

Assessment of Delayed Hydride Cracking in Used CANDU Fuel Bundles during Dry Storage

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ABSTRACT

Title: Assessment of Delayed Hydride Cracking in Used CANDU Fuel Bundles during Dry Storage
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Abstract

This report summarizes various experimental and modelling activities performed to evaluate if Delayed Hydride Cracking (DHC) could be occurring in used CANDU fuel stored in Ontario Power Generation Dry Storage Containers. The conclusion of this work is that DHC is not expected in the used CANDU fuel.

DHC requires a hydrogen concentration in a material that exceeds its solubility, a stress gradient that concentrates the dissolved hydrogen thus forming solid hydrides, and a sufficient stress to fracture the formed hydrides. If all conditions are present, DHC is expected to occur. Early studies identified that the hydrogen concentration in used fuel was sufficient for DHC, but it was uncertain whether the magnitude of the stress in CANDU fuel would be sufficient for DHC under dry storage conditions.

To investigate this further, AMEC NSS developed finite element stress models of CANDU fuel bundles used by Ontario Power Generation and Bruce Power. The stress models were developed so they could account for the condition of used CANDU fuel. A key area of the fuel bundle that is a concern for DHC is the endplate-to-endcap weld where there is a very sharp weld discontinuity. A case study of the fuel in dry storage conditions was performed to determine the stress intensity factors at the tip of the weld discontinuity. The results suggest stress intensity factors no larger than $3 \text{ MPa m}^{1/2}$.

In understanding if this stress intensity factor is sufficient to initiate DHC, Kinectrics developed a methodology and apparatus to determine the critical stress intensity factor for initiation of DHC at the endplate-to-endcap welds. Both 28- and 37-element fuel designs from different manufacturers were tested. The results of the testing suggest the critical stress intensity factor is in the range of $7.6 \text{ MPa m}^{1/2}$ to $13.6 \text{ MPa m}^{1/2}$.

Since the calculated stress intensity factor is well below the critical stress intensity factor, DHC is not expected to occur during dry storage. Though there are some uncertainties remaining in this work, they are not considered to be significant enough to affect the conclusion that DHC is not expected during dry storage, given the indicated margin between the applied and critical stress intensity factors.

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1. INTRODUCTION

This technical report summarizes the results of analysis to evaluate if Delayed Hydride Cracking (DHC) could be occurring in used CANDU fuel bundles during storage in a Dry Storage Container (DSC). This work is part of the Used Fuel Integrity (UFI) program at the Nuclear Waste Management Organization (NWMO), which was initiated to provide assurance that used CANDU fuel that has been stored in a dry environment for up to 100 years can be safely and efficiently handled during post-storage operations.

Past investigations into the condition of CANDU fuel during dry storage have provided confidence that bundles do not significantly degrade and their integrity is maintained under normal storage conditions (Lovasic and Gierszewski, 2006; Lampman and Daniels, 2005). However, more recent work in the UFI program indicated that fuel bundles in dry storage might be susceptible to DHC, particularly in the endcap-to-endplate welds (commonly termed the *assembly welds*). As a result, a strategy to investigate this degradation mechanism was developed in order to assess whether DHC could occur during dry storage (Lampman and Daniels, 2005).

Parametric ANSYS Finite Element (FE) models of 28- and 37-element CANDU fuel bundles were developed to calculate the stresses within used fuel bundles during dry storage (Lampman and Popescu, 2008; Popescu and Lampman, 2010; Lampman and Popescu, 2010). Several experiments were performed with unirradiated fuel bundles to validate the models. A sensitivity study of the model was performed to determine the parameters that have the greatest effect on the bundle response and determine the best-fit parameters to be used in the finite element models. Comparison of the unirradiated fuel bundle models with the experimental data shows good agreement for axial and bending loads.

The finite element fuel bundle models were updated to account for the effect of irradiation on material properties. Calculations of the stress distribution were performed for different post-discharge bundle geometries modelled under initial dry storage conditions. Calculation of the stress intensity factor at the weld discontinuities of the assembly welds were performed for comparison with the critical stress intensity factors required for delayed hydride cracking.

In parallel with the development of the finite element fuel bundle stress model, Kinectrics developed a test apparatus and test methodology to determine the critical stress intensity factor required to initiate DHC and the delayed hydride crack velocity (DHCV). Testing was performed on both 28- and 37-element non-irradiated fuel bundles from both General Electric – Hitachi (28- and 37-element) and Cameco Fuel Manufacturing (37-element) to investigate different bundle designs and manufacturing.

An earlier study performed by AMEC NSS within the UFI program reviewed the condition of all discharged fuel from Ontario Power Generation (OPG) and Bruce Power (BP) reactor cores (Lazaroski *et al.*, 2005). This work identified two basic "populations" of used fuel discharged from OPG and BP cores: the segregated population consisting of approximately 0.2% of all discharged fuel and the remaining bulk population. The segregated population generally consists of bundles which are defected or damaged and will be stored separately from the bulk population. The focus of the work described in this document is to examine the potential for DHC in the much larger bulk population of fuel that will be stored in a DSC.

2. BACKGROUND

The average CANDU fuel bundle is discharged from the reactor with a burnup of approximately 210 MWh/kgU after a residence time of up to one to two years. The coolant temperature in the core is between 260°C and 300°C and the external pressure applied to the bundle's sheath is about 10 MPa. Recently, analysis of the range of wear and deformation of all the fuel bundles discharged from Pickering, Darlington, and Bruce stations was performed for the period from 1972 to 2004 (Lazaroski *et al.*, 2005). This report indicates that the majority of the approximately 1.5 million fuel bundles discharged in this period has nominal wear and deformation, but that there is a small proportion that may have some significant wear and deformation. This report summarizes work performed to evaluate if the presence of DHC in the discharged fuel bundles is possible during storage in an Ontario Power Generation DSC.

2.1 Storage of Used CANDU Fuel in Dry Storage Containers

When a fuel bundle is discharged from a reactor, it undergoes a brief temperature excursion of approximately three minutes up to a temperature typically less than 400°C before placement in the Irradiated Fuel Bay (IFB). The fuel bundles are then cooled by the surrounding water in the IFB at a temperature of approximately 25°C for a minimum period of 10 years. There is no evidence of fuel degradation during this period.

When the fuel bundles are removed from the IFB at OPG and Bruce Power nuclear power plants, they are loaded into fuel storage modules (with the exception of Darlington NGS where the bundles are immediately stored in the modules after discharge from the reactors), which are then loaded into the DSCs. The structure and contents of a typical DSC is shown in Figure 1.

A fuel storage module consists of an array of 48 horizontal tubes, each of which accommodates two used fuel bundles. Each DSC is loaded with four modules in the fuel bay. The DSC is then removed from the IFB, drained, and vacuum dried. This process results in another temperature excursion of the used fuel, but the fuel is not expected to reach temperatures greater than 150°C (Lovasic and Gierszewski, 2006) and will gradually decrease with storage time.

Used fuel may be stored in DSCs for extended periods as great as 100 years before it would be transferred to the next phase of waste management. An evaluation of different degradation mechanisms that may be present in used CANDU fuel during this period has been performed by Lampman and Daniels (2005). The storage environment and conditions are generally considered to be benign and do not promote degradation of the fuel. However, uncertainties in the understanding of the stress levels in fuel bundles stored horizontally were identified. Specifically, it was uncertain at the time whether the bending stress at the assembly welds of used CANDU fuel bundles would be sufficient to initiate delayed hydride cracking.

2.2 Delayed Hydride Cracking

DHC is a slow, hydrogen-assisted cracking mechanism that is essentially driven by hydride formation and fracture in high stress fields. Hydrogen and deuterium that is in solution in the Zircaloy-4 matrix can diffuse to high stress locations. As it reaches the high stress area, the hydrogen precipitates out of solution if the local concentration is greater than the solubility of the Zircaloy-4 matrix. This results in hydride platelets that are oriented perpendicular to the stress gradient at the crack tip. As more hydrogen migrates to this region, the hydride grows until it becomes unstable due to its size in the matrix and fractures, resulting in the crack advancing.

