Canadian Nuclear Safety Commission | Commission canadienne de sûreté nucléaire

CMD 26-M10.3

Date: 2026-02-23

Written Submission from Dr. Frank Greening

Mémoire de Dr. Frank Greening

In the matter of the

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Status of licensee research and development commitments on elevated hydrogen equivalent concentration in pressure tubes

État des engagements en matière de recherche et développement de titulaires de permis sur les concentrations élevées d'hydrogène équivalent dans les tubes de force

Commission Meeting

Réunion de la Commission

March 2026

Mars 2026

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Canada

From: Frank Greening
Sent: February 23, 2026 8:15 AM
To: Interventions / Interventions (CNSC/CCSN)
Cc:

Subject: Intervention for CNSC Heq Hearing
Attachments:

To whom it may concern,

Please consider this email as an intervention to the public meeting slated for March 23rd, 2026, to discuss the status of Bruce Power and OPG research and development on elevated Heq in operating pressure tubes.

Intervention:
On February 1st, 2023, under control document Ref. 2023-H-103, we have the following announcement by the CNSC:

> *The CNSC is to conduct a Hearing in Writing on application from Bruce Power Inc. to amend the power reactor operating licence for Bruce Nuclear Generating Stations A and B to reflect recent Commission decisions. The CNSC will conduct a hearing based on written submissions to consider an application from Bruce Power Inc. (Bruce Power) to amend the power reactor operating licence for the Bruce Nuclear Generating Stations (NGS) A and B.*
> *The proposed amendment is as follows: to remove licence condition 15.3, Pressure Tube Fracture Toughness, and to include all fitness-for-service requirements applicable to pressure tubes under licence condition 6.1, Fitness for Service.*

The events leading up to this Hearing in Writing are as follows. On July 5, 2021, Bruce Power reported that measurements obtained from a Unit 6 pressure tube after 271,729 hot hours of operation showed Hydrogen Equivalent Concentrations ([H_{eq}]) above the generic predictions and exceeding the Licence Condition 15.3 [H_{eq}] limit of 120 parts per million (ppm – by weight). Bruce Power reported that pressure tube B6S13 has a [H_{eq}] of 211 ppm at the burnish mark and 212 ppm at the burnish mark plus 10 mm. Also, on July 8, 2021, Bruce Power reported that measurements obtained from a Unit 3 pressure tube showed [H_{eq}] above the generic predictions and above the Licence Condition 15.3 [H_{eq}] limit of 120 ppm. For the Unit 3 pressure tube B3F16, Bruce Power indicated a preliminary measurement of 131 ppm [H_{eq}].
Additionally, on March 24th 2022, Bruce Power issued the Event Initial Report CMD 22-M16 stating:

> *Following the July 2021 discovery of elevated Heq near the outlet rolled joint, Bruce Power performed additional surveillance testing on the removed PT B6S13 and discovered that elevated Heq also exists near the inlet end of the PT. The reported Heq level from a through-wall punch sample was 126 ppm at approximately 10 mm in board of the burnish mark.*
> *Bruce Power does not have a mechanistic understanding of the phenomenon nor validated models as a result of this finding. In other words, their Heq model is invalid because the outputs of the Heq models do not align with the B6S13 measurement of 126 ppm at the inlet end of the*

PT. These Heq outputs are used as inputs into Fitness for Service Assessments such as leak-before-break (LBB) and fracture protection (FP) assessments. The uncertainty of the Heq inputs impact the LBB and FP assessments. CNSC staff are of the opinion that Bruce Power cannot confidently perform these assessments until the Heq phenomenon is understood and modelled.

The licence condition for Bruce A & B with respect to pressure tube fitness for service has always been based on two CSA Standards as follows:

CSA Standard N285.4:

12.3.5.2 Acceptance criteria

The determination of H_{eq} shall be considered acceptable when

(a) the predicted concentration value at the end of the next periodic measurement interval is below the level at which hydrides are present at sustained operating conditions; and

(b) the measured/determined rates of change in H_{eq} are less than those defined in the following Table:

Maximum channel outlet temperature	Maximum allowable rate of change in H_{eq} concentration per 10 000 hot operating hours
< 315 °C	3 ppm H_{eq}
< 305 °C	2 ppm H_{eq}
< 295 °C	1 ppm H_{eq}

CSA Standard N285.8:

Table 5
Distribution of maximum allowable hydrogen equivalent concentration
(See Clause 8.2.)

