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LOCA Criteria Basis and Test Methodology

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Cover photos: ring compression test (JAEA, Japan); cross-sections of fuel cladding oxidised under LOCA conditions (ANL, United States).

Foreword

Acceptance criteria for emergency core cooling systems (ECCS) define the maximum temperature and degree of oxidation in order to avoid excessive embrittlement and hence failure of the fuel cladding, which would affect core cooling in the case of a loss-of-coolant accident (LOCA). The criteria are mainly based on experimental data obtained in the 1970-80s. The semi-integral quench test, the quench test without mechanical loading, the ring compression test (RCT), the bending test and the impact test have been performed to evaluate structural integrity and embrittlement of the cladding under LOCA conditions, and consequently different test methodologies have been used for determining the cladding embrittlement criteria. The trend towards high burn-up and the use of new cladding alloys has increased the need for international discussions on these test methodologies and acceptance criteria.

The NEA Committee on the Safety of Nuclear Installations (CSNI) noted that these different approaches produced nearly identical limits. It therefore asked the CSNI Working Group on Fuel Safety (WGFS), a body tasked with advancing the understanding of fuel safety issues by assessing the technical basis for current safety criteria and their applicability to high burn-up and to new fuel designs and materials, to write the present technical opinion paper on the LOCA criteria basis and test methodology. The purpose of this paper is to review the LOCA criteria basis and the different test methodologies, and to provide recommendations to the international community on how the results of these different methodologies can be applied for regulatory purposes.

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Executive summary

The emergency core cooling system (ECCS) acceptance criteria define the maximum temperature and degree of oxidation in order to avoid excessive embrittlement and hence failure of the fuel cladding, which would affect core cooling in the case of loss-of-coolant accident (LOCA). The criteria are mainly based on experimental data obtained in the 1970-80s. The trend towards high burn-up and the use of new cladding alloys has increased the need for international discussions on the test methodologies and acceptance criteria on cladding embrittlement during a LOCA.

The semi-integral quench test, the quench test without mechanical loading, the ring compression test (RCT), the bending test and the impact test have all been performed to examine structural integrity and embrittlement of the cladding under LOCA conditions. Results from RCT were the main basis for the criteria in the United States and many other countries. Japan and Russia originally based their criteria on similar data and considerations; however, they changed their bases, taking into account additional information from the semi-integral quench test. Historically, other test methodologies and parameters have also been tried for determining the embrittlement criteria.

The NEA Committee on the Safety of Nuclear Installations (CSNI) noted that these different approaches produced nearly identical limits in review of high burn-up RIA (reactivity initiated accident) and LOCA criteria [1]. The CSNI Working Group on Fuel Safety (WGFS) proposed to write the present technical opinion paper (TOP) on the LOCA criteria basis and test methodology, and CSNI approved the proposal. The purpose of this paper is to review the LOCA criteria basis and the different test methodologies, and to provide recommendations to the international community on how the results of these different methodologies can be applied for regulatory purposes.

The quench test without mechanical loading is the first approach for evaluation of the quench-bearing capability of the cladding and addresses only thermal stress. The semi-integral quench tests under axial constraint provide information on the structural behaviour of the fuel rod under simulated LOCA conditions and on the strength of the fuel rod, allowing the occurrence of the fracture, if any, at the weakest point. This kind of test simulates the LOCA sequence up to the quench phase. A fracture/no-fracture criterion can be directly derived by incrementing the high temperature oxidation time. More investigations are needed to define an appropriate loading to assess the cladding integrity during and after the quench phase.

The RCT has the advantage of requiring smaller specimen volume, easier specimen preparation and easier specimen set-up in the test apparatus. At the same time, it may be considered as a "local material property" test, since it addresses the behaviour of a small length of cladding, without fuel. The stress applied during the test is also localised and may be higher than that generated during quenching. The aim of this mechanical test is to produce a limit such that the cladding remains ductile, and in doing so, to avoid discussions about the loads that the cladding must sustain during or after a LOCA. Among ring compression tests conducted in different laboratories, it is important to have a common definition of ductile-to-brittle transition. In addition, more data and discussion are required about test procedures and conditions, such as oxidation temperature, test temperature, sampling position from oxidised/ruptured cladding and cooling scenario, in order to obtain a clear consensual criterion for cladding embrittlement.

Since specimens for RCT are generally taken from non-ballooned region, they may not fully represent the behaviour of the cladding in the balloon area nor fully represent other phenomena (e.g., secondary hydriding). To overcome this problem, the bending test can be used. In this sense, the bending test is simply considered another method of observing the mechanical response of the cladding, but now includes the ballooned region. There is only limited experience in the use of the bending test for the investigation of cladding behaviour in the ballooned section compared to RCT or semi-integral tests. It would be important to carry out bending tests in different laboratories in order to analyse the applicability of this type of tests for the establishment or confirmation of LOCA criteria.

When comparing the results of RCT and semi-integral testing the following must be taken into account:

- The two methods consider different mechanisms in order to evaluate the embrittlement process of Zr cladding in high temperature steam. RCT samples are usually oxidised in steam, as are in the quench tests. Semi-integral quench tests may better represent the behaviour of fuel rod under LOCA accidents including clad ballooning and secondary hydriding on the inner surface of the cladding after burst.
- The two methods give information on somewhat different phenomena at different degrees of embrittlement. The final result of RCT is the determination of ductile-to-brittle transition. The transition can be determined from load-displacement curves and the conditions are very reproducible. On the other hand, the semi-integral quench tests are "strength tests" to examine resistance of the oxidised cladding to stress caused by thermal shock and other mechanical loadings. The fracture of embrittled cladding may be less reproducible.

Furthermore the RCT and semi-integral quench tests, which are carried out in different laboratories or in different test series, may not be conducted in the same way. This can lead to rather different results.

The present TOP introduces and compares the advantages and drawbacks of the existing test methodologies, and gives recommendations for each methodology.

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However, it does not specify any standardised procedure for the experimental conditions to establish LOCA criteria because of several reasons:

- Each experimental set-up and procedure has special advantages and limitations. Test conditions and methods of data evaluation vary in different laboratories involved in testing.
- Data produced in different laboratories and facilities on the embrittlement of Zr cladding under similar conditions are generally comparable to each other. Consequently, the differences in embrittlement test methodologies do not seem to be of primary importance. However, the impact of test conditions, for example, slow-cooling versus quenching, needs to be addressed while drawing conclusions for LOCA criteria.
- The establishment of LOCA criteria includes conservative assumptions which are not the same in all countries and which need specific interpretation of experimental data.

