed to the certain value (1.0, 1.1, 1.15, 1.2...) to simulate the power uprating. Also, to maintain the heat balance against this change the steam and feedwater flow rate of secondary system were increased with the same ratio of uprating.

Fig. 2 shows the results of the MARS simulation for the transient fuel rod temperature and ECR during LBLOCA, with increases in the reactor power. It can be shown seen clearly from Fig.2 that the new ECCS temperature limit window can allow the power increase to 10%. Without the newly proposed temperature limit, it would not be possible to allow any room for a potential power uprate unless there is a reduction in the temperature margin below $\sim 104^{\circ}\text{C}$ ($= 1204^{\circ}\text{C} - 1000^{\circ}\text{C}$).

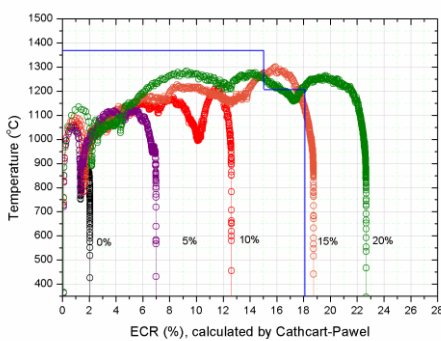


Fig.2 Comparison of the newly proposed acceptable ECCS criteria (blue lines) with the actual fuel rod temperature and ECR during the LBLOCA transience with increases in the reactor power.

3. Conclusions

It is considered to increase the temperature limit of the current ECCS criteria upto 1370°C as long as ECR

is below 13%. This could lead to an increased safety margin that can be readily translated into a potential power uprate upto 10%. Such a safety margin increase is anticipated to increase even more if there is an increase in the ECCS capacity (e.g. water inventory increase in safety injection tank or flow rate increase of low pressure safety injection pump). The influence of these safety system capacity on the safety margin with the newly proposed criteria are under investigation.

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