Position relative to pressure tube inlet burnish mark, z_b/L_b*	Maximum allowable hydrogen equivalent concentration, ppm
0.00	70
0.05	70
0.10	71
0.15	72
0.20	72
0.25	74
0.30	75
0.35	77
0.40	80
0.45	82
0.50	85
0.55	88
0.60	91
0.65	93
0.70	95
0.75	96
0.80	98
0.85	99
0.90	99
0.95	100
1.00	100

* *Linear interpolation between tabular values may be used.*

The three fuel channel temperatures listed in Clause 12.3.5.2 of CSA N285.4, shown above, – namely, 315 °C, 305 °C, and 295 °C – correspond to the conditions for Darlington, Bruce and Pickering NGS fuel channel outlets, respectively. Thus, we see that CSA N285.4 sets the maximum allowable rate of increase in H_{eq} per 10,000 hot hours, ($\Delta H_{eq}/10^4$ HH), to be 3 ppm, 2 ppm and 1 ppm, for Darlington, Bruce and Pickering respectively.

In order to investigate if Bruce Power is in non-compliance with the CSA N285.4 Standard, I have reviewed the available data for Heq in Bruce B pressure tubes since the start of their commercial operations in the mid-1980s. The results of this review, and in particular estimates of $\Delta H_{eq}/10^4$ HH as a function of the hot hours of exposure of the pressure tubes, are summarized in Table 1, below.

Table 1: Average $\Delta H_{eq}/10^4$ HH (in ppm) for Bruce Units 5 - 8

Bruce B Units 5 - 8 Average Hot Hours	$\Delta H_{eq}/10^4$ HH (ppm)
50,000	0.4
100,000	1.0
150,000	2.0
200,000	3.4
250,000	4.9
300,000	6.5 [1]

Ref 1 = Extrapolated Value

The data in Table 1 show two important trends:

(i) After approximately 150,000 hot hours of exposure the $\Delta H_{eq}/10^4$ HH values are consistently *above* the Bruce B limit of 2 ppm per 10,000 hot hours

(ii) The *rate* of hydrogen pick up steadily *increases* as the hot hours of exposure increase

CSA Standard N285.8 was revised in 2019 to an Heq of 80 ppm at a pressure tube's inlet and 120 ppm Heq at its outlet. Nevertheless, it is clear that the July 2021 high Heq concentrations measured at the inlet, (126 ppm), and the outlet, (212 ppm), of the B6S13 pressure tube place Bruce Power in violation of the requirements of the revised CSA Standard N285.8.

Thus, we see that Bruce Power is in non-compliance with regard to both the CSA N285.4 and the CSA N285.8 Standards. However, as noted above, the fact that the *rate* of hydrogen pickup is not only well *above* the 2 ppm per 10,000 hot hours licence limit, <u>but is steadily *increasing*</u>, is worrisome and needs to be explained. An important clue as to *why* $\Delta H_{eq}/10^4$ HH is increasing may be found in data on the oxidation kinetics of Bruce pressure tubes derived from a C-13 oxide dating technique I developed in the 1990s. This technique is based on SIMS (Secondary Ion Mass Spectrometry) depth-profiling, as described in the following references:

1. *The Detection and Interpretation of Carbon-13 Isotope Effects in the Oxide Scales of Irradiated Zr-2.5 wt% Nb Pressure Tubes.* OHRD Report 91-93-P, (June 1991).
2. *Post Irradiation Investigations of Corrosion and Deuterium Pickup by Zr-2.5 wt% Nb Alloy Pressure Tubes: Isotope Tracers in Inside Oxides.* OHT Report A-NFC-96-200-P, (December 1996).