1. Background

The postulated loss-of-coolant accident (LOCA) is one of the design basis accidents (DBA) for water cooled reactors. A LOCA is caused by a break in the reactor coolant pressure boundary. It is assumed that the reactor is automatically shut down, but the temperature of the reactor core continues to rise due to the radioactive decay in the fuel as well as the reduction or loss of coolant. At the time this event was defined as a DBA it was generally accepted that preventing fuel from melting and avoiding excessive fuel dispersal would be necessary to minimise radiological consequences to the public. For this reason, it was required to design emergency core cooling systems (ECCS) to ensure that the fuel could be sufficiently cooled during all phases of the DBA.

During a LOCA, the cladding may be heated up to temperatures over 1 000 °C and it reacts with coolant. The alpha-phase zirconium starts to transform to the beta phase when the temperature reaches about 800 °C. At the same time, oxygen dissolved in the metal stabilises the alpha phase, and a layer of alpha phase with a high content of oxygen begins to grow on the beta phase underneath the oxide scale. The oxide itself is very brittle and oxygen also embrittles the alpha phase of the base metal. The severely oxidised cladding may shatter when the hot fuel rod is quenched back to low temperatures. Therefore, a limit clearly must be placed on the amount of growth of oxygen-stabilised alpha phase and oxide layer and diffusion of oxygen into the beta phase. This limit is necessary to ensure that the load-bearing prior-beta layer is thick and has low oxygen concentration sufficient to preserve structural integrity of the fuel rod when the fuel rods are quenched and subjected to various types of stresses.

In 1971, the United States Atomic Energy Commission (USAEC) promulgated acceptance criteria for the ECCS which stated that “the clad temperature transient is terminated at a time when the core geometry is still amenable to cooling, and before the cladding is so embrittled as to fail during or after quenching” [2]. These criteria were subjected to a rulemaking hearing in 1973 that was extensively documented in the *Journal of Nuclear Safety* in 1974 [3, 4].

At that time, it was well understood that embrittlement of zirconium-alloy cladding is caused by the diffusion of oxygen into the metal underneath the surface oxide rather than by the surface oxide itself. That is, although the best measure of the heat generation from zirconium metal-water reaction (hence, core heat-up) is total oxidation, cladding structural integrity is really controlled by the small fraction of oxygen atoms that is dissolved in the remaining beta zirconium layer. It appears that, out of simplicity and in absence of a better method of calculating oxygen distribution in the metallic layers, the USAEC regulations used the time required to accumulate 17% equivalent cladding reacted (ECR) based on Baker-Just model as a surrogate measure of the time required to diffuse enough oxygen into the underlying metal to embrittle it. In doing so, the United States regulations also imposed an additional limit on peak cladding temperature (PCT) of 2 200 °F (1 204 °C). Other countries basically followed the United States though some adopted slightly different limits on the basis of their own data and considerations.

The trend towards high burn-up and the use of advanced cladding alloys has increased the need for international discussions on the test methodologies and acceptance criteria on cladding embrittlement during a LOCA. Different methodologies have been adopted to derive the LOCA criteria in the NEA member countries. The purpose of this technical opinion paper (TOP) is to review the LOCA criteria basis and the different test methodologies, and to provide recommendations to the international community on how the results of these different methodologies can be applied.

2. Existing test methodologies

For the safety demonstration, several types of tests including the semi-integral quench test and the quench test, the ring compression test, the bending test and the impact test have been performed to evaluate the structural integrity and embrittlement of the cladding. Detailed descriptions of the tests are summarised in the WGFS's "Technical Note on LOCA Fuel Cladding Test Methodologies – Compilation of Responses and Recommended Test Characteristics" [5].

2.1. Quench test

2.1.1. Semi-integral quench test

Short test fuel rods (about 200 to 600 mm) fabricated with cladding tubes and UO_2 pellets (or dummy pellets) are exposed to steam at high temperatures and finally quenched by the flooding water in the semi-integral quench test [6-10]. The rods balloon and rupture during the heat-up, and they are isothermally oxidised at pre-determined temperatures for various periods.

Hotter fuel rods and cooler guide tubes show different axial expansion and shrinkage. Since some phenomena, such as ballooning of the rod and the rod-grid chemical interaction, can restrict the axial displacement of the fuel rod in the spacer grids, the fuel rod may be axially constrained. The axial constraint is remarkable in the fuel bundle in which the guide tubes are mechanically fixed to the spacer grids. Because of these factors, fuel rods during reflooding will be subjected not only to complex thermal shock and hydraulic loads but also to the axial tensile load due to the constraint. Hence, the test rods are often cooled and quenched under restrained conditions in the semi-integral tests. In a high-burn-up fuel rod, pellet-cladding bonding can be so tight that axial shrinkage may be restricted though the bonding may be vanished during the LOCA transient. Mechanical loading during and after the quench phase is considered complicated in the bundle geometry, and therefore it is difficult to evaluate conditions and level of the mechanical loading. For example, possible loading in the radial direction which is suggested by the in-pile tests [11, 12] is not considered in the above tests.

If the cladding is embrittled by oxidation and secondary hydriding, it may fracture on quenching. The cladding fracture generally depends on the oxidation amount and the mechanical load. The results are summarised on maps showing fracture/no-fracture conditions relevant to oxidation amount and oxidation temperature (or pre-existing hydrogen in the case of high burn-up fuel cladding).

2.1.2. Quench test without mechanical loading

The degree of a grid-to-rod lockup due to any kind of chemical or eutectic interaction is a matter of controversy. Also, if the rods would not balloon within the spacers (the spacer vanes cool down the fuel rod locally), the grid to rod interaction would be very small and the axial constraint applied to the rod would be negligible [13, 14]. In this respect, the thermal shock and hydraulic loads induced by the quench may be the main loadings applied to the fuel rod during the quench phase. The quench test without mechanical loading which has been performed in France [15] could be useful in investigating the thermal stress bearing capability. Short cladding samples (non-irradiated and pre-hydrided) are exposed to double-side steam oxidation and then quenched directly by dropping the samples into cold water. From the test, limits can be determined based on the ECR value corresponding to the cladding fragmentation caused by thermal stresses on quenching or on the ECR value leading to the first leak (first micro-crack detected in the cladding using a post quench helium leak test). The rod balloon and rupture were not simulated and, therefore, the influence of the secondary hydriding was investigated by pre-hydriding up to 5 000 ppm in the previous study [15].

