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HYDROGEN IN CANDU FUEL ELEMENTS

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ABSTRACT

Unirradiated and irradiated CANDU* fuel cladding was tested to compare the role of stress-corrosion cracking and of hydrogen in the development of fuel defects. The results of the tests are compared with information on fuel performance in-reactor. The role of hydriding (deuteriding) from the coolant and from the fuel element inside is discussed, and the control of "hydrogen gas" content in the element is confirmed as essential for defect-free fuel performance. Finally, implications for fuel element design are discussed.

INTRODUCTION

In 1984, Unit 3 at the Bruce-A Nuclear Generating Station (NGS) experienced systematic fuel failures among bundles built by Combustion Engineering-Superheater (CES). CES was one of three fuel manufacturers that had fuel in all four Bruce-A units at the time. Unit 3 also had fuel from the other two manufacturers: General Electric of Canada (GEC) and Westinghouse Canada (WECAN), now named Zircatec Precision Industries (ZPI). This fuel did not show systematic failures.

The Bruce bundle has the end bearing pads brazed on outer fuel elements in staggered locations. Because of this, the two ends of each outer element have different geometry and properties. At the "out-board" end, where the end bearing pad is located close to the end cap, the braze heat-affected zone (HAZ) extends to the very end of the sheath. The other end ("in-board") has the end bearing pad away from the end cap, such that the as-received (AR) condition of the tubing is preserved at the sheath end. Thus, at the "out-board" side, the end cap is welded to the braze HAZ of the sheath, while at the "in-board" end, it is welded to the AR sheath.

The CES fuel failures were characterized by circumferential cracking in the end cap weld at the "out-board" end of high-powered outer fuel elements.

The results of the post-irradiation examinations (PIE) were inconclusive. The suspect defect mechanism was either stress-corrosion cracking (SCC) or delayed hydride cracking (DHC) [1-3]. The investigation did not explain why the defects preferentially occurred

* CANDU: CANada Deuterium Uranium; registered trademark.

- among CES bundles;
- at Bruce-3 unit; and
- at the out-board ends of outer elements.

CANDU fuel is built to common technical specifications that specify several of the requirements as performance requirements. This gives the manufacturer some flexibility for detailed design variations (e.g., in the design of the end cap weld, pellets, or fuel-element internal geometry). Also, at the Bruce-A NGS, the conditions have been maintained within common specifications that permit slight differences between reactor units (e.g., in hydrogen level in the coolant).

The results of the Bruce-3 incident investigation suggested that differences in fuel-element detailed design and in in-reactor conditions played roles in the primary defect mechanism.

To understand the primary defect mechanism, the CANDU Owners Group (COG) embarked on an experimental program designed primarily to investigate the development of similar defects, and to compare the role of SCC and of hydrogen (i.e., DHC) in unirradiated and irradiated fuel cladding.

During the test program, the following information became available, indicating that hydrogen can play a significant role in the development of primary defects:

- a) excess hydrogen due to the underbaking of the graphite Canlub coating was believed to be responsible for systematic fuel failures at Point Lepreau in 1991/92 [4], and
- b) defects related to primary hydriding (blisters) were found in a prototype 43-element fuel bundle subjected to severe overheating in the NRU reactor in 1990 [5].

In this paper, we compare the test results with relevant information on CANDU fuel performance in-reactor. The objective is to evaluate the existing fuel-design requirements related to hydrogen.

COG EXPERIMENTAL PROGRAM

As-manufactured, hydrided and irradiated cladding material was tested in a neutral environment and in iodine vapour. Material and experimental methods are described in [3, 6].

Hydrogen (deuterium) was introduced into the cladding material using four different methods:

- a. Pre-hydriding ("static hydriding") of unirradiated cladding: Hydrogen was introduced into and redistributed in the specimen before the application of the mechanical load. Hydrogen concentrations ranged from low (solubility limit at 300°C) to as high as 300 ppm.
- b. "Dynamic hydriding": Hydrogen was introduced
 - b1) at the cladding outside (in a Ni layer at the outside surface), or
 - b2) at the cladding inside surface (as undercured Canlub),
 and redistributed (adsorbed and redistributed) in the cladding after the application of the mechanical load (i.e., during the test).

- c. Deuteriding from the coolant, and hydriding from element inside, of reactor fuel cladding in-reactor.