The formation of radiogenic C-13 by the O-16(n,α)C-13 nuclear reaction allows a pressure tube's oxidation rate to be determined. C-13 SIMS data are available for a number of Bruce A and Bruce B pressure tubes, as in the examples shown in Figures 1a and 1b, below:

Figure 1a: Pressure Tube B3U11

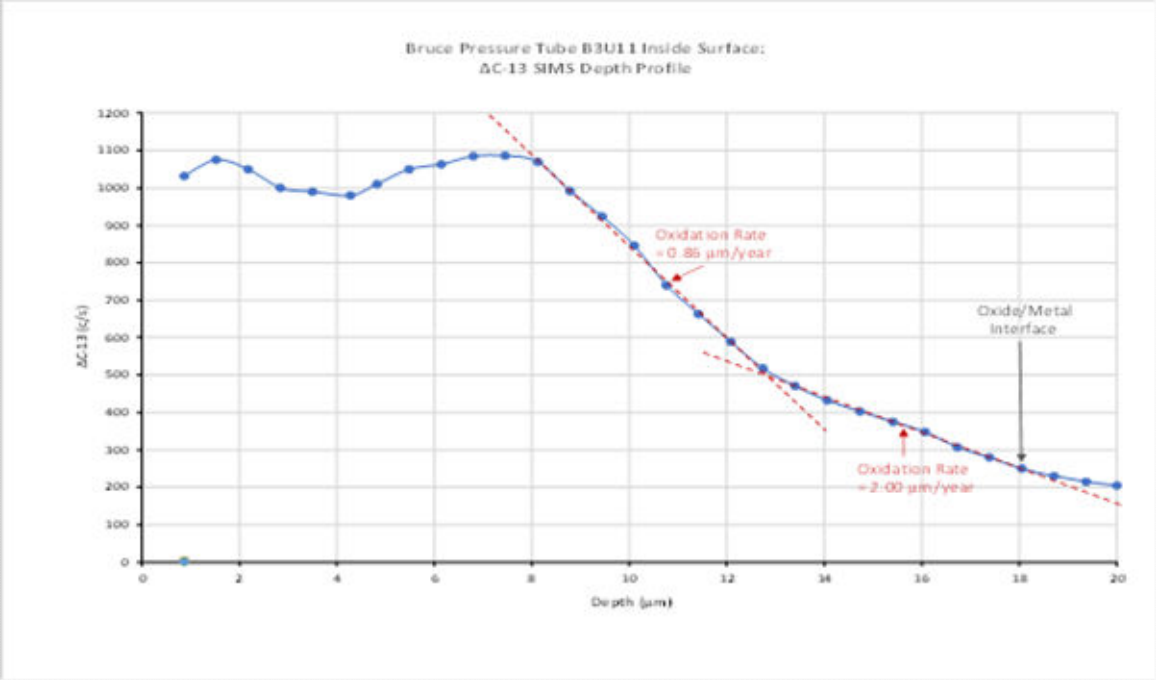
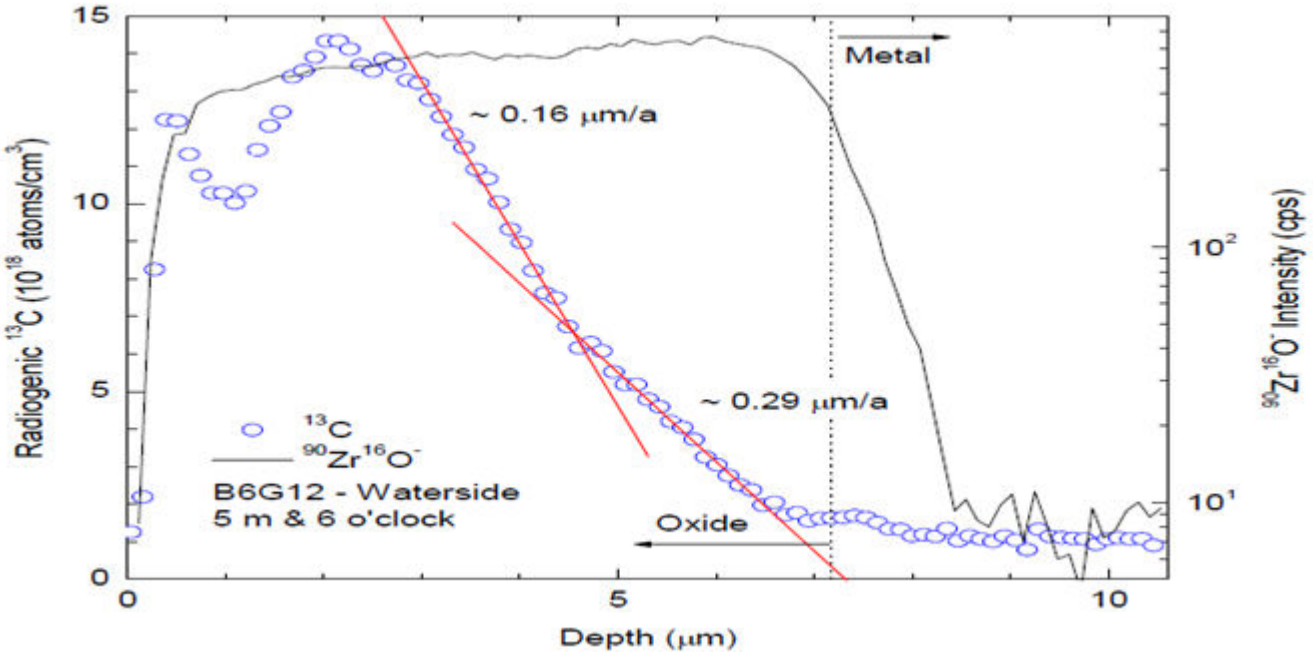