2.2. Post-quench tests

2.2.1. Ring compression test

The ring compression test is a standard ductility screening test used for materials that transition from ductile to brittle failure. It is not designed to represent any particular loading conditions. Ring-compression test results were used in 1973 to define the ECR and PCT limits to ensure post-quench ductility.

However, the methods used by Hobson [16, 17] to determine oxidation temperatures and levels leading to brittle behaviour were crude. Rings were crushed to complete failure – often four cracks at four positions relative to the loading platen. The pieces were reassembled. If the cracked pieces formed a circular shape vs. an oval shape, they were classified as brittle. In addition, a microscopic examination of the rupture area was performed. Load-displacement curves for these tests were used to define zero ductility with results from the macro and microscopic observations but the curves were never published though they were considered in the 1973 hearing.

In the recent work at Argonne National Laboratory (ANL) [18], ring specimens were compressed at a low-displacement rate in a testing machine. A first significant load drop would indicate a through-wall crack along the length of the sample. The sample was hypothetically unloaded from the peak load before the load drop by using the slope of the elastic region of the load-displacement curves to determine the offset displacement. This offset displacement is normalised to the pre-test outer diameter of the cladding to determine a relative plastic strain (offset strain). For oxidised samples, controlled unloading is not possible just prior to failure because the displacement at which failure will occur is unknown and the offset strain is overestimated. Accordingly, a limiting offset strain of 2% is assumed in the determination of the ductile-to-brittle transition. In other words, samples with ≥2%

offset strain are considered ductile. For failure with offset strains <2%, it is not clear if they are ductile or brittle. Therefore, a complementary criterion was set on the permanent strain (with a limit at 1%) and the associated procedure in this test series to identify if these samples were ductile or brittle.

It should also be noted that the 17% ECR[1] limit was derived on the basis of zero ductility data obtained from the slow compression test only (i.e., apparently excluding similar data obtained from high strain rate or impact-type ring-compression test). This exclusion was not discussed in the 1973 hearing.

There seems to be no precise definition on what residual ductility metric should be applied in the ring compression test. In general, the residual ductility decreases rapidly in a relatively narrow ECR interval and the critical ECR may be derived by extrapolating the fall in residual ductility to zero or 2% limit on offset strain or 1% limit on permanent strain [18, 19]. Another definition of ductile-brittle transition has been proposed with use of the strain energy per mm of specimen length spent during the compression test up to the formation of the first through-wall crack [20]. In the Czech Republic, an additional empiric criterion called “K” has been developed [21-23].

2.2.2. Bending test

Ring compression specimens have been taken from the ballooned region of ruptured and oxidised cladding by both Chung [6] and Uetsuka [7]. Results have clearly shown the presence of brittle zones in and near the balloon. Consequently, there is a need for a suitable test to investigate the post-quench mechanical behaviour of ballooned fuel rods. A limited number of bending tests have been conducted at ANL [18] and USNRC plans to conduct additional bend tests at ANL and Studsvik.

Tube bending test is a possible test methodology to confirm the embrittlement criteria. The cladding is stressed axially which is perhaps a more realistic loading situation in relation to LOCA loads, such as spacer grid interactions, flow-induced vibrations, or seismic accelerations. In the bend tests the stress distribution is nominally uniform over the specimen thickness while its magnitude and sign varies around the circumference:

- In the 3-point bending tests the failure location is expected to be at the location of the applied load; for investigation of the behaviour of the balloon, this bias in the failure location minimises the interest for such tests. However, this test is adequate for investigation of the behaviour of the rest of the cladding [24, 25].
- In 4-point bending tests, a constant bending moment is applied between the two inner supports. This test, with fuel pellet inside, is most prototypical for investigating the behaviour of the ballooned region of a fuel rod. Thus, the sample is allowed to fail at its weakest point, which would normally be in the

1. ECR: equivalent cladding reacted (fraction of cladding thickness oxidised assuming that all absorbed oxygen forms stoichiometric ZrO_2).

burst region, the neck-and-beyond region or anywhere within the balloon, depending on thickness as well as oxygen and hydrogen content in the post-quench sample.

Application of various bending test results in regulatory analysis may presume some knowledge of LOCA loads. In generating bending test results, it should be noted that the failure location and failure energy will depend significantly on the orientation of the bending loads with respect to the burst opening. The lowest failure energy is expected if the 4-point bend test is conducted with the burst opening under bending tension. Like the ring compression test, it may be appropriate to simply use the bending test to establish ductile-brittle transition rather than establish the lowest failure energy with incomplete knowledge of LOCA loads. Finally, the bend test is clearly the most expensive test in that it requires the greatest length of tube specimens per test. However, the bending test has the significant advantage of testing a specimen that includes the ballooned region.

2.2.3. Impact test

Impact testing has been widely used in the programme conducted in the early 1980s at ANL by Chung and Kassner to investigate the post-quench load bearing capability of oxidised cladding [6]. It should be noted that most of these specimens survived quench, even at high levels of oxidation. Consequently, there was interest in establishing the mechanical margin-to-failure in those specimens that survived. The results of these tests were considered in the review of the technical basis of embrittlement criteria in the 1988 hearing, but the original limits remained unchanged.

Such tests are also performed in the French *Commissariat à l'énergie atomique* (CEA) [26, 27] and the results were compared to 3-point bending tests and RCTs.

There are also practical reasons to perform impact tests in the LOCA context: some of the loads of concern may be in the form of impact, for instance rod-to-rod impact in ballooned regions during quenching or post-quench handling.

The impact test may be considered as a high-strain-rate 3-point bend test, with failure location biased to the location of applied impact. This is, however, of little importance for assessing the mechanical behaviour in homogeneously oxidised regions (outside the balloon). The advantage of impact testing, in comparison with tube bending, is to possibly perform several impact tests on the same cladding tube after survival to impact at the chosen energy, and to test each part of the tube independently from the others, as done by Chung and Kassner [6].

2.2.4. Ring tensile test

The ring tensile test is almost as simple to perform as a ring compression test. This test is often used to evaluate stress-strain relation in the hoop direction of the fuel cladding. The main difference between the ring compression test and the ring tensile test is the stress distribution over the specimen wall thickness. In the compression test it is a typical bending stress which goes from compression to tension. In the ring tensile test the stress distribution is approximately a uniform tensile stress though bending stress is applied at the early stage of the test due to stretching of the ring. The absolute values of recorded load-displacement curves in ring tensile, ring compression and bending tests are different due to different loading conditions and geometrical arrangements. The ductile-to-brittle conditions derived from any of these tests should be close to each other, since the loss of ductility does not depend on the applying test method, it characterises the cladding state after the oxidation process.