Methods a, b1 and b2 worked with a uniform temperature (300°C) throughout the specimen. In case c, the (relatively slower) deuteriding and hydriding took place in a temperature gradient. There were no stresses in the cladding during the "static hydriding" (method a). The "dynamic" methods (b1, b2) were redistributing hydrogen in the presence of tensile stresses. The in-reactor deuteriding/hydriding (c) proceeded under loads from coolant pressure and pellet volume changes.

The test results are presented as % of cracked or fractured specimens in Figure 1 (Expanding mandrel tests) and as defect thresholds in Figure 2 (Internal pressurization tests). Note that tests with cladding produced by all three Canadian manufacturers are merged in these two summaries. Compared to the effect of material condition (as-manufactured, irradiated, pre-hydrided) and test condition (neutral environment, iodine vapour), the effect of the weld design was of lesser importance. Therefore, for the purpose of this paper, we decided to disregard it.

A strengthening effect of pre-hydriding is apparent; this confirms earlier results [7]. The (slow) deuteriding and hydriding in-reactor has, in general, a similar effect on the defect threshold of the irradiated material. Dynamic hydriding from the outside surface resulted in some alignment of hydrides in high-stress areas; however, fracture has not been achieved. Dynamic hydriding from the inside results in a small reduction of the threshold, if compared with the as-manufactured condition. It results in a large reduction, if compared with the pre-hydrided condition.

INFORMATION FROM REACTOR FUEL

Experience with fuel performance in-reactor yields additional information on hydrogen/deuterium behaviour in the fuel elements. In particular, we will pay attention to the following:

- deuteriding from the coolant (through the cladding outside surface);
- hydriding from sources inside the fuel element (through the inside surface):
 - . formation of hydride blisters from moisture inside the fuel elements,
 - . hydriding of the cladding in fuel elements with undercured Canlub,
 - . sheath hydriding at high temperatures; and
- distribution of hydrogen in the sheath under stress and temperature gradients.

DEUTERIDING FROM THE COOLANT

Three factors strongly influence the ingress of deuterium in the cladding [8, 9]: the presence of neutron fluence, the condition of the coolant, and the condition of the cladding.

Figure 3 shows in-reactor hydriding rates for AR and braze HAZ parts of the sheath in low-power and high-power positions, and compares them with out-reactor rates. Note that for high-power positions, the average deuteriding rate is ~0.6 ppm D/day for braze HAZ of the sheath, and ~0.3 ppm D/day for AR sheath. After longer residence times, the differences in deuterium contents of HAZ and AR parts of the sheath become smaller, as illustrated in Figure 4.

HYDRIDE BLISTERS FROM MOISTURE

One of the lessons learned from the early experience with CANDU fuel (prior to the introduction of Canlub) was that excess moisture content inside a fuel element can lead to the formation of hydride blisters, cracks in the sheath, and eventually defects.

A volume increase accompanies the formation of hydrides in Zircaloy [10]. When hydrogen from the moisture enters the sheath, it is often localized in a highly hydrided zone. Because of the interaction with other parts of the sheath, the hydrided zone is under compressive stresses, surrounded by low-hydrogen sections under tensile stresses. This leads to hydrogen diffusion into areas under tensile stresses, and eventually to growth of cracks and through-wall fractures. To prevent this, a limit was introduced for the content of hydrogen gas per fuel element [11].

PRIMARY HYDRIDING FROM CANLUB

Uncured or undercured Canlub contains hydrogen that gets released from its bond in the Canlub layer early after bundle loading in the reactor at normal operating conditions. This "hydrogen gas" is available for absorption by the sheath. This is the mechanism responsible for the 1991/92 defects in Point Lepreau. Up to 4 mg of hydrogen gas have been found in as-manufactured fuel elements with undercured Canlub. Defects have been detected in bundles that were exposed to a steady high power above a power-burnup threshold line. Unlike the SCC defects, the sheaths with undercured Canlub did not need any power ramp to defect [4]. Figure 5 compares thresholds for "undercured Canlub defects" with power-ramp defects [11]. The ramped powers and power ramps for Bruce-3 defects are also shown in the figure.

Hydride blisters with the accompanying cracks have been found in the sheaths of two elements of a prototype 43-element bundle that was severely overheated during irradiation at Chalk River Laboratories (CRL) [5].