Figure 1b: Pressure Tube B6G12



The C-13 plots in Figures 1a and 1b exhibit the following features for the B3U11 and the B6G12 pressure tubes:

(i) The B3U11 tube's ID oxide is 20 µm thick, compared to only a 7 µm thick oxide for the B6G12 tube

(ii) An approximate doubling of the pressure tubes oxidation rate is observed after about 13 µm of growth for the B3U11 tube, compared to a similar doubling for the B6G12 tube, but after only about 5 µm of growth

These observations show that there is a great deal of variability in the oxidation rate of the inside surfaces of Bruce pressure tubes. However, these data also show that there is a tendency for the oxidation rate to increase at higher hot hours of exposure, and this occurs *regardless of the initial rate of oxide growth*. This acceleration in the rate of oxidation of pressure tubes is very significant because it has implications for the

associated rate of H/D pickup by these tubes. This is because zirconium alloy pressure tubes pick up deuterium during zirconium corrosion in D_2O through the following two-step processes:

$Zr + 2D_2O = ZrO_2 + 2D_2$ ……(1)

$Zr + x/2\ D_2 = ZrD_x$ ……..(2)

It may be shown that the deuterium concentration for 100 % pickup by formation of 1 µm oxide is 14.3 ppm. However, measurements of pressure tube oxide thicknesses together with the associated deuterium concentrations generally show pickups in the range 5 - 15 % of the theoretical maximum.

A predictive model, referred to as the *Design Equation*, for corrosion and deuterium ingress by Zr-2.5Nb pressure tubes, was first developed by AECL in the late 1990s – see for example the 1996 COG Report No. COG-95-596/RC-1551. Specifically for Bruce tubes, the *Design Equation* predicts accelerating oxidation kinetics and accelerating deuterium pick up for exposures of more than about 150,000 hot hours. Hence, it is important to note that the acceleration of H/D pickup by aging pressure tubes was predicted by the Canadian nuclear industry more than 25 years ago. And, the fact that accelerated corrosion and H/D pickup have been a constant feature of CANDU pressure tube aging is confirmed in an article published by Chalk River Nuclear Laboratories – See CNL Nuclear Review Vol 5 (1), June 2016:

> *Along the main body of a pressure tube the deuterium concentration increases and peaks near the outlet end. Approximately 2% - 10% of the deuterium generated by the corrosion process is absorbed. In general, the deuterium concentration in the main body of the pressure tube increases with time. At the 1.5 m axial location the increase is approximately linear with time, whereas at the 4 m and 5 m axial locations the uptake rate is increasing with time.*