2.3. In-pile integral test

In-pile integral tests like those performed in the power burst facility (PBF), PHEBUS and Halden reactor [28] are useful to simulate more prototypical conditions, which cannot be reproduced in the out-of-pile tests (e.g. irradiation of the fuel rod, power generation inside the UO_2 pellet by fission or relocation of fragmented fuel pellets into the ballooned area). Nevertheless, the test conditions are not always such that the experimental outcomes can be used directly to represent LOCA conditions. In particular many of the in-pile LOCA experiments, except the PHEBUS tests [11, 12] are single rod tests (which tends to enhance the ballooning and make the high temperature oxidation phase less prototypical), with no depressurisation and/or quench phases. As a consequence, those in-pile integral tests have to be properly instrumented, analysed and interpreted with validated codes, so as to allow a reliable transposition from the in-pile conditions to the in-reactor conditions [11, 12, and 29].

3. Main results from different methodologies

3.1. Quench test

The semi-integral quench tests have been performed at ANL and the Japan Atomic Energy Agency (JAEA) (previously JAERI) with unirradiated cladding specimens in the 1980s [6, 9]. The researchers reported that the fracture threshold lied between 38 and 40% ECR under unrestrained conditions. This result is consistent with the quench tests without mechanical loading conducted in France [15], except for the highly pre-hydrided cladding (2 000-5 000 ppm H) which exhibited the lower threshold of about 20% ECR upon direct quenching from the oxidation temperature. The JAEA tests have also been conducted under restrained conditions and it was shown that the threshold decreases to about 20% ECR under the fully restrained condition.

In response to the recent concern regarding the high burn-up, the hydrogen effect on fracture/no-fracture threshold has been also investigated, because hydriding has a great impact on cladding ductility in general [30]. JAEA results indicate that the fracture/no-fracture threshold on the oxidation amount decreases as both the initial hydrogen concentration and axial restraint load increase. However, the fracture threshold was found sufficiently higher than the 17%-limit even if the restraint of about 540 N was applied in the tests. The quench tests have been performed with new alloys such as M5® and HANA claddings. No obvious alloy effect has been found in the fracture conditions [31, 32].

Recently, the semi-integral quench tests with the same axial restraint condition were performed with high burn-up fuel cladding (66-77 GWd/t, <840 ppm of hydrogen) at JAEA [33]. The cladding alloys were MDA, ZIRLO, M5, NDA, and Zircaloy-2. The cladding samples were oxidised at temperatures from about 1 190 to 1 210 °C and to the oxidation amounts from about 18 to 38% ECR-BJ[2] (13 to 28% ECR-CP[3]). Results indicate that the effects of high burn-up are not significant in terms of oxidation, ballooning, and rupture behaviour. The fracture/no-fracture boundary is not reduced significantly by the high burn-up and the use of new alloys in the examined burn-up range, although it may be somewhat reduced with hydriding developed during the reactor operation, as observed in the unirradiated Zircaloy-4 cladding pre-hydrided to the same level. In other words, the fracture boundary of

2. ECR calculated with the Baker-Just equation.
3. ECR calculated with the Cathcart-Powel equation.

the high burn-up cladding under the tested conditions is higher than the limit in the Japanese LOCA criteria (15%-ECR), in the burn-up range up to about 80 MWd/kg and/or up to about 840 ppm of hydrogen.

3.2. Post-quench mechanical test

3.2.1. Ring compression test

Ring compression tests of oxidised specimens were recently conducted with unirradiated and irradiated cladding that had been sectioned from high burn-up fuel rods (63–70 MWd/kg) at ANL [18]. Results of the tests indicated that hydrogen produces the main burn-up effect on embrittlement and the embrittlement threshold (the permanent strain <2%, see Section 4) obviously decreases with the hydrogen content. In addition, embrittlement of the cladding is dependent on cooling conditions. As a consequence, for high-burn-up Zircaloy-4 and ZIRLO with high hydrogen content (<700 ppm), post-LOCA embrittlement thresholds are bounded by 8–9% (no quench) and 5% ECR-CP (quench at 800 °C), which is much lower than the current United States safety limit. For high-burn-up M5 with lower hydrogen content (<150 ppm), the embrittlement threshold is bounded by 18% (no quench) and 14% ECR-CP (quench at 800 °C).

At ANL specimens of different alloys have been tested in the same equipment which makes the results comparable with regard to the performance of the different alloys. One of the first findings in that research was that the embrittlement threshold for unirradiated conventional Zircaloy (Zr-1.4%Sn) is not fixed at exactly 17% as defined in the United States regulations. Namely, three different manufacturing tube specimens of Zircaloy-4 exhibit thresholds from 15.6 to 19%. In other tests involving the alloys M5 and E110, which have nominally the same (Zr-1%Nb) composition, very different embrittlement thresholds were exhibited. Zircaloy-2 and ZIRLO showed embrittlement thresholds (19%) that are very similar to that of the other modern cladding materials that are manufactured with similar techniques. Consequently, the variations in embrittlement threshold can be attributed to by manufacturing differences rather than specific alloy composition.

Ring compression tests and related detailed analyses have been performed at CEA to compare ductility reduction of oxidised and quenched Zircaloy-4 and M5 specimens and the information has been obtained regarding the hydrogen effect and the mechanism of post-quench embrittlement of the alloys [24, 26]. Similarly, AEKI has performed the tests with oxidised E110 and Zircaloy-4 cladding and demonstrated embrittlement of these alloys [20, 34].

3.2.2. Impact test

In 1980, ANL performed pendulum impact tests at 23 °C on pressurised Zircaloy-4 tubes that were burst, oxidised, cooled at ≈5 °C/s, and survived quenching thermal shock [6]. The results of the 0.3-J impact tests indicate that the 17%-ECR limit is adequate to prevent a burst-and-oxidised cladding from failure under 0.3-J impact at 23 °C, as long as peak cladding temperature remained <1 204 °C. The results from the same tests were converted to a failure-survival map based on average hydrogen

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content of the impact-loaded local region and the thickness of transformed-beta layer containing <0.7 wt.% oxygen. On the basis of the map, ANL proposed to replace the 1 204 °C PCT and 17% ECR criteria by a unified criterion which specifies that the thickness of transformed-beta layer containing <0.7 wt.% oxygen shall be >0.3 mm. This approach was not adopted.