Tests done on graphite coating of fuel elements for the Italian research reactor "Cirene" [13] demonstrate that the Canlub layer contains hydrogen even after good curing, and that this hydrogen gets released (becomes "hydrogen gas") if the graphite layer reaches temperatures above the curing temperature. A correction for differences between the CANDU and Cirene graphite layer (diameter, thickness, curing temperature) gives the quantity of "additional hydrogen gas" available for high-temperature transients as 0.4 mg per fuel element in a 37-element bundle.

HYDROGEN (DEUTERIUM) DISTRIBUTION IN A SHEATH WITH STRESS AND TEMPERATURE GRADIENTS

Sawatzky [14, 15] developed a model that explains why hydrogen concentrates in zones at low temperatures or under high tensile stresses.

In the absence of significant stress gradients, temperature gradients across the sheath wall govern the distribution of hydrogen (deuterium) in the sheath. Typically, H and D contents are the highest at the sheath outside surface, and the lowest at its inside surface.

Hydrogen and deuterium analyses of intact elements in CES fuel involved in the Bruce-3 incident indicate some deviations from the usual pattern. Figures 6 and 7 show the distribution of hydrogen and deuterium between the end caps, braze HAZ and AR sheath of such fuel elements. For comparison, similar data are shown for WECAN fuel that was used in the Bruce-3 reactor under comparable conditions to the CES fuel, and performed well.

In CES fuel, the content of H in the end caps (both "in-board" and "out-board") is unusually high. In view of Sawatzky's model, this indicates high tensile stresses in the end caps right after loading in the reactor.

Figure 8 shows the increases of isotopic hydrogen contents in WECAN and CES fuel elements. The increases, defined as differences between the contents found during PIE of discharged fuel, and in as-manufactured fuel, can be made up of two contributions: hydrogen gas ingress (from the element inside) in-reactor, and hydriding (from the outside) of the discharged fuel in the bay. Thus, the values found are an upper bound for hydrogen gas ingress in-reactor.

The total for WECAN elements (1.05 mg) is obviously affected by this additional hydriding after discharge; to obtain an estimate of hydrogen gas, it should be corrected downward – there is no reason to expect hydrogen gas contents as high as 1 mg/element in this fuel. Note that the total for CES fuel elements (0.78 mg) is below the specification limit for hydrogen gas even without correction for possible hydriding after discharge.

DISCUSSION

COG Program: DHC vs. SCC

One of the objectives of the tests was to compare SCC and DHC as defect mechanisms for fuel cladding, and for the end cap welds in particular. The tests demonstrated that at 300°C, the presence of a SCC-active species always reduced the defect threshold significantly. The presence of hydrogen caused a limited reduction of the defect threshold in some cases, but increased the threshold in many others. There are earlier reports with similar observations. For example, Reference [7] states that hydrides increase the SCC resistance, and eventually limit the effect of iodine to pitting; i.e., render Zircaloy resistant to SCC by iodine.

To explain this, together with other effects of hydrogen described in this paper, we have to consider not only DHC, but also other phenomena related to hydrogen.

The Role of Hydrogen in the Development of Fuel Defects

Within the scope of this paper, we consider three categories of hydrogen-related mechanisms:

- hydrogen as the cause of mechanical loads (stresses),
- hydrogen as the element involved in the mechanism of crack growth, and
- hydrogen as the hydride-forming component; i.e., properties of alpha-Zircaloy with hydrides.

The formation of blisters in the sheath falls into the first category. Compared to the original alpha solution, solid hydrides of the blister cause a significant increase in volume [10]. This

imposes a stress field on both the blister and the surrounding microstructure. The stresses are predominantly compressive in the blister, and tensile in the surrounding parts of the sheath.

DHC belongs to the second category. Under tensile stresses in front of a crack tip, DHC is the mechanism of hydrogen migration, hydride growth, realignment and crack propagation. The stresses may come from a blister present in the sheath, or from pellet expansion.

To characterize the third category, let us consider the hexagonal solid solution of alpha Zr with large particles of hydrides that do not dissolve completely during changes of stress or temperature. Hexagonal alpha-Zircaloy has high resistance to crack growth across the basal planes of its grains. There is a strong crystallographic correlation between the hydrides and alpha zirconium. Unless there are steep stress gradients in the solid solution, hydrides have platelet morphology, and their habit plane is parallel with the basal plane of alpha zirconium [10]. Such precipitation increases the resistance to crack growth in the direction normal to the basal plane. Because the tubing for CANDU sheaths is manufactured with a texture where the basal planes have hoop-axial orientation (basal poles have radial orientation), resistance to crack growth across the wall (i.e. in the radial direction) increases because of the presence of hydrides.