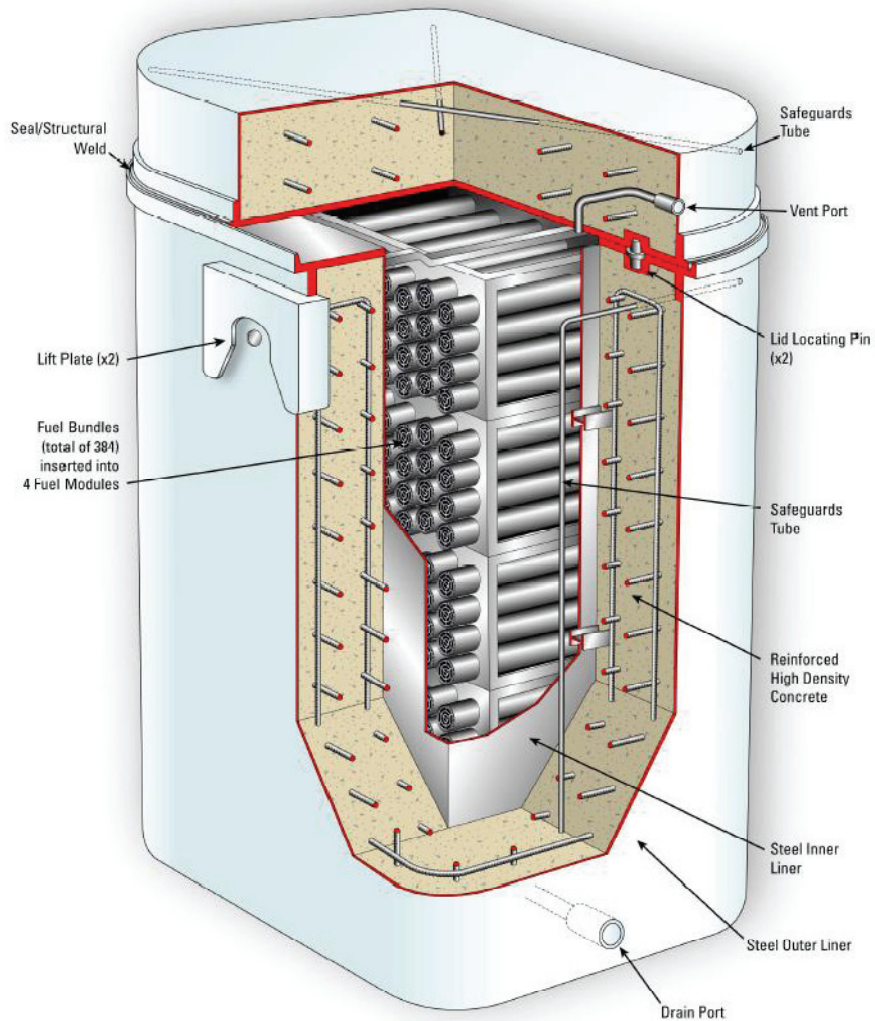


Figure 1: Design of the Canada Power Generation Dry Storage Container

This process repeats itself until either the crack has progressed through the material or the stress is relaxed. Rashid *et al.* (2000) have identified five basic conditions that must be met for DHC to occur in used fuel cladding. Some of the conditions are specific to used fuel cladding considerations, but they can be generalized to two basic conditions for DHC to occur at the assembly welds:

- Sufficient hydrogen and deuterium must be present in the material so that hydrides will form. Therefore, the hydrogen concentration must exceed the solubility limit at the material's temperature.
- A sufficient stress must be present to drive hydrogen migration to the crack tip and fracture the subsequent hydride.

The first condition for DHC to occur appears to be satisfied in discharged CANDU fuel bundles. During operating conditions, fuel bundles absorb deuterium due to oxidation of the Zircaloy-4 by the coolant. Post-Irradiation Examination measurements of hydrogen and deuterium concentrations indicate that the average equivalent hydrogen concentration in the bundle

ranges from 45 ppm to 85 ppm by weight. The results of McMinn *et al.* (2000) indicate that the maximum solubility of irradiated Zircaloy-4 at 150°C is approximately 20 ppm by weight. Therefore, the concentration of hydrogen and deuterium in used CANDU bundles is high enough that, given sufficient stress, DHC can occur at the start of storage in a DSC.

The second condition is the presence of a high enough stress field to drive diffusion and fracture of the hydrides. Reviews of available experimental work did not identify information on the critical stress intensities needed for DHC of irradiated Zircaloy-4 at temperatures below 150°C. However, previous assessments of delayed hydride cracking (DHC) have been made for used fuel cladding by Ikeda (2002) and Rashid *et al.* (2000). These assessments are based on information about DHC at normal operating conditions in Zr-2.5%Nb and there is little evidence to predict the behaviour of DHC at dry storage conditions. For the Rashid *et al.* report, the focus was on LWR and BWR fuel where degradation of the cladding was examined. The report indicates that DHC will not occur in the cladding due to low stress levels. This is expected to be true for CANDU fuel element sheaths as well. However, CANDU bundles contain assembly weld regions that provide a greater stress enhancement than cladding and thus make the welds more susceptible to DHC.

The presence of DHC during dry storage is a critical concern. If it occurs, then loss of fuel integrity is expected which would have an impact on bundle load and vibration limits. However, it was unclear whether this would occur during dry storage. This document discusses an investigation to examine if DHC is possible at the start of dry storage. To do this, the following parameters were evaluated for a storage temperature of approximately 150°C:

- the critical stress intensity factor required for DHC in assembly welds , and
- the stress intensity factors in used CANDU fuel assembly welds during storage in a DSC.

This report summarizes the work done to establish these parameters and evaluates the possibility of DHC occurring during dry storage.

3. FINITE ELEMENT APPROACH TO MODELLING CANDU FUEL BUNDLES

To understand if DHC can occur within the used fuel bundle, an understanding of the stress levels within the bundle, and specifically, the stress intensity factor (K_I) at the location of crack initiation, is required. To calculate K_I values in assembly welds of dry-stored used CANDU fuel bundles, AMEC NSS has developed finite element fuel bundle models and performed calculations of K_I values of used fuel bundles in fuel storage module tubes. This work has been documented in detail in several reports (Lampman and Popescu, 2008; Popescu and Lampman, 2010; Lampman and Popescu, 2010), and this report only summarizes key developments and findings.

3.1 Description of Finite-Element CANDU Fuel Bundle Models

Full finite element models of the 28- and 37-element CANDU fuel bundles were created using the ANSYS finite element software package. Parametric models were developed to allow for variations in the fuel bundles' geometry, composition, and material properties. This allowed for modelling of used fuel geometries, variations in the modelled components of the fuel bundle, such as the fuel pellets, and modelling of irradiated and unirradiated Zircaloy-4 material properties.

The overall solid model of the 28-element fuel bundle is shown in Figure 2 and one corresponding finite-element model is shown in Figure 3. The models contain all the components of the fuel bundle, including spacer pads to model contact between adjacent fuel elements. Many different types of finite elements are used in the model to optimize its size and convergence time. Additionally, to minimize the size of the model and solution times, linear finite elements (*i.e.*, no mid-size nodes) were used throughout. The size of the model depends on the parameter values, but for most analyses the 28-element model contained between 300,000 to 500,000 degrees of freedom.

Details of the finite-element model are given in the development and sensitivity study reports (Lampman and Popescu, 2008; Popescu and Lampman, 2010). However, several aspects of the model are notable in discussing the final analysis results:

- The fuel element models could be varied on an element-by-element basis. Therefore, fuel elements which contained modelled UO_2 pellets could be inserted into the model where needed, while other elements would not contain modelled pellets. This allowed for the maximum modelling options to reduce the model size while incorporating the appropriate details where needed, such as for an element where a load is directly applied. This also allows for the development of new fuel element models, which can be easily incorporated into other analyses, as needed.
- Fuel element models allow for initial, pre-load bowing of the element. This allows for the modelling of used fuel geometries, as shown in Figure 4, prior to loading into the module tube.
- The effect of irradiation and temperature on the Zircaloy-4 material can be captured by changing the elastic modulus in the model. In this analysis, unirradiated and irradiated material properties were taken from MATPRO (MATPRO, 2003).