CEA has performed impact tests with pre-notched samples which were oxidised from one side and finally quenched. For the given geometry and test conditions used, the fractograph analysis showed a threshold for ductile to brittle fracture of 0.05 J/mm^2. Furthermore, an embrittlement criterion based on oxygen content, hydrogen content, ZrO_2, alpha (O) and prior-beta thicknesses was proposed [27]. However, this new criterion can be considered as a refinement of the previous criterion proposed by ANL, that is, "the thickness of transformed (prior)-beta layer containing <0.7 wt.% oxygen shall be >0.3 mm" [5].

4. Rationale for LOCA criteria basis

Results from ring compression tests became the main basis of the criteria in the United States and many countries. Japan and Russia also established their criteria based on similar data and considerations; however, they changed their bases later, taking account of additional knowledge, from semi-integral quench tests. The rationales behind the two methodologies are briefly summarised below. Historically, some other test methodologies and parameters have been used for determining the embrittlement criteria.

4.1. Rationale for use of semi-integral test

After the establishment of the LOCA criteria in the 1970s mainly based on the results from the ring compression tests, experimental data on fuel behaviour in a LOCA were additionally accumulated in the 1980s. One of the major findings was secondary hydriding caused by inner surface oxidation of the cladding around the rupture opening [7]. It was shown that embrittlement of an oxidised cladding is enhanced by secondary hydriding. In addition, it was reported that cladding deformation due to ballooning and rupture has the great influence on the cladding embrittlement. Therefore, the sequences of ballooning, rupture, oxidation, secondary hydriding and thermal shock should be considered in evaluating cladding embrittlement in a LOCA [9]. If one considers that the greatest impact is the thermal shock and the mechanical loading during quench, the integral (or semi-integral) quench test simulating the LOCA sequences provides key information for integrity of the cladding in a LOCA.

Conditions for extensive fragmentation of the fuel rod materials, which would harm the coolability of the reactor core, may be used to ensure the safety in a LOCA. However, it is difficult to quantify the “extensive fragmentation”. Therefore, a fracture/no-fracture boundary determined by the semi-integral quench test can be used to preserve coolable geometry of the core under LOCA conditions with sufficient margin.

If one accepts the premise that the greatest impact is the thermal shock and the other loads can be ignored, the quench test protocol based on loss of leak tightness detection could be the approach to demonstrate a long-term coolable geometry by guaranteeing no fuel rod fragmentation at all. Otherwise, one must seek alternative test methods that are not so dependent on a better understanding of mechanical loading during the accident.

4.2. Rationale for use of ring compression test

In the early 1970s, Hobson and Rittenhouse performed ring compression tests at 23–150 °C with Zircaloy-4 cladding tubes that were oxidised in steam on two sides, followed by direct quenching from high temperatures (927-1 315 °C) into cold water. They determined zero-ductility threshold using the beta-layer fraction in the wall thickness of the oxidised cladding as an indicator. The 17%-ECR criterion was primarily based on the results of their post-quench ductility tests, taking into account other data, including results from quench tests of Hesson *et al.* [36] and ring compression tests after single-side oxidation of Combustion Engineering.

The USAEC staff believed that quench loads are likely the major loads, but the staff did not believe that the evidence was as yet conclusive enough to ignore all other loads. They also believed that it might not be possible to anticipate and calculate all of the stresses to which fuel rods would be subjected in a LOCA and afterwards. Therefore, retention of ductility was considered the best guarantee against potential fragmentation under various types of loading (thermal-shock, bundle constraints, hydraulic loads, rod-to-rod impact, and handling), and the results from the ring compression tests were adopted as the main basis of the criteria.

Results from unconstrained quench tests were considered only corroborative and reassuring. The USAEC staff wrote during the 1973 hearing that “the loads due to assembly restraint and rod-to-rod interaction may not be small compared to the thermal shock load and cannot be neglected”. Lack of data on loading level and fracture conditions appears to be the reason why the use of quench test data was not accepted for regulatory purposes at that time.

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5. Comparison and discussion

5.1. RCT vs. semi-integral quench test

When comparing the results of RCT and quench testing the following must be taken into account:

- The two methods consider different mechanisms in order to evaluate embrittlement process of Zr cladding in high temperature steam:
  - RCT samples are usually oxidised in steam (as are integral tests).
  - Semi-integral quench tests may better represent the behaviour of fuel rod under LOCAs including clad ballooning and secondary hydriding on the inner surface of the cladding after burst.
- The two methods give information on somewhat different phenomena at different degrees of embrittlement:
  - The final result of RCT is the determination of ductile-to-brittle transition (zero ductility or embrittlement threshold). The transition can be determined from load-displacement curves and the conditions are well reproducible.
  - The semi-integral quench tests with mechanical loading are "strength tests" to examine resistance of the oxidised cladding to axial stress caused by thermal shock and the other mechanical loadings. Radial loading which may be applied in the bundle geometry is not currently taken into account. The fracture of embrittled cladding may be less reproducible.

Table 1. Comparison of RCT and semi-integral quench tests

	RCT	Semi-integral
Embrittlement due to high-temperature Zr oxidation	Yes	Yes
Effect of in-service corrosion and hydriding	Yes	Yes
Ballooned region is tested	No*	Yes
Effect of secondary hydriding	No*	Yes
Ductile-to-brittle transition of Zr cladding is determined	Yes (ductility test)	No
Conditions of fuel rod fracture are observed (resistance to axial stress is examined)	No	Yes (strength test)

* It is possible to be conducted or examined, but it has not been applied to demonstrate the safety.

Furthermore the RCT and semi-integral quench tests carried out in different laboratories or in different test series may not be conducted in the same way. This can lead to different results.