Deuteriding from the Coolant

The condition of the coolant, such as the degree of radiolysis, temperature, and coolant chemistry, governs the amount of deuterium available for ingress into the cladding [8].

Deuterium pickup by the cladding then depends on the cladding surface condition and/or microstructure. Figures 3 and 4 confirm the preferential deuteriding of the braze HAZ of the sheath, likely related to the higher oxidation rate of the outside surface in braze HAZ parts of the sheath [9]. A gradual decrease of the concentration differences between HAZ and AR can be explained by deuterium diffusion along the concentration gradient in the axial direction, and by the effect of higher D concentrations in the sheath surface on the pickup rate.

Because of the heat flow from the pellets to the coolant, there are temperature gradients across the sheath. In the absence of stress gradients, this results in a deuterium concentration gradient in the wall, keeping most of the deuterium in cooler outside layers [14, 15].

The rate of deuterium ingress in the sheath is relatively low. At the rates that can be derived from Figure 3 for high-power positions, it would take approximately two full-power days to reach the D solubility limit in a layer at the OD as thin as 1% of wall thickness. Because of a high neutron flux there, the stress relaxation rate is high, and no serious tensile stresses should be expected in the section of the wall between the deuterided layer and the inside surface.

We can thus conclude that deuteriding through the outside surface does not constitute a root cause of fuel defects. At most, it may act as an additional factor, if other causes for crack growth in the cladding are present.

Hydriding from the Fuel-Element Interior

There are three sources of hydrogen gas inside the fuel element: i) pellets, ii) filling gas, and iii) Canlub. As long as the manufacturing process is under control, the hydrogen content in pellets

and in the filling gas should be low. However, Canlub contains significant quantities of hydrogen, even after good curing. Some of this hydrogen is released at normal operating temperatures, some at higher temperatures.

Thus, there is hydrogen inside the fuel element available for pickup by the cladding immediately after, or soon after, bundle loading in the channel. Figure 6 shows that, for CES fuel, the hydrogen pickup at the cladding inside surface can be localized, depending on the presence of tensile stresses and strains. The combination of these two conditions, the quantity of hydrogen gas and degree of localization, should be considered when the effects of hydrogen gas are being assessed.

Factors Affecting the Risk of Canlub-related Hydriding Defects

Canlub is used in CANDU fuel sheaths to improve their resistance to SCC. In some respects, improvements of this SCC-protective function of Canlub may not be consistent with the desire to minimize the content of hydrogen gas. Therefore, a balanced approach has to be adopted.

Sheath protection from SCC improves with Canlub thickness [16]. On the other hand, the quantity of residual hydrogen, and with it the quantity of hydrogen gas from Canlub, is also proportional to Canlub thickness. This may become important in the development of CANDU fuel for high burnup.

Chan et al. [17] have presented evidence that the SCC-protective effect of Canlub depends on the presence of some hydrocarbons in the layer (i.e., on Canlub curing conditions): a low degree of curing gives the best SCC-protection, a high degree reduces the protecting effect. This, of course, means that the "best Canlub" will have relatively high residual hydrogen.

Curing under conditions of mass production offers additional complications. A batch of tubes in vacuum is difficult to heat uniformly to curing temperature. There will be a distribution of temperatures and times-at-temperature during the curing cycle, resulting in a distribution in the degree of curing within a production batch of sheaths.

Bruce-3 Incident Re-examined

The data presented in this paper provide an insight into the mechanisms responsible for the CES defects that occurred in Bruce-3.

Considering the results of chemical analysis of CES bundles discharged from Bruce-3 (Figures 6 and 8), the hydrogen gas content in as-manufactured fuel elements was obviously below 1 mg/element. Thus, it satisfied the specification requirement.