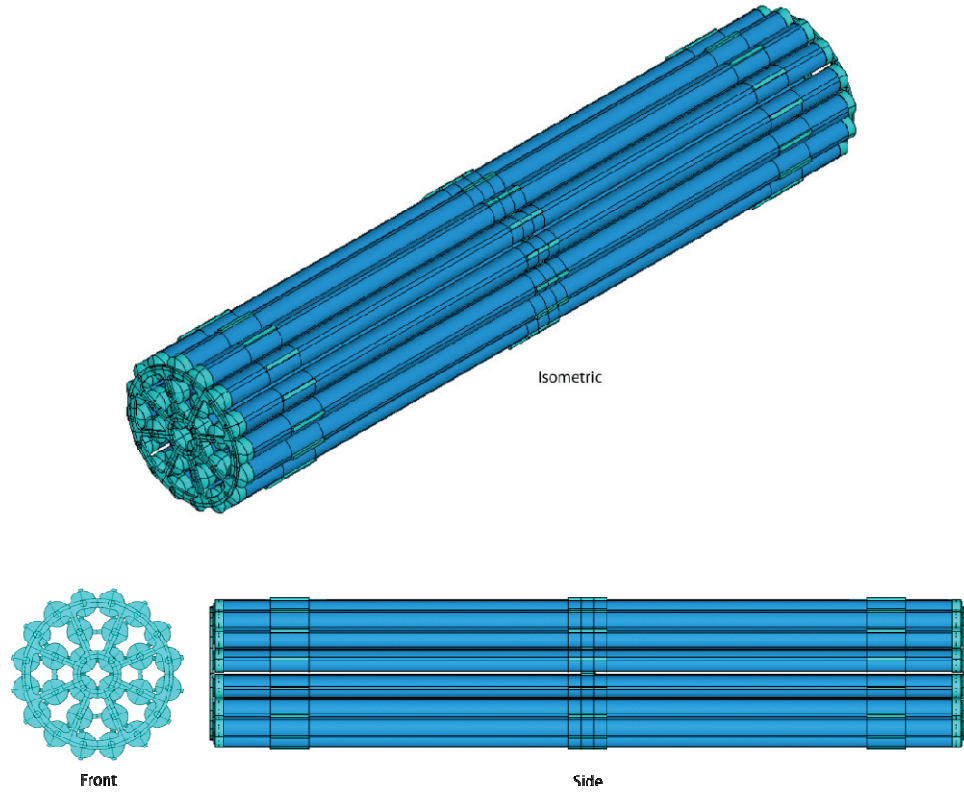


Figure 2: 28-Element Fuel Bundle Solid Model

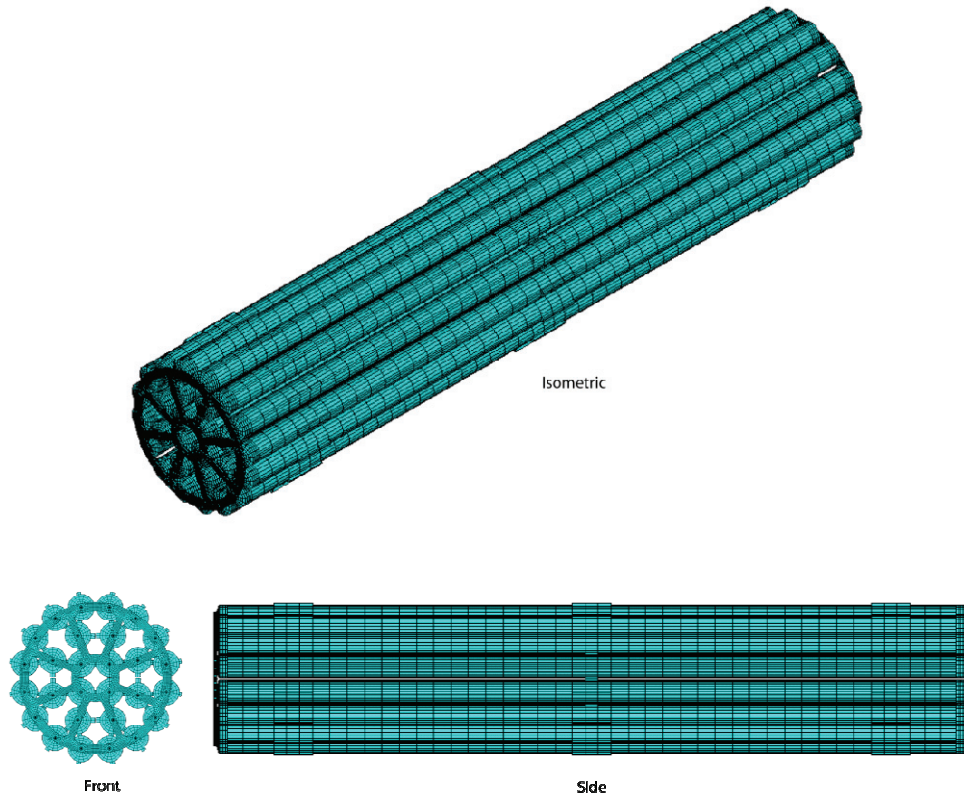


Figure 3: 28-Element Fuel Bundle Finite Element Model

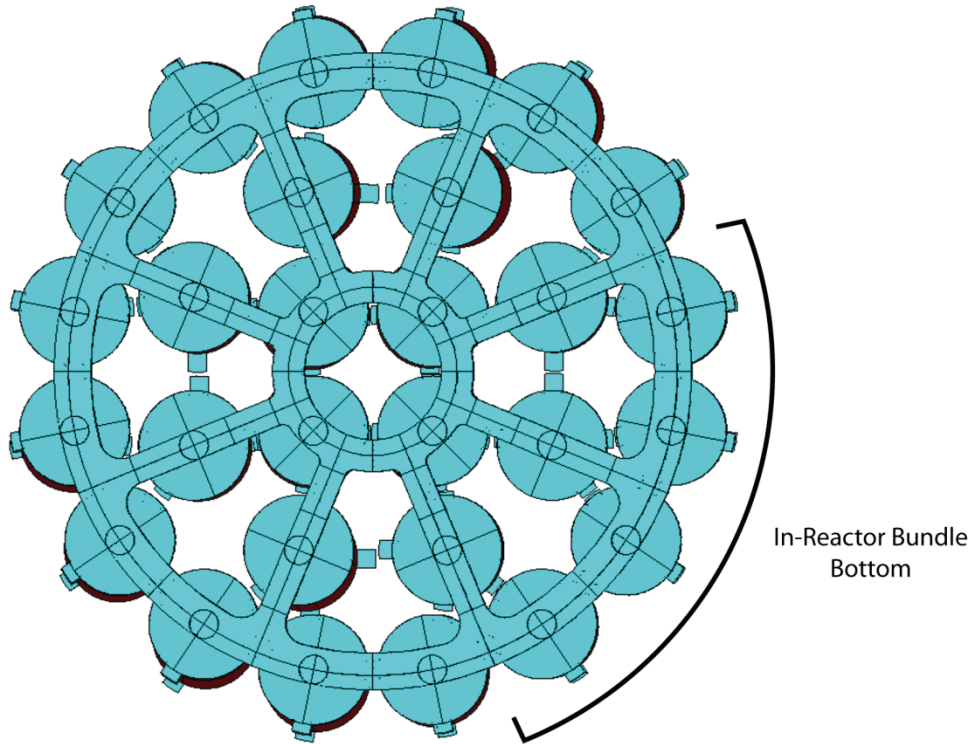


Figure 4: Post-Discharge Geometry of a Used Fuel Bundle Showing Orientation in the Reactor Core

Experimental data on fuel element deformation due to applied mechanical loads was collected by Stern Laboratories to validate the fuel element models. Single elements were removed from unirradiated bundles, the endplates on both sides were clamped, and a load was applied at the center of the element perpendicular to its axis. Measurements of applied loads and resultant deformation were recorded. A sensitivity study was performed with the fuel bundle model for comparison against the experimental data. The results of this work for the 28-element fuel model are shown in Figure 5 where the sensitivity cases are seen along with the best estimate case. The parameter values used for the best estimate were reviewed and are within manufacturing allowances and agree well with known measurements of individual parameters. Therefore, the best estimate sensitivity case was used for this analysis.

The bundle model allows for the calculation of deformation of fuel bundles in dry storage. However, it does not calculate K_I values for the specific assembly weld geometries. To do this, a submodel of the assembly weld including the weld discontinuity, as shown in Figure 6, was developed and validated. The calculated deformation around the assembly weld from the bundle model was mapped onto the assembly weld submodel and K_I values around the weld discontinuity were calculated.

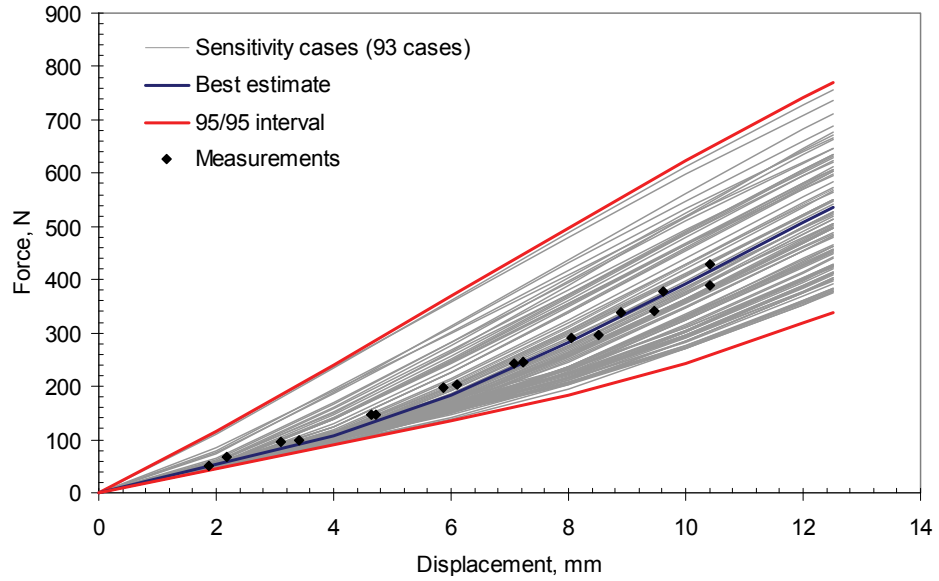


Figure 5: Sensitivity Study Results for 28-Element Fuel Bundle Model

3.2 Evaluation of K_I in Used Fuel Bundles

The details of the case study to evaluate stress levels in dry-stored CANDU fuel and the associated K_I values in the assembly welds is documented in Lampman and Popescu (2010). The analysis focussed on an evaluation of the 28-element fuel bundle model.