- In case of RCT, both the oxidation temperature and the compression testing temperature have important effects on the final results. The compression testing temperature is important, since the ductile-to-brittle transition at elevated temperature (i.e., higher than room temperature) corresponds to a higher ductile-to-brittle transition ECR.
- If the cladding sample is quenched by cold water after oxidation (which may take place with either test method), its internal structure will not change, while in case of slow cool-down the diffusion of oxygen, hydrogen and some alloying elements can lead to structural changes and these changes can also have an effect on the conditions of ductile-to-brittle transition. The effect of cooling conditions (cooling rate and quenching temperature) have been investigated with non-hydrided and hydrided samples in different laboratories, using different test protocols [6, 27, 37, and 38]. However, the results do not always agree. Therefore, more investigations are necessary to obtain the clear picture about the influence of the cooling conditions on ductility reduction of the high burn-up fuel cladding.
- Figure 1 illustrates additional issues in comparing information from ANL and CEA. It should be understood that the test protocols used by CEA and those used at ANL are different. First, the CEA tests are one-sided oxidation tests whereas the ANL tests are double-sided tests. Second, the CEA testing temperature is ramped very quickly to 1 200 °C (2 194 °F) whereas the ANL tests approach this peak temperature more slowly to better simulate expected LOCA conditions. As explained in the previous two sections, it is not sufficient to examine only ECR. The temperature at which the oxidation occurs plays an equally important role in establishing whether the material is brittle or ductile. Third, for most tests, CEA quenches the sample from 1 200 °C (2 194° F) while ANL slowly cools from 1 200 °C (2 194 °F) or sometimes quenches from 800 °C (1 472 °F). Some CEA tests however were performed with slow cooling before quench [27, 38].
- In case of quench testing, the experimental conditions may vary even more than in RCT. The simplest quench test can be performed with empty cladding tubes or fuel rods with open ends and in this case very high degree of oxidation must be reached to observe fracture. The quenching from one vs. two side(s) as well as presence or no-presence of the (dummy) pellets may have effects on cooling and quench characteristic. The simulation of ballooning before quench results in secondary hydriding and the material gets brittle at an even lower degree of oxidation. Quench tests can be carried out with or without additional load (or constraint) and of course the high constraint leads to fuel fracture at a lower degree of oxidation.
- The schematic comparison of the listed effects is illustrated in Figure 2.

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Figure 1. CEA and ANL temperature histories in oxidation for RCT test

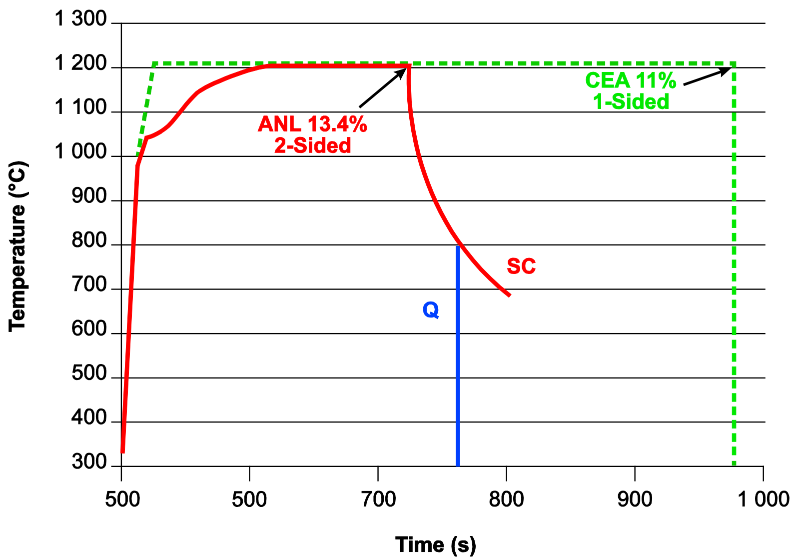
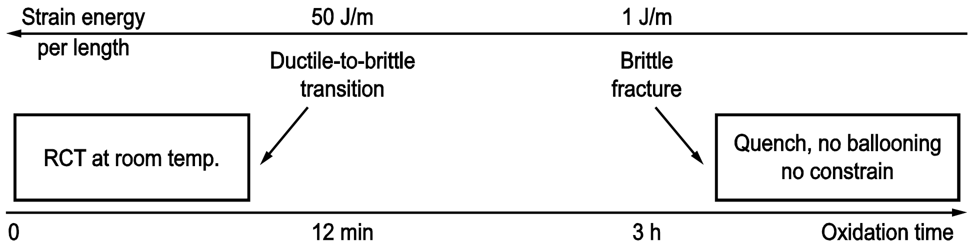


Figure 2. Comparison of test parameters

Pre-hydriding	is more severe than	As-fabricated material
Mechanical testing at room temperature		Mechanical testing at 135°C
Testing ballooned region		Testing undeformed cladding
Testing with load		Testing without load

A recent study [34] tried to estimate how far the ductile-to-brittle transition and conditions of fuel fracture during quench are from each other. According to experimental results the strain energy per unit length for the ductile-to-brittle transition is about 50 J/m and the brittle failure corresponding to water quench would take place below 1 J/m. It was pointed out that the ductile-to-brittle transition for Zircaloy-4 can be reached in 12 minutes at 1 000 °C under isothermal conditions, while fracture during integral testing without constraint is associated with oxidation for three hours at the same temperature (Figure 3). So the difference is very large if secondary hydriding does not take place.

Figure 3. Oxidation time of Zicaloy-4 cladding at 1 000 °C to reach ductile-to-brittle transition and brittle fracture during quench without constraint



As described in Sections 3.1 and 3.2.1 the behaviour of high burn-up cladding was rather different in the Japanese integral and the United States RCT tests. The hydrogen content in the high burn-up cladding used in both test series was about 700-800 ppm, which may result in reduction of Zr cladding ductility.

In case of RCT, the embrittlement threshold for the high burn-up cladding is reached after a short oxidation time compared to as-received cladding and the limiting ECR drops to a low value. In case of integral tests the pre-hydrided cladding must reach much higher degree of embrittlement in order to reach fracture. The contribution of pre-hydriding to the final brittle state of the cladding appears to be smaller than that in RCT, and the main effects are related to oxidation, ballooning and secondary hydriding. So in the integral tests the pre-hydrided cladding gets the necessary embrittlement level at an ECR value slightly lower than that of as received cladding.

As mentioned above, the two methods give information on different phenomena at different degrees of embrittlement and adopted different experimental conditions. However, it can be generally said that the cladding embrittlement is more sensitive to pre-hydriding in RCT.

5.2. Bending test or RCT

Compressing a ring induces hoop-bending stresses that change from tensile to compressive across the wall of the cladding. Other testing techniques used to determine the transition from ductile-to-brittle behaviour include loading modes that are similar (e.g., axial bending) or more severe (e.g., axial or hoop tension). Both France and Russia have found that ductility and embrittlement results determined from axial bending tests were comparable to those determined from ring compression tests [24]. As such, it appears that if oxidised cladding embrittles in the hoop direction, it will also embrittle in the axial direction. This similarity is expected because embrittlement in the prior-beta phase is independent of loading direction.