The internal design of the CES fuel element at that time included several factors that caused a buildup of high tensile stresses in the end caps and end cap welds during irradiation [1, 3, 6]: a short axial gap, the use of standard pellets at stack ends (large pellet diameter facing the end cap), and high pellet density. The fact that a large fraction of hydrogen gas was picked up by the end caps (both out-board and in-board) indicates that high tensile stresses existed in the end caps early after bundle loading in the reactor, i.e., in the initial low-power position. Some of the stresses got relaxed through time in this initial position, but the shift to a high-power position

(position 3 to position 7, in most cases) brought them back to high levels.

At the time of the shift, the fuel burnup was 40 to 120 MWh/kg U – enough to make fission products available in quantities sufficient for SCC. With reference to the COG testing program (Figures 1 and 2), SCC is the low-threshold defect mechanism under such conditions. We therefore conclude that SCC was the principal crack-growth mechanism. However, in the initial low-power position, hydrogen was entering the end caps and welds under high tensile stresses, and hydrides were precipitating stress-aligned. Thus, hydrogen was likely an additional factor involved in the initiation of cracks.

High tensile stresses in the end cap and end cap weld, related to insufficient clearances and high densities, were the root cause of the defects.

Fuel Design

In the existing technical specification, the importance of the "hydrogen gas" content in fuel elements is recognized. The limit, max. 1 mg H/fuel element for 28- and 37-element fuel bundles, has been in place for a number of years, with fuel performance generally free of hydrogen-related defects. The experience with undercured Canlub in Point Lepreau [4] also indicates that the limit is justified, at least for present fuel designs and conditions of use. The population where defects occurred had hydrogen gas distribution far beyond 1 mg/fuel element, and defects were found in outer elements at high linear powers only.

There are, however, several design-related areas where attention should be paid to the role of hydrogen gas in the fuel element:

- a. Development of Canlub. Changes in Canlub composition, thickness and curing conditions will have their impact on the quantity of hydrogen gas and on its release from the layer.
- b. Development of CANDU fuel for high burnup. A "better" and probably thicker Canlub layer may be needed, to last to higher burnups than those at present, with the consequence that the fuel element will contain a higher quantity of hydrogen gas.
- c. Uranium content in fuel elements. The combination of hydrogen gas availability in the fuel element, and tensile strain/stress at the cladding inside surface, can lead to critical conditions, and therefore deserves careful assessment. At present, technical specifications for CANDU fuel do not address this factor directly.

CONCLUSIONS

The results of the COG test program were reviewed in combination with information on the effects of hydrogen and deuterium on fuel performance in-reactor.

To understand the effects of hydrogen, we considered the following three processes:

- hydrogen-related volume changes and accompanying stresses,
- hydrogen-related crack growth, and

- microstructural strengthening by hydride platelets.

Deuteriding from the coolant and hydriding from the fuel-element interior were compared, and their potential as causes of fuel defects was assessed.

The existing specification limit for hydrogen gas in fuel elements has been reviewed. The experience with fuel performance, including hydrogen-related defects, indicates that the present limit for hydrogen gas is justified, at least for present fuel designs and conditions of use.

The mechanisms involved in the 1984 Bruce-3 defects were reviewed. We have concluded that the root cause of the defects was high tensile stresses related to an improper internal design of the fuel elements, and the principal cracking mechanism was stress-corrosion cracking.

In programs where there is a potential for larger hydrogen gas releases (such as the development of Canlub and the development of CANDU fuel for high burnup), attention should be paid to the risk of hydrogen-related defects.

Additional hydrogen gas may be released during high-temperature transients. The risk of fuel defects should be assessed for such cases.

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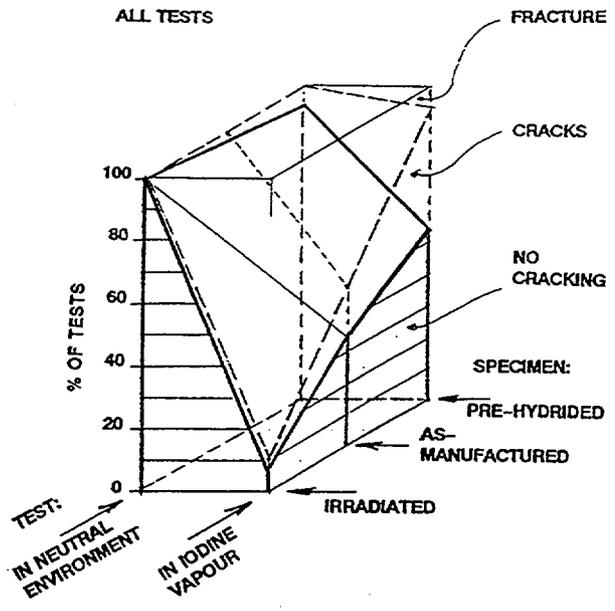


Figure 1 - SUMMARY OF EXPANDING MANDREL TESTS (COG).