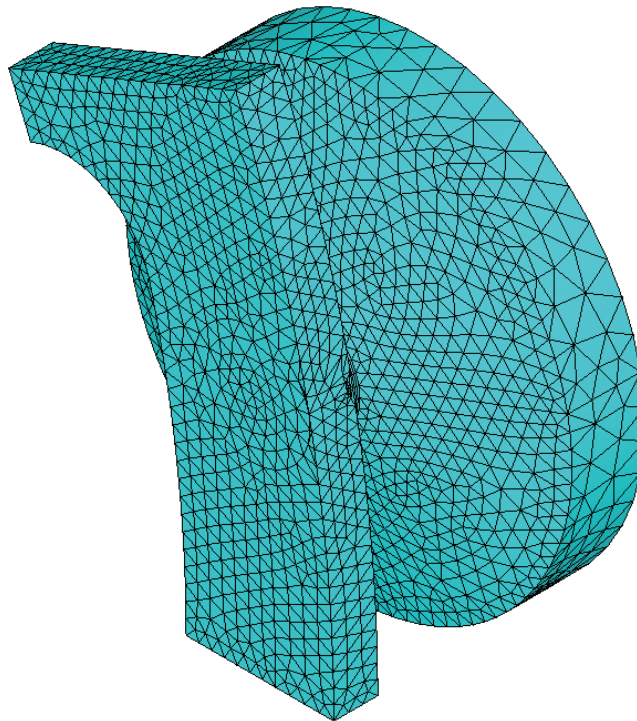


Figure 6: 28-Element Assembly Weld Finite Element Submodel

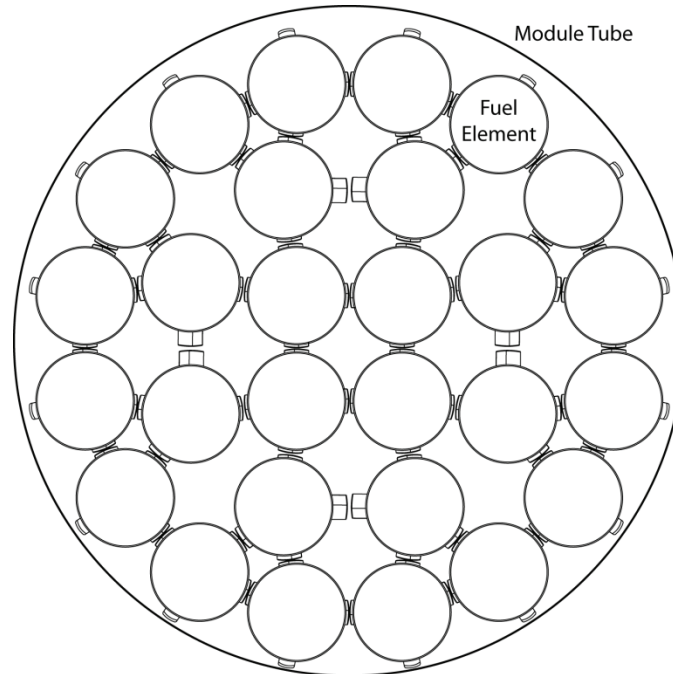


Figure 7: Geometry of a Nominal Bundle in a Module Tube

The irradiated 28-element fuel bundle model was used to calculate the stress distributions and K_I values for several dry storage scenarios. For all calculations, the hollow fuel element model, which ignores stiffening effects of fuel pellets, was used to give conservative results¹. The following analysis cases were evaluated:

- Irradiated fuel bundle in free space with a typical post-discharge geometry (an example is given in Figure 4).
- Irradiated fuel bundle in a module tube with an as-manufactured geometry.
- Irradiated fuel bundle in a module tube with a post-discharge geometry.
- Irradiated fuel bundle in a module tube with a limiting element bow due to crept fuel channel.

The geometry of a fuel bundle in a module tube is shown in Figure 7. It has been assumed in this analysis that the orientation of the fuel bundle in the module tube is identical to the orientation of the bundle in the fuel channel.

For all analyses, the stress distribution in the endplate was plotted (an example is given in Figure 8) to determine at which assembly weld the axial stress was the greatest. The welds with the greatest axial tensile stress were identified. From this information, several assembly

¹ The material properties and geometry of irradiated fuel pellets is variable and difficult to model. During irradiation, the microstructure of the pellets varies across its radius and the pellets will crack. To account for this uncertainty, they were not modelled, thus increasing the predicted element deflections and associated K_I values at the assembly welds.

welds were chosen for further analysis to determine the maximum K_I around the assembly weld. The results of this for the four cases evaluated are given in Table 1.

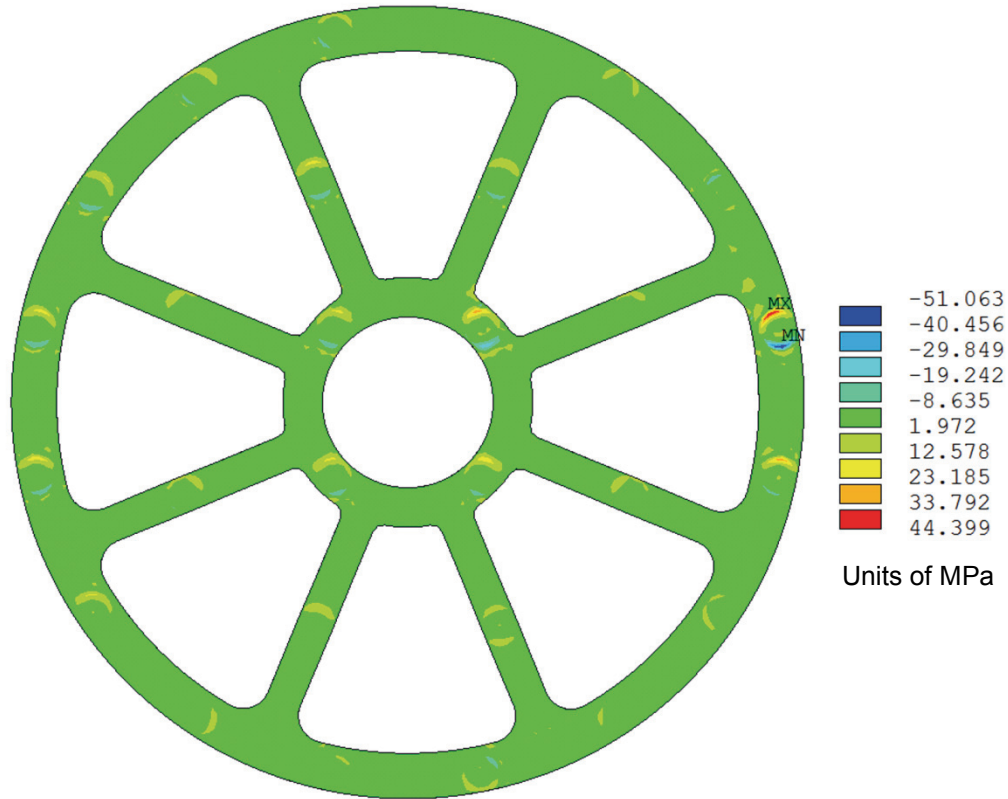


Figure 8: Axial Component of Stress Distribution in the Endplate for a Used Fuel Bundle in a Module Tube

Table 1: Summary of Calculated Stress Intensity Factors for Dry Storage Case Study

Analysis Case (28-Element Fuel Bundle)	Bundle Ring	Maximum K_I (MPa m ^{1/2})
Post-Discharge Geometry in Free Space	Outer	1.48
	Intermediate	0.83
Nominal Geometry in Module Tube	Outer	1.01
	Intermediate	0.97
Post-Discharge Geometry in Module Tube	Outer	2.45
Crept-Channel Geometry in Module Tube	Outer	1.53
	Intermediate	3.04

4. CRITICAL STRESS INTENSITY FACTOR FOR DHC

In parallel with the development of the finite element fuel bundle stress model, Kinectrics developed a test apparatus and test methodology to determine the critical stress intensity factor required to initiate DHC and measure the delayed hydride crack velocity (DHCV). Testing was performed on 28- and 37-element unirradiated fuel bundles from General Electric Hitachi (GE-H) and Cameco Fuel Manufacturing (CFM) to investigate differences in DHC initiation between different bundle designs and manufacturers.