Four-point bending test of ballooned fuel cladding could combine the main advantages of RCT and semi-integral tests, since it can include the simulation of Zr embrittlement due to both oxidation and secondary hydriding (similarly to semi-integral test) and it can indicate the loss of ductility in the weakest section of the fuel rod (similar to RCT).

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6. Recommendation

6.1. Quench test

By conducting quench tests without mechanical loading, the thermal shock effect on the cladding can be investigated with different levels of hydrogen content. If the threshold is based on the first leak detected, it gives some margins with respect to the fracture or fragmentation threshold. This test is the first approach to evaluate the quench-bearing capability of the cladding and addresses only thermal stress.

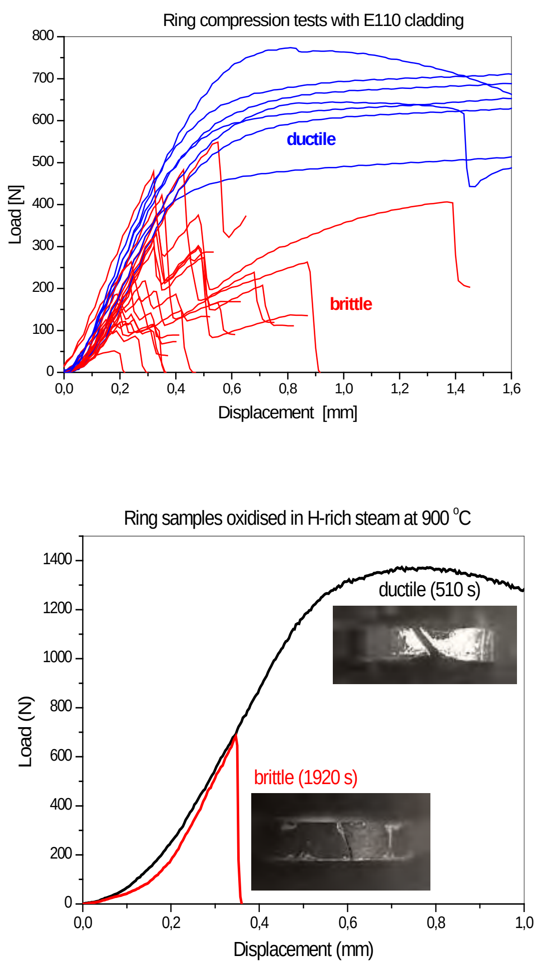
The semi-integral quench tests under constraint provide information on the structural behaviour of the fuel rod under simulated LOCA conditions and strength of the fuel rod allowing the occurrence of the fracture, if any, at the weakest point (for example, located at the burst location or at the neck regions in JAEA test where the secondary hydriding leads to high hydrogen concentrations). This kind of test simulates the LOCA sequence up to quench phase. A fracture/no-fracture criterion can be directly derived by incrementing the high temperature oxidation time. Additional sequence (loading) might be necessary to assess the post-quench cladding integrity. The investigations should be continued regarding the "pellet-cladding bonding" and "plant vibration" effects. More investigations are needed to define an appropriate loading (or a consensus based conventional loading) to be applied to the fuel rod during or after the quench phase when defining the test protocol that will be used to determine the design LOCA limits.

6.2. Ring compression test

First the test has advantages like requirement of smaller specimen volume, easier specimen preparation and easier specimen set-up in the test apparatus. At the same time, it may be considered as a "local material property" test, since it addresses the behaviour of a small length of cladding, without fuel. The stress applied during the test is also localised and may be higher than that generated during quenching. The aim of this mechanical test is to produce a limit such that the cladding remains ductile, and doing so, to avoid discussions about the loads that the cladding must sustain during or after a LOCA.

Among ring compression tests conducted in different laboratories, it is important to have a common definition of ductile-to-brittle transition. In addition, more data and discussion are required about test procedures and conditions, such as oxidation temperature profile, one-side or two-side oxidation, test temperature (room temperature or 135° C), sampling position from oxidised/ruptured cladding and cooling scenario ("with quench or without quench"), in order to obtain a clear consensual criterion for cladding embrittlement.

Figure 4. Load displacement curves showing the ductile-to-brittle transition in RCT (top) [39] and ring tensile (bottom) [40] tests



Zero ductility (or ductile-to-brittle transition or embrittlement threshold) could be determined on the basis of load-displacement curves. The curve for a ductile material is characterised by a ductile plateau after elastic section and when this plateau is missing the material has some degree of embrittlement (Figure 4). The load displacement curves of compression, tensile and bending tests have the same character, and any of them can be used. The zero ductility can be evaluated by the

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shape of the curve without need to introduce specific parameters as e.g. offset strains or strain energy which are derived from the curves.

6.3. General opinion

Two basic types of tests have been proposed and are in use in LOCA research. A mechanical test based on ductility measurements (e.g., ring compression, three- or four-point bend, and impact tests) and a test based on resistance to thermal shock during quenching of a heated fuel rod (with or without mechanical loading).

The integral-type experiments with Zr cladding and fuel rods pointed out that the weakest part of the cladding during a LOCA would be the ballooned section, especially the “neck” region where secondary hydriding takes place. The traditionally used ring compression test refers to the Zr cladding behaviour outside the ballooned region, while the semi-integral quench tests with short fuel rods can simulate not only ballooning but also secondary hydriding.

In the mechanical tests, it is important to note, despite the fact that the methods used to measure ductility are different, the conclusions reached in different laboratories on the same cladding materials do not substantially differ from each other. However, experience has shown that the results of ductility tests are very sensitive both to the way the test samples are prepared and the way the tests are performed. The preparation of samples includes the selection of their geometry, surface treatment, pretest oxidation or corrosion, and pre-hydriding.

At the same time, parameters such as oxidation time and temperature and heating as well as cooling rate have a significant impact on the final ductility of tested samples. Moreover, the techniques involved in mechanical testing of oxidised cladding and processing and presentation of results represent an additional source of potential discrepancy between different laboratories. Thus, for an appropriate understanding, interpretation, and/or a comparison of results from different laboratories, it is important that these parameters be known and unified to the extent possible.

Moreover, it has been noted that the test parameters (e.g., quench or slow cool from oxidation temperature) can have an impact on test results as significant as the test method.

The main objective of LOCA criteria is to prevent fuel rod fracture which may affect coolable geometry of the core. Such fuel rod fracture conditions can be derived from experiments using any one of several test methods, and the results tempered by choice of test parameters, depending on the degree of conservatism desired.