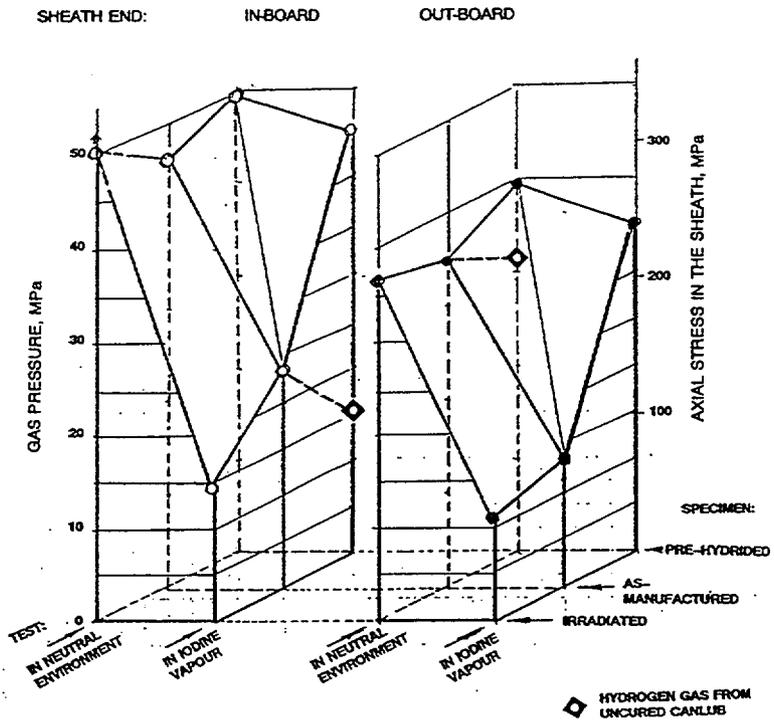


Figure 2 - INTERNAL PRESSURIZATION TESTS: SUMMARY OF DEFECT THRESHOLDS

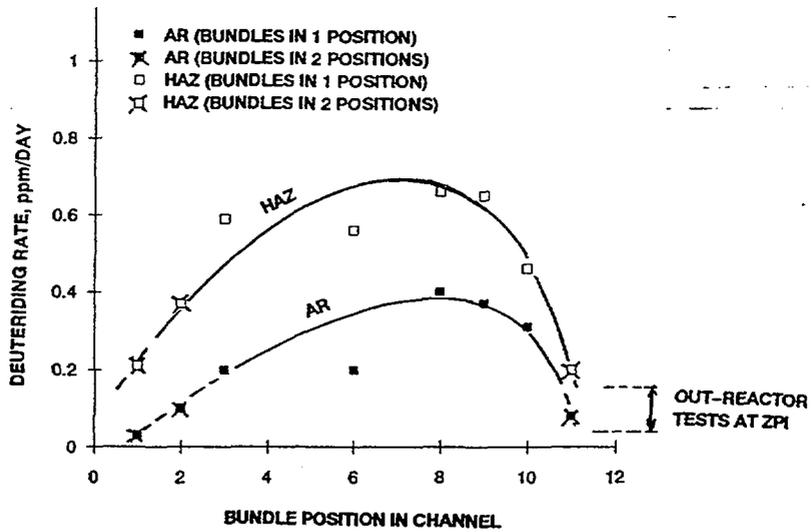


Figure 3 - EFFECT OF NEUTRON FLUENCE ON HYDRIDING RATES (COG).