4.1 Test Methodology

The test apparatus and procedure, as well as the results of testing of elements from different CANDU fuel bundle manufacturers, have been described in a series of reports issued by Kinectrics (Shek and Wasiluk, 2009; Shek, 2010). The test procedure and results are summarized briefly here.

All testing was performed on samples taken from unirradiated fuel bundles without fuel pellets that were acquired from GE-H (28- and 37-element bundles) and CFM (37-element bundles). The bundles were produced using standard manufacturing processes and the geometry and material properties of the assembly welds is consistent with that of fuel bundles manufactured for use in the reactor cores.

The individual test samples are prepared from the fuel bundles. To simulate hydrogen and deuterium concentrations in used fuel, the test bundle assembly welds are first hydrided to a target concentration of 40 ppm by weight using the electrolytic hydriding and thermal diffusion method (Shek and Wasiluk, 2009). This concentration was chosen to be in excess of the terminal solid solubility for hydride precipitation at the test temperature (150°C) to ensure initiation of DHC would be independent of the hydrogen concentration. The individual test samples were then removed from the fuel bundle using electric discharge machining to prepare samples as shown in Figure 9.

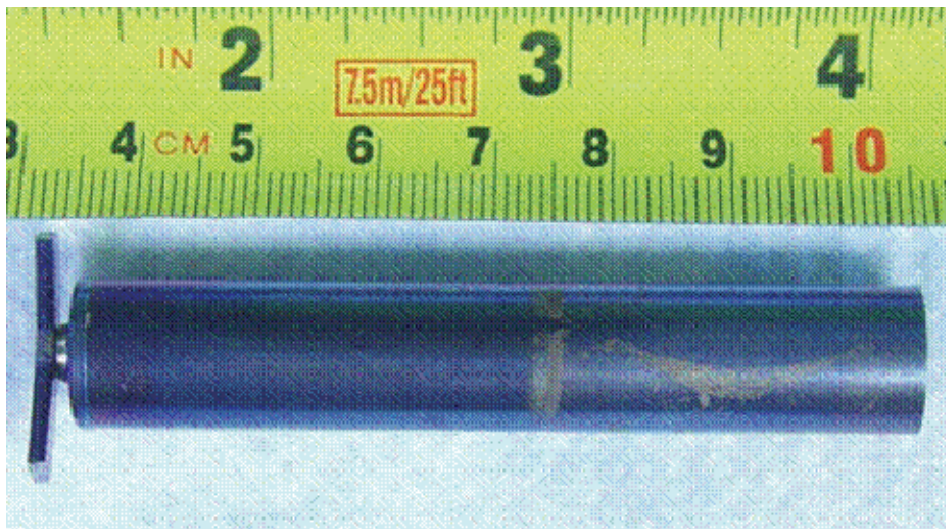


Figure 9: 37-Element Sample for K_{IH} Test (Shek, 2010)

The test apparatus is illustrated in Figure 10. The fuel element sample is secured to the loading apparatus by the portion of the endplate still attached to the sample. The sample was loaded by pulling on a cable placed around a groove in a collar attached to the fuel element. This induces a tensile bending stress in the region of the assembly weld on the opposite side of the direction of the applied load. The test sample, loading collar, and endplate clamp are all placed inside a furnace to maintain the test temperature of 150°C throughout the test. The endplate clamp can be rotated so that the direction of loading around the weld, and thus location of maximum tensile stress, can be varied.

DHC initiation is determined using an increasing load technique. In this technique, an initial applied load insufficient to initiate DHC is applied to the sample and increased in small steps at regular intervals until cracking is detected. Monitoring of crack growth is performed using the direct current potential drop technique (Shek and Wasiluk, 2009). Each applied load was held for approximately 50 hours to provide sufficient time for hydrides to form and cracking to initiate. If no cracking was detected, the load was increased by approximately 5% and held again to monitor for crack growth. Once crack growth has been detected, the applied load is noted for further analysis to calculate the critical stress intensity factor. In some test cases, the tests were extended beyond simply determining the DHC initiation load to also measure the stable DHC crack growth velocity. For the determination of DHCV, after the initiation load was determined, the applied load was increased by approximately 30% and held for 50 hours.

At several points throughout the test the sample is heated to approximately 240°C. This first occurrence of this is immediately after the first load is applied. This served two purposes: first, it preconditions the sample by ensuring the solubility of hydrogen is minimized, and secondly, it heat tints² the assembly weld discontinuity so that the initial geometry and crack depth are

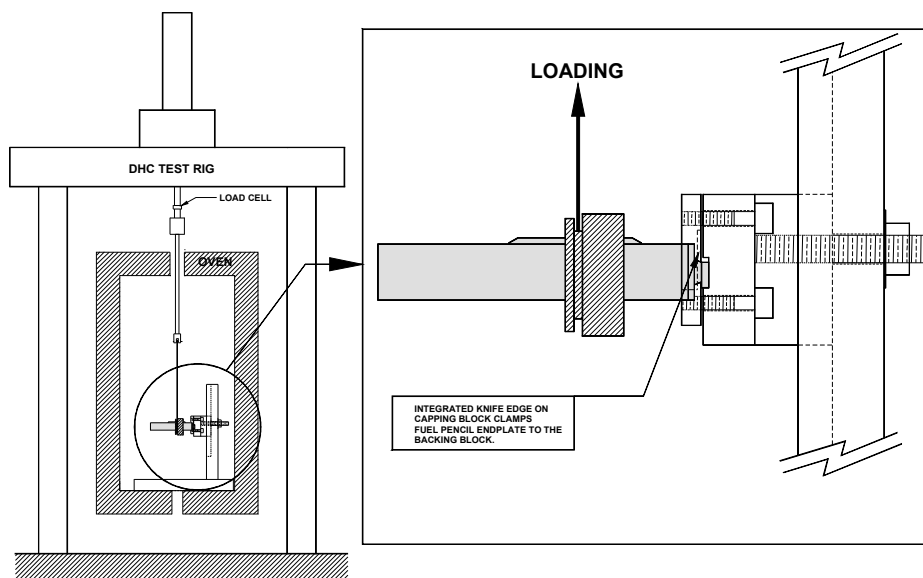


Figure 10: DHC Test Apparatus (Shek, 2010)

² The heat-tinting cycle oxidizes the exposed crack surface, thus affecting its visible hue (*i.e.*, lightness, or darkness, of the surface).

known. The second occurrence of the heat cycle is performed if a DHCV measurement is going to be made. Prior to increasing the applied load by 30%, the sample is heated to heat tint the exposed crack. Finally, at the completion of the test, another heat cycle is performed to further tint the final cracked surface.

After the loading of the sample is complete, it is removed from the apparatus and the endplate is detached from the endcap, exposing the surface of the weld discontinuity and DHC crack. An example of this fracture surface is shown in Figure 11. From the heat-tinting cycles, the initial depth of the weld discontinuity, the DHC crack growth from the initiation test, and the DHCV crack growth can be determined. Additionally, in most cases, it is clear where DHC growth first occurred: either on the top of the sample, or to the side.

A finite element model of the sample is then used to determine K_{IH} from each test. The applied load measured when crack growth first occurs and the measured depth of the weld discontinuity from the heat tinting boundaries are input into the model and K_{IH} is calculated (Shek, 2010; Wasiluk, 2010). For the DHCV values, they are determined from the measured crack growth during the DHCV test and the elapsed time for this test.

4.2 Test Results

The tests performed, and some key results, are summarized in Table 2 and Table 3. In total, there were two series of tests performed: initial commissioning tests with a GE-H 37-element bundle (Table 2); and the K_{IH} study tests to primarily determine K_{IH} under dry storage conditions and evaluate differences of fuel bundle design and manufacturer on DHC initiation (Table 3), which included a set of GE-H 37-element tests, a set of GE-H 28-element tests, and a set of CFM CANDU-6, 37-element tests. For all tests, elements from the outer ring of the bundle were used. In the K_{IH} study GE-H tests (Table 3), DHC was initiated on the inner side of the element facing the interior of the bundle (ID loading location) as well as the outer side (OD loading location).

From the K_{IH} study tests, there does not appear to be an observable difference in the calculated K_{IH} values between the different fuel designs and manufacturers as there is a greater scatter in the results within one test series than between different series. The calculated K_{IH} value for the assembly welds were between $9.4 \text{ MPa m}^{1/2}$ and $13.6 \text{ MPa m}^{1/2}$.

There also does not appear to be a difference between K_{IH} values on the ID and OD side of the weld. Since K_{IH} is a material property, differences in material geometries between the ID and OD side don't appear to significantly affect the material formed from the welding process. However, a difference is noticed in applied loads required to initiate DHC. A greater load appears to be required to initiate cracking on the outer side of the element than the inner.