7. Summary

This technical opinion paper reviews the loss-of-coolant accident (LOCA) criteria basis and the different test methodologies, and provides recommendations to the international community on how the results of these different methodologies can be applied for regulatory purposes.

The quench test without mechanical loading is the first approach for evaluation of the quench-bearing capability of the cladding and addresses only thermal stress. The semi-integral tests under constraint provide information on the structural behaviour of the fuel rod under simulated LOCA conditions and strength of the fuel rod allowing the occurrence of the fracture, if any, at the weakest point. This kind of test simulates the LOCA sequence up to quench phase. A fracture/no-fracture criterion can be directly derived by incrementing the high temperature oxidation time. Additional sequences (e.g. loading) might be necessary to assess the post-quench cladding integrity. The investigations should be continued regarding the "pellet-cladding bonding" and "plant vibration" effects. More investigations are needed to define an appropriate loading to be applied to the fuel rod during or after the quench phase when defining the test protocol that will be used to determine the design LOCA limits.

The ring compression test (RCT) has advantages of requiring smaller specimen volume, easier specimen preparation and easier specimen set-up in the test apparatus. At the same time, it may be considered as a "local material property" test, since it addresses the behaviour of a small length of cladding, without fuel. The stress applied during the test is also localised and may be higher than that generated during quenching. The aim of this mechanical test is to produce a limit such that the cladding remains ductile, and in doing so, to avoid discussions about the loads that the cladding must sustain during or after a LOCA. Among ring compression tests conducted in different laboratories, it is important to have a common definition of ductile-to-brittle transition. In addition, more data and discussion are required about test procedures and conditions, such as oxidation temperature, test temperature, sampling position from oxidised/ruptured cladding and cooling scenario, in order to obtain a clear consensual criterion for cladding embrittlement.

Since specimens for RCT are generally taken from non-ballooned region, they may not fully represent the behaviour of the cladding in the balloon area nor fully represent other phenomena (e.g., secondary hydriding). To overcome this problem, the bending test can be used. In this sense, the bending test is simply considered another method of observing the mechanical response of the cladding, but now includes the ballooned region. There is only limited experience in the use of the bending test for the investigation of cladding ductility in the ballooned section compared to RCT or semi-integral tests (and, bending test as RCT, does not represent any particular loading conditions that can be expected in LOCA transient). It would be important to carry out bending tests in different laboratories in order to analyse the applicability of this type of tests for the establishment of LOCA criteria.

When comparing the results of RCT and semi-integral testing the following must be taken into account:

- The two methods consider different mechanisms in order to evaluate the embrittlement process of Zr cladding in high temperature steam.
- The two methods give information on somewhat different phenomena at different degrees of embrittlement.

Furthermore the RCT and semi-integral tests carried out in different laboratories or in different test series are not conducted in the same way and it can lead to rather different results.

The present review introduces and compares the advantages and drawbacks of the various test methodologies, but does not specify any standardised procedure for the experimental conditions to establish LOCA criteria because of several reasons:

- Each experimental set-up and procedure has special advantages and limitations. Test conditions and methods of data evaluation vary in different laboratories involved in testing.
- Data produced in different laboratories and facilities on the embrittlement of Zr cladding under similar conditions are generally comparable to each other. Consequently, the differences in embrittlement test methodologies do not seem to be of primary importance. However, the impact of test conditions, e.g. slow-cooling versus quenching, needs to be addressed while drawing conclusions for LOCA criteria.
- The establishment of LOCA criteria includes conservative assumptions which are not the same in all countries and which need specific interpretation of experimental data.

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List of abbreviations

AEKI	*Atomenergia Kutatóintézet*/Atomic Energy Research Institute (Hungary)
ANL	Argonne National Laboratory
CEA	*Commissariat à l'energie atomique*
CSN	*Consejo de Seguridad Nuclear* (Spain)
CSNI	Committee on the Safety of Nuclear Installations
DBA	design basis accident
ECCS	emergency core cooling system
ECR	equivalent cladding reacted
EDF	*Électricité de France*
ENSI	Swiss Federal Nuclear Safety Inspectorate
GRS	*Gesellschaft für Anlagen und Reaktorsicherheit*
IRSN	*Institut de radioprotection et de sûreté nucléaire*
JAEA	Japan Atomic Energy Agency
LOCA	loss-of-coolant accident
LWR	light water reactor
NEA	Nuclear Energy Agency
NRI	Nuclear Research Institute (Czech Republic)
OECD	Organisation for Economic Co-operation and Development
PCT	peak cladding temperature
PSI	*Paul Scherrer Institut* (Switzerland)
RCT	ring compression test
RIA	reactivity initiated accident
TOP	technical opinion paper
USAEC	United States Atomic Energy Commission
USNRC	United States Nuclear Regulatory Commission
VTT	*Valtion Teknillinen Tutkimuskeskus*/Technical Research Centre of Finland
WGFS	Working Group on Fuel Safety

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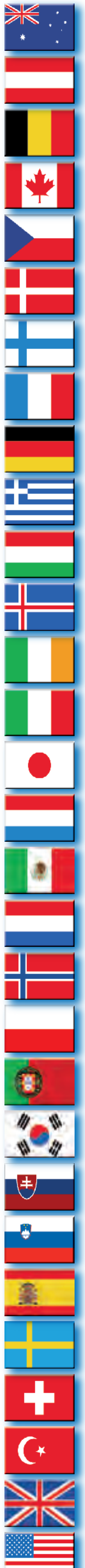

CSNI Technical Opinion Papers No. 13

Acceptance criteria for emergency core cooling systems (ECCS) define the maximum temperature and degree of oxidation in order to avoid excessive embrittlement and hence failure of the fuel cladding, which would affect core cooling in the case of a loss-of-coolant accident (LOCA). The criteria are mainly based on experimental data obtained in the 1970s-80s. Several types of tests have been performed to evaluate structural integrity and embrittlement of the cladding under LOCA conditions, and consequently different test methodologies have been used for determining the cladding embrittlement criteria. The current trend towards high burn-up and the use of new cladding alloys has increased the need for international discussions on these test methodologies and acceptance criteria. In response, the NEA Committee on the Safety of Nuclear Installations (CSNI) and its Working Group on Fuel Safety produced this technical opinion paper, which should be of particular interest to nuclear safety regulators, nuclear power plant operators and fuel researchers.

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