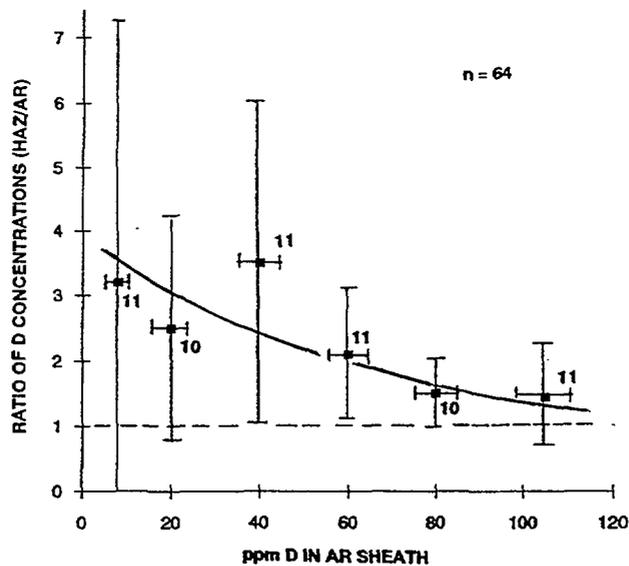


Figure 4 - EFFECT OF RESIDENCE TIME ON THE DIFFERENCES IN D CONTENT IN THE BRAZE HAZ AND AR PARTS OF THE SHEATH. Pickering, Bruce and Point Lepreau Fuel.

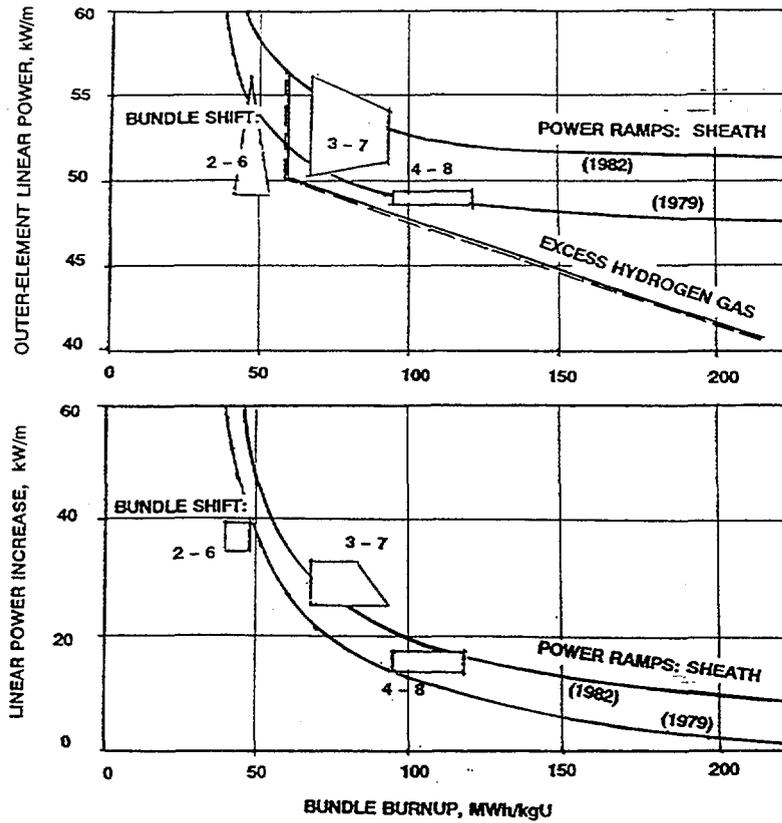


Figure 5 - DEFECT MAP FOR 37-ELEMENT FUEL BUNDLES:

- Power-Ramp Thresholds 1979 and 1982 [12];
- Defect Thresholds for Excess Hydrogen Gas in Point Lepreau Fuel [4];
- End Cap Weld Defects, Bruce-3 Incident (COG).

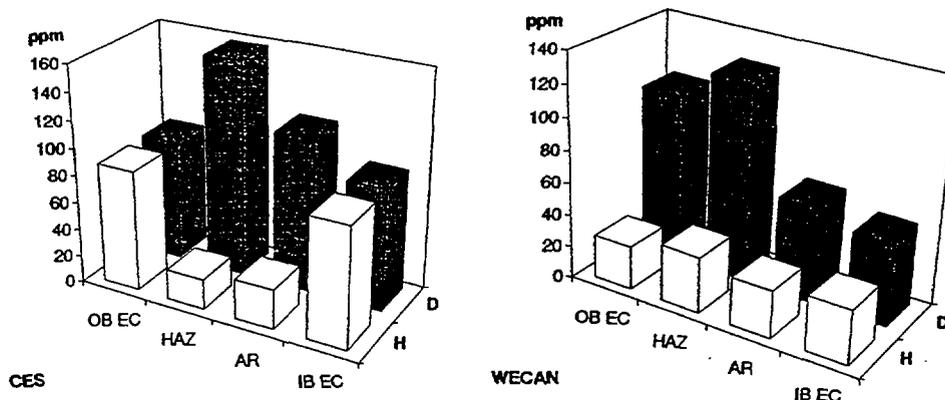


Figure 6 - DISTRIBUTION OF H AND D IN BRUCE CLADDING.