The applied loads required to initiate DHC in the CFM fuel bundle are higher than the GE-H bundles, but the K_{IH} appears to be the same, although this observation is made with a small number of results. This is most likely explained by the lack of an observed weld discontinuity in the CFM bundle: the material property is the same as the GE-H bundles, but the stress riser is not present in the CFM bundle.

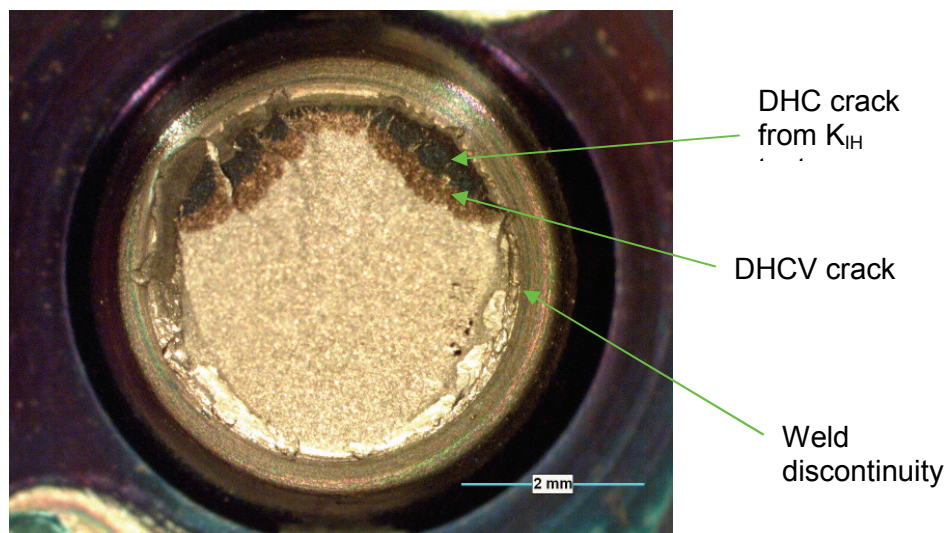


Figure 11: Metallographic Cross-Section of Assembly Weld after Inner Diameter K_{IH} and DHCV Test (Shek, 2010)

There is a difference in the K_{IH} values determined between the commissioning tests and the K_{IH} study tests. In the commissioning tests, the location of initiation considered in the calculation of K_{IH} was always at the top of the element. From two of the samples post-test weld examinations, 07-09 and 07-140, initiation appears to have started at approximately 45° ³, with applied loads of 65 N and 62 N and K_{IH} values of $8.3 \text{ MPa m}^{1/2}$ and $7.9 \text{ MPa m}^{1/2}$, respectively. For the K_{IH} study test samples 09-106 and 09-120, DHC initiation started at 45° and this was accounted for in the K_{IH} calculations. The applied loads were 64 N and 63 N with K_{IH} values of $11.2 \text{ MPa m}^{1/2}$ and $10.7 \text{ MPa m}^{1/2}$, respectively. Therefore, for the same sample type, there is agreement with measured load values for initiation, but the calculated values (bending stress and K_{IH}) differ.

The reason for the differences in the K_{IH} values between the two test series may be due to the difference in the bundles used between the series for test samples. However, the similarity in the measured applied loads and very similar weld geometries of the samples suggest the material performance was the same. One difference between the test series was the period the sample was held after DHC was initiated. In the qualification tests, the samples appear to be held at-load for approximately 200 to 800 hours, whereas the K_{IH} study test series samples were held at-load for 200 to 260 hours after initiation. It is possible the additional crack growth period may complicate the ability to determine where the crack growth originated.

In terms of the location for DHC initiation during the K_{IH} study tests, the 37-element samples all initiated to the side of the plane in which the load was applied (denoted as 45° in Table 3). This is consistent with the modelling work performed by Kinectrics (Wasiluk, 2010). For the 28-element samples, the ID initiation was in the plane of the applied load (denoted as 0° in Table 3), but to the side for the OD test. This is likely also explained by the work of Wasiluk (2010). The modelling work illustrated that when the weld extended to the edge of the endplate, and the endplate was not fixed at the weld, the maximum stress intensity factor around the weld was 45° from the endplate edge. However, when the weld did not extend to the endplate edge, or the

³ The report does not explicitly mention the location of initiation for the commissioning tests, but the crack depths measured were all taken approximately 45° from the top of the element and the crack had grown the most at these points.

endplate was fixed at the weld, the maximum K_I around the weld was at 0° . The 37-element outer-ring weld has a diameter that is nearly the width of the endplate, and therefore, both ID and OD loading should see DHC initiation at the sides. However, the 28-element weld diameter is smaller than the endplate width, and the OD side of the weld extends to the edge of the endplate, whereas the ID does not. This appears to explain why the location of DHC initiation varies from the ID to OD side for the 28-element bundles, whereas it does not for the 37-element bundle.

Table 2: Results of DHC Apparatus and Methodology Commissioning Tests

Test #	Test Bundle		Element	Loading Location	DHC Initiation		K_{IH} (MPa \sqrt{m})	Location for DHC Initiation
	Manu- facturer	# Elements			Load (N)	Bending Stress (MPa)		
07-09	GE-H	37	6	-	65	184	8.3	-
07-67	GE-H	37	2	-	62	163	7.7	-
07-104	GE-H	37	13	-	62	167	7.9	-
07-140	GE-H	37	15	-	62	170	7.6	-
07-164	GE-H	37	8	-	62	155	13.6	-

Table 3: Results of K_{IH} Study Tests

Test #	Test Bundle		Element	Loading Location	DHC Initiation		K_{IH} (MPa \sqrt{m})	Location for DHC Initiation
	Manu- facturer	# Elements			Load (N)	Bending Stress (MPa)		
09-106	GE-H	37	2	ID	64	282	11.2	45°
09-120	GE-H	37	5	ID	63	282	10.7	45°
09-136	GE-H	37	8	ID	72	334	11.1	45°
09-148	GE-H	37	15	ID	76	367	13.1	45°
09-124	GE-H	37	17	OD	83	346	12.2	45°
09-125	GE-H	28	2	ID	53	227	10.1	0°
09-130	GE-H	28	4	ID	53	214	10	0°
09-144	GE-H	28	15	ID	56	220	13.6	0°
09-172	GE-H	28	6	ID	53	225	9.4	0°
09-182	GE-H	28	8	OD	67	285	11.6	45°
09-135	CFM	37	6	ID	73 ¹	253	NA ²	45°
09-186	CFM	37	18	ID	85	277	12.2	45°
09-204	CFM	37	5	ID	80	255	12.1	45°
09-208	CFM	37	16	ID	80	325	NA ³	45°
10-16	CFM	37	9	ID	93 ¹	324	NA ²	45°

Table Notes:

¹ Crack initiation not picked up by potential drop measurement, load at initiation back calculated based on crack size and typical crack velocities to determine time at which cracking most likely initiated.

² K_{IH} not reported since it is based on extrapolated load.

³ Endplate portion of this sample contained portion of the radial web so finite element model could not be used to calculate K_{IH} .

5. DISCUSSION

The purpose of the finite element stress modelling presented in Section 3 and the DHC critical stress intensity testing in Section 4 is to evaluate if it is possible DHC cracking could be present in used CANDU fuel in dry storage containers. Comparison of the modelled K_I values for 28-element bundles with the experimental K_{IH} values provides a means to evaluate the presence of DHC in Pickering NGS used fuel. The work performed for this program strongly suggests that DHC is not expected to occur in 28-element fuel bundles. However, some uncertainties exist that were not addressed in this work.

5.1 Finite Element Maximum Assembly Weld Stress Intensity Factors

Full-bundle mechanical stress modelling of 28-element CANDU fuel models for varying conditions in horizontal dry storage was summarized in Section 3. Stress distributions were computed throughout the bundle, and for assembly welds with the largest stresses, the maximum stress intensity factor at the tip of a 0.5 mm weld discontinuity was calculated. For the purposes of dry storage, DHC of an outer-ring weld is of key importance, as it could result in a loose element that affects handling of the fuel bundle. It may be possible that a cracked weld on the interior of the bundle affects the bundle's response to impacts and vibration, but it is unlikely it would affect handling of the bundle structure.

One scenario evaluated a nominal bundle geometry (*i.e.*, no initial deformation and perfectly straight elements) in a DSC module tube. In this scenario, the maximum K_I value was calculated to be $1.01 \text{ MPa m}^{1/2}$ in an outer-ring element. The maximum tensile axial component of the stress field across the weld was found to be approximately 20 MPa and the element mid-plane displacement was approximately 0.3 mm.

The nominal bundle geometry is not considered a realistic scenario for the majority of used CANDU fuel bundles because post-irradiation examinations show that the bundles creep during irradiation. Using post-irradiation examination information as inputs into the fuel bundle geometry, another scenario was evaluated where a fuel bundle with a post-irradiation geometry was modelled in a DSC module tube. In this scenario, the maximum K_I value calculated was $2.45 \text{ MPa m}^{1/2}$ for an outer-ring element. The maximum tensile axial stress component across the weld was 44 MPa and the element mid-plane displacement was approximately 0.9 mm.

One other scenario was investigated where a bundle with an identical amount of bow of each element was modelled inside a DSC module tube. The amount of bow was taken to be that necessary to conform with current sag profiles of CANDU fuel channels. This scenario was to represent a situation where a bundle from an aged, sagged pressure tube is placed in a straight module tube. The geometry of the bundle is not truly representative of the bundle geometry from an aged channel as it does not consider diametrical expansion of the pressure tube and its effect on the bundle's geometry. However, it was investigated to see if ageing of the reactors could affect DHC initiation in used fuel. The calculations indicated the maximum K_I value would be for an intermediate-ring element with a value of $3.04 \text{ MPa m}^{1/2}$. However, the maximum K_I for an outer-ring element was $1.53 \text{ MPa m}^{1/2}$ with a maximum axial stress component of approximately 28 MPa and a mid-element displacement of approximately 0.6 mm.

The K_I calculations performed suggest the outer-ring values in used fuel may be as large as $2.5 \text{ MPa m}^{1/2}$. As well, the effects of post-discharge geometry on stress intensities at the assembly welds may also be larger than stresses induced by constraint of the bundles in module tubes

under gravitational loading, assuming there is no significant re-orientation of the used fuel in dry storage.

5.1.1 Effect of Zircaloy-4 Material Property Variations

The stress model developed for this work assumed there was no variation in the material properties of the Zircaloy-4 throughout the bundle (Popescu and Lampman, 2010). This is known not to be true of manufactured CANDU fuel bundles: the fuel sheath is specially textured compared to the rest of the Zircaloy-4 material which results in different mechanical properties, and there are multiple heat affected zones—assembly welds, closure welds, and appendage brazing areas—that would also affect the material properties. However, the unirradiated model validation exercises showed excellent agreement between the modelled predictions of element bending and the experimental measurements (Popescu and Lampman, 2010). This suggests that modelling the variations in Zircaloy-4 material properties throughout the bundle is not required to predict the bundle's response to element bending loads.

On a more local level, material property variations throughout the assembly weld would be expected to have an observable effect on DHC initiation. The DHC testing summarized in this report calculated K_{IH} values from measured applied loads on the test samples. A finite element model of the sample was prepared which accounted for an idealized geometry of the sample (*i.e.*, a smooth, circular crack front), but it did not model the material variations in the weld as the material properties are unknown. Similarly, the assembly weld submodels used to calculate K_I values accounted for an idealized weld geometry and homogeneous material properties. As such, the K_I and K_{IH} values determined in this work may not be true values for the CANDU assembly welds, but they were determined using the same assumptions and can be used for comparative assessment.

5.1.2 Effect of Fuel Pellets on Fuel Element Bending

The fuel pellets within the fuel sheath have a large effect on the stiffness of an element. The fuel element model contained individual pellets and their contacts with adjacent pellets and the surrounding sheath. This was required to match the mechanical response of the unirradiated fuel element during validation tests. Prior to irradiation, the fuel pellets are solid, homogeneous, and have a well defined geometry. However, after irradiation, the pellets contain radial and circumferential cracks and chips and the grain structure is no longer homogenous in the pellet. Insufficient data exists on mechanical behaviour of irradiated fuel pellets to accurately model their behaviour. Due to the uncertainties in modelling irradiated pellets, they were removed from the irradiated fuel models used to evaluate stresses and K_I values in dry-stored CANDU fuel bundles.

To investigate the effect irradiated pellets may have on the fuel element bending and assembly weld K_I values, a comparison of bending of a fuel element without pellets and one with solid pellets in very near contact with the sheath was performed (Lampman and Popescu, 2010). Under identical loading conditions, the introduction of the pellets reduced the mid-element displacement, or bending, by approximately 40% and the maximum assembly weld K_I reduced by approximately 50%. The solid pellet in contact with the sheath is probably a bounding case for stiffening of the element, but this suggests that the presence of irradiated fuel pellets would reduce the K_I values at the assembly welds, possibly by a factor of two. Therefore, the modelled response of used CANDU fuel bundles in dry storage without the pellet-to-sheath interaction is expected to be conservative with respect to DHC.

5.1.3 Effect of Weld Morphology and Crack Depth

The DHC testing performed by Kinectrics (Shek, 2010) provided a large amount of good information on the assembly weld morphology. The variability in the morphology was clear from the post-test weld cross sections. In some cases, the weld and discontinuity seemed to have an elliptical shape in the plane of the weld, whereas in other cases the shape was more round. Kinectrics investigated the effect of elliptical versus round weld profiles on the computed K_I values and found the difference to be within approximately 10% of each other (Wasiluk, 2010). Therefore, there is an uncertainty in stress intensity factors calculated using idealized geometries compared to the real geometries, but the effect appears to be minor.

A feature of the assembly welds that would have a more significant effect on the calculated stress intensity factors is the discontinuity depth. The stress model calculations assume a depth of 0.5 mm. However, the discontinuity depths appear to be mainly in the range of 0.2 mm to 0.3 mm (Shek, 2010). The relationship between stress intensity factor and crack depth for a circumferential crack in a rod is,

$$K_I \propto \sqrt{c},$$

where c is the crack depth. Therefore, accounting for a more realistic weld discontinuity depth, the K_I values would be reduced by approximately 25% to 35%.

5.2 28-Element DHC Initiation Test Measurements

As given in Table 3, the unirradiated DHC testing of 28-element CANDU fuel found K_{IH} values in the range of 9.4 MPa m^{1/2} to 13.6 MPa m^{1/2} for DHC initiation in outer-ring elements (Shek, 2010). Based on the tests performed, it is not possible to determine if there is a difference in initiation between the inner and outer diameter of the endplate. The 28-element values are consistent with the K_{IH} values ranging between 10.7 MPa m^{1/2} to 13.1 MPa m^{1/2} for the tested 37-element fuel samples from the K_{IH} study tests (Shek, 2010). Values as low as 7.6 MPa m^{1/2} were recorded by Kinectrics for a GE-H 37-element fuel during the commissioning tests, but the location of crack initiation is not clearly understood and could increase the K_{IH} values. Apart from the commissioning test results, it appears there is no systematic difference in the K_{IH} value between the different fuel designs and manufacturers.

While the K_{IH} values appear to be consistent between manufacturer and fuel design in the K_{IH} study test series, there are observable differences in initiation load and bending stress. For the 28-element bundle, the initiation load and bending stress for the inner diameter side of the endplate was approximately 53 N and 220 MPa, respectively. However, for the outer diameter side of the endplate, the equivalent values were slightly higher at 67 N and 285 MPa and the crack initiated 45 degrees from the radial direction, which is likely a result of the different loading

Table 4: Summary of DHC Critical Stress Intensity Factors for Irradiated Zircaloy-2

Material	Temperature (°C)	Irradiation	Hydrogen (ppm)	K_{IH} (MPa m ^{1/2})
Zircaloy-2 Cladding (Efsing and Pettersson, 2000)	200	33.9 MWd/kgU	560–1,000	12.5 ± 0.2
		35.9 MWd/kgU	1,200–1,900	12.7
	300	33.9 MWd/kgU	560–1,000	9.9 ± 0.3
		35.9 MWd/kgU	1,200–1,900	9.9 ± 0.2
Electron Beam Welded Zircaloy-2 (Schofield et al., 2002)	200–255	3–5 x 10 ²⁵ m ⁻²	55	8–12

conditions. This observation suggests greater bending of the element is required to initiate DHC for elements bowed into the bundle than away. An increased load to initiation of DHC on the outer diameter of the 37-element bundle was also observed, and differences in initiation loads between the 37-element bundle manufacturers are apparent.

5.2.1 Effect of Irradiation

The K_{IH} values determined from this work are for unirradiated material, and a question remains as to the effect irradiation will have on the critical stress intensity factor. There does not appear to be any published data on irradiated Zircaloy-4 K_{IH} values. However, some information on irradiated Zircaloy-2⁴ cladding and electron-beam welded material is available and is presented in Table 4.

The work on high-hydrogen, high-irradiation Zircaloy-2 cladding⁵ (Efsing and Pettersson, 2000) found K_{IH} values similar to the unirradiated Zircaloy-4 weld results. Efsing and Pettersson commented that the effect of irradiation in Zircaloy material was observed to be less than theoretical models. For the 200°C temperature tests, the effect of irradiation lowered the measured K_{IH} by approximately 10% of the unirradiated value with the effect increasing at higher temperatures. The work on high-irradiation electron beam welded Zircaloy-2 (Schofield *et al.*, 2002) measured K_{IH} values within a range very similar to those measured as part of this work.