CES and WECAN Fuel, Comparison of End Caps (EC), HAZ and AR (COG).

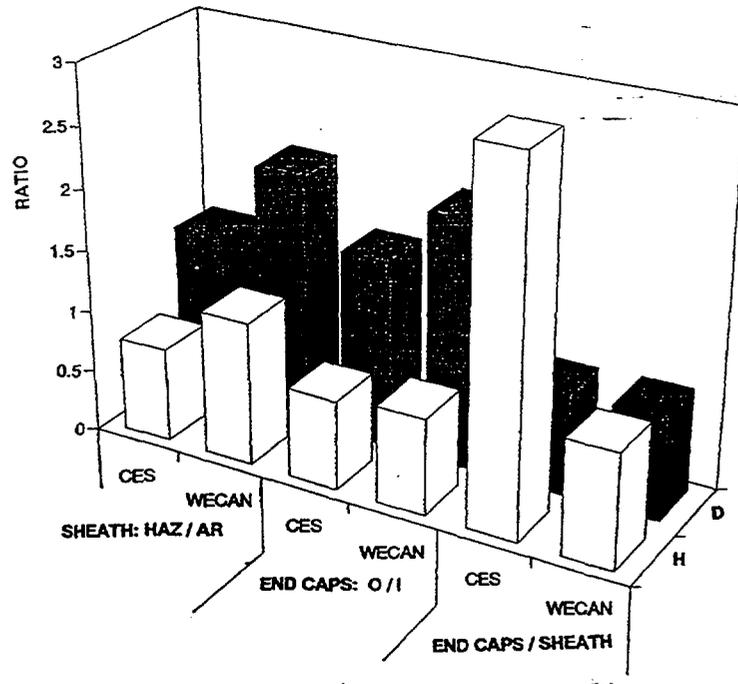


Figure 7 - DISTRIBUTION OF H AND D IN BRUCE FUEL ELEMENTS. Comparison of CES and WECAN Bundles (COG).

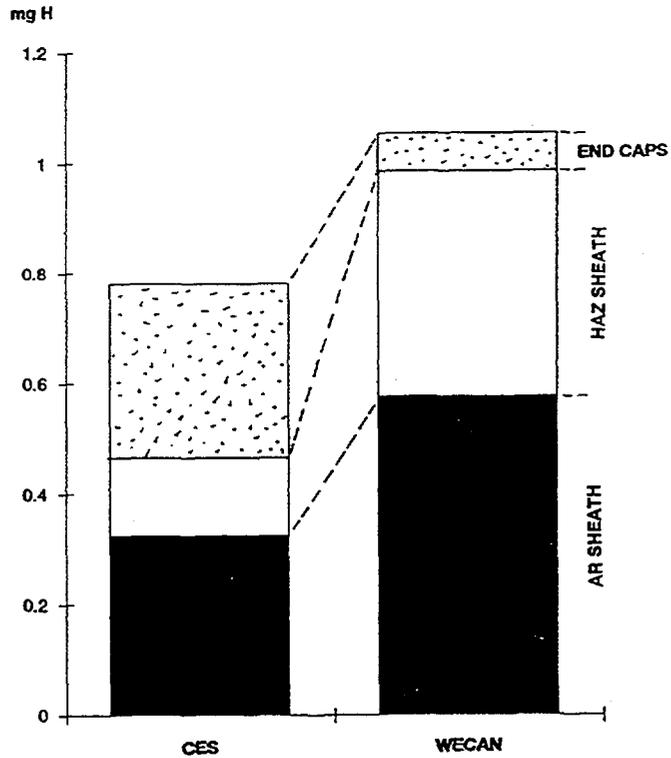


Figure 8 - INCREASES OF H CONTENT IN THE CLADDING OF BRUCE FUEL (COG). (Differences between As-Manufactured and Discharged Intact Fuel Elements.)