DHC initiation is sensitive to the material and micro-structure, and the K_{IH} values determined for Zircaloy-2 are not directly applicable to Zircaloy-4. However, given the alloys are very similar and the apparent similarity in K_{IH} values in the literature and determined by Kinectrics (Shek, 2010), it is expected that the effect of irradiation will be similar on K_{IH} between the two alloys. Therefore, irradiation will slightly lower the measured unirradiated assembly weld K_{IH} values, possibly by an approximate amount of 1-2 MPa m^{1/2}.

5.3 Evaluation of the Presence of DHC during Dry Storage

The results of the CANDU fuel bundle stress model analysis on 28-element fuel bundles suggest stress intensity factors in the assembly welds would not exceed 3 MPa m^{1/2}. Based on the conservatism in the models from lack of fuel pellets and deeper cracks than measured, the upper bound may be more realistically around 1 MPa m^{1/2}. The K_{IH} testing suggested stress intensity factors in the range of 9.4 MPa m^{1/2} to 13.6 MPa m^{1/2} would be required to initiate DHC in the assembly welds. The qualification tests suggest lower K_{IH} values in the range of 7.6 MPa m^{1/2} to 13.6 MPa m^{1/2}, although the location of initiation of the cracking is not well understood in these tests. Considering the effect of irradiation, the K_{IH} values would be lower, probably by 1-2 MPa m^{1/2} based on Zircaloy-2 testing. The difference between the expected K_I and K_{IH} values is still significant and DHC is not expected during dry storage of used 28-element CANDU fuel bundles in a DSC.

⁴ Zircaloy-4 is a very similar alloy to Zircaloy-2 with the same crystal structure, a slightly higher iron content (approximately 0.04 to 0.11 %wt), and the removal of Nickel (accounting for approximately 0.08 %wt of Zircaloy-2) to reduce the amount of hydrogen uptake (Wah Chang, 2003).

⁵ The samples have high levels of hydrogen and higher irradiations when compared to commercial CANDU fuel.

This conclusion makes sense from the perspective of total bending of the fuel element. Large deflections of the fuel elements were noted during the DHC testing. A 2.5 mm deflection of the free end of a 37-element fuel element sample (approximately 70 mm from the weld) resulted from the 48 N loading case (Shek, 2010). All tested samples required larger loads, and thus larger deflections, to initiate DHC. Therefore, a fuel element would need to bend by possibly more than 9 millimetres at the element mid-point to initiate DHC. This level of bending is not expected, and may not even be possible in constrained module tubes.

The stress intensity factors in used 37-element fuel bundles were not evaluated in this work. However, the conclusion that DHC is not present is expected to hold as the K_{IH} values appear to be the same, the weight and overall dimensions of the bundle are similar, and the weld stress intensity factors are unlikely to be several multiples greater than the 28-element values as the bending constraints posed by the element interactions and module tube dimensions are similar.

5.3.1 Remaining Uncertainties

A key uncertainty remaining from this work is the effect of residual stresses in the assembly weld region due to manufacturing processes. It was noted from the DHC testing program (Shek, 2010) that once the intact bundle was cut, the elements sprung out from the bundle's axis. This suggests that there is a significant residual stress induced in the bundle from manufacturing. However, the magnitude of the residual stress and its effect on delayed hydride cracking at the assembly welds is uncertain.

It is expected that the magnitude of the residual stress field will be reduced during irradiation. During irradiation, the fuel is exposed to a high neutron flux at nearly 300°C for approximately one year before discharge. During this period, the bundles creep into a new shape determined by the pressure tube constraints. This has been observed in post-irradiation examinations of used CANDU fuel. The residual stress in the bundle from manufacturing is expected to relax during the creep process. However, the amount of creep relaxation was not analyzed in this work and remains unknown⁶.

The actual orientation of the fuel bundles in dry storage modules is unknown. It was assumed in this analysis that the orientation in storage is identical to the orientation in the fuel channel, but if this is altered, it may be possible to reach higher stress intensity factors at the weld discontinuity. However, the K_{IH} testing also indicates that mid-element displacements larger than several millimetres, which is much greater than calculated in the stress models, would be required to initiate DHC. This increased level of deformation is not expected by varying the orientation of the bundle in dry storage.

Another uncertainty in this analysis is the effect of varying used fuel geometries. During irradiation, the spacer pads, which maintain the separation of adjacent fuel elements, wear and this would have an effect on the amount of element bending that may occur. A relatively minor amount of Post-Irradiation Examination data exists and it was used to evaluate the effect of permanent element bowing on DHC, but other bundle conditions—such as mechanical damage due to interaction with fuel channel or fuel handling objects—have not been accounted for. Much more detailed Zircaloy-4 material properties would be needed, such as non-linear properties, to account for this, but this data does not exist for irradiated Zircaloy-4. These

⁶ Anecdotally, fuel bundles have been manufactured and used for demonstration purposes for decades and there are no known instances of unexpected cracking of the assembly welds.

mechanical damage observations on used fuel are rare, but such a condition may possibly promote DHC.

While the bowing of used CANDU fuel elements has been considered in the models, the deformation of endplates has not. Currently, the modelled endplates are perfectly flat, but during manufacturing the endplate at the welds tends to be pushed into the bundle relative to the non-weld endplate sections. Furthermore, irradiation can lead to endplate doming (concave or convex distortion) or parallelograming (axial skewing) of the endplates. The effects of this geometry on DHC initiation during dry storage have not been analyzed.

The effect of irradiation on the Zircaloy-4 material properties is also a source of uncertainty. An expected effect on the K_{IH} value for the weld material has been discussed based on a literature review of similar materials, but direct observation of this effect was not made. The effect of irradiation on material properties used in stress modelling was taken from MATPRO (MATPRO, 2003), which should be reasonable, but verification of the properties was not possible. The magnitude of the effect of the UO_2 fuel pellets on element bending is unknown, but a conservative approach was taken to bound the stress analysis so the irradiation uncertainties are relatively minor.

6. CONCLUSIONS

The work summarized in this document was performed as part of the NWMO's Used Fuel Integrity program to evaluate the condition of used fuel during dry storage. More specifically, this work examined whether DHC is a likely degradation mechanism during dry storage in OPG Dry Storage Containers for periods extending up to 100 years. The results of the experimental DHC initiation study and finite element stress modelling of CANDU fuel bundles suggest that DHC will not occur in used fuel during dry storage.

The work performed on DHC initiation has focussed on the condition of used fuel at the start of dry storage where the temperature of the fuel is between 130°C to 150°C. The post-discharge geometry of the fuel elements has been considered and the stress distribution throughout the bundle was calculated for 28-element fuel bundles resting horizontally in dry storage containers. The calculated stresses and deformations are relatively low with no mid-element bending more than 1 mm. The assembly welds of the fuel bundles are known to have weld discontinuities, which are effectively cracks due to the sharpness of the discontinuity tip, and the stress intensity factors at the crack tips resulting from element bending were calculated. All calculated values were relatively low and remained below 3 MPa m^{1/2}. These calculations were based on conservative models without the stiffening effect of the UO₂ fuel pellets within the fuel elements and deeper weld discontinuity depths than observed in this program.

To evaluate whether the calculated stress intensity factors were sufficient to initiate DHC, an experimental program was created to determine critical stress intensity factors for DHC in CANDU fuel assembly welds. This work suggests that the K_{IH} value for the unirradiated Zircaloy-4 assembly weld is in the range of 7.6 MPa m^{1/2} and 13.6 MPa m^{1/2}. The effect of irradiation on K_{IH} was not tested, but based on similar materials it is expected to lower K_{IH} by a relatively minor amount. The CANDU 6 fuel bundles manufactured by Cameco Fuel Manufacturing did not appear to contain a weld discontinuity and initiation loads were higher, suggesting they are less sensitive to DHC.

There are several remaining uncertainties in this analysis. The effect of residual stresses from the manufacturing process has not been evaluated. While unknown, it is not expected to have a significant effect on initiation due to creep relaxation of the fuel bundles while in the reactor cores. The effect of used fuel with abnormal geometries, such as observable mechanical deformation, is also unknown. However, as determined by Lazaroski *et al.* (2005), the percentage of fuel with abnormal geometries is less than 1% of all used fuel, and the majority of fuel within the 1% exhibits very minor deformation unlikely to result in DHC initiation.

Comparison of the conservatively calculated maximum stress intensity factor of 3 MPa m^{1/2} with the minimum critical stress intensity factor of 7.6 MPa m^{1/2}, which may be lowered by approximately 2 MPa m^{1/2}, required to initiate DHC indicates that DHC is not likely an active degradation mechanism for dry-stored CANDU fuel in a Dry Storage Container as there is sufficient margin to account for remaining uncertainties.

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