My emphasis in red

Nevertheless, it is of interest to see if this view of H/D pickup is currently also held by Bruce Power, especially in light of the discovery of very high Heq in some of its pressure tubes. In the months following the July 2021 reporting of high Heq concentrations in two Bruce pressure tubes, there have been several CNSC Meetings/Hearings to discuss these findings. At the Public Meeting held on September 3rd 2021, Bruce Power was asked by CNSC Commissioner Lacroix for its interpretation of the high $[H_{eq}]$ observed in some of its operating pressure tubes, to which Bruce Power replied:

> "*We're not seeing a change in the rate of hydrogen uptake. What we're seeing is a redistribution (of the hydrogen) to the cooler region at the top of the pressure tube. So, it's not an acceleration but a redistribution*".

Based on the data presented in the first part of this intervention, the fact that Bruce Power still believes the high Heq data reported for some of its pressure tube is <u>not</u> due to accelerated pickup, but is due to a *redistribution* of ingressed deuterium, is fraught with difficulties. And, it is important to note that Bruce Power provides no evidence or proof for its belief that there is no acceleration in H/D pickup by its pressure tubes. It is simply stated as a fact, when it is nothing more than a plausible hypothesis – *and a plausible hypothesis <u>is not necessarily true</u>*.

However, there are plenty of other reasons, including real physical evidence, to accept as a well-established fact that accelerated H/D pickup *<u>is</u>* occurring in Bruce pressure tubes, and has been for some time. For example, please consider:

(i) The data reported for ΔHeq previously shown in Table 1 of this intervention

(ii) A major problem with Bruce Power's redistribution hypothesis may be seen by eliminating the effects of diffusion and calculating the actual amount of H/D entering the B6S13 pressure tube. Thus, using data from Table 2, found in OPG's September 3rd 2021 *Written Submission* to the CNSC, CMD 21-M37.2, we may calculate an *average* concentration of H/D at a distance *x* from a pressure tube's inlet using the H/D concentrations measured at the 6 and 12 o'clock locations as follows:

$[C_{Av}(x)] = \{[C_6(x)] + [C_{12}(x)]\}/2$

> Then, for B6S13, the average H/D concentration near its outlet burnish mark, [C_{Av}(*BM*)], is equal to (211 + 59)/2 or 135 ppm, *which is significantly above the CSA N285.8 limit of 120 ppm*. From this result I conclude that the postulated redistribution of H/D is unable to explain how or why the ***average*** circumferential [Heq] at the burnish mark of tube B6S13 is in excess of the CSA N285.8 limit.

At this point I would say that Bruce Power *must* accept that accelerated H/D pickup is occurring for the B6S13 and B3F16 pressure tubes. However, when Bruce Power states that: "*We're not seeing a change in the rate of hydrogen uptake. What we're seeing is a redistribution (of the hydrogen) to the cooler region at the top of the pressure tube. So, it's not an acceleration but a redistribution*." One needs to ask: What is the relevance of this statement? Because it has no bearing on the fact that Bruce Power was non-compliant with CSA N285.8. following the July 5th, 2021, discovery of elevated Heq concentrations in Unit 6 pressure tubes. Indeed, in a letter from Bruce Power to the CNSC dated July 15th, 2021, – See CNSC document CMD 21-M37.1 – we read, (with my emphasis in red):

> Bruce B Power Reactor Operating Licence, (PROL 18.01/2028), Conditions 6.1 and 15.3, require that Bruce Power, "*…implement and maintain a fitness-for-service program*" and that "***before*** *hydrogen equivalent concentrations exceed 120 ppm,* [Bruce Power] *shall demonstrate that pressure tube fracture toughness will be sufficient for safe operation beyond 120 ppm.*"

However, ***nowhere*** in Bruce B's current PROL, (18.01/2028), or in CSA Standard N285.8 is there a stipulation that exceedances of a Unit's 120 ppm Heq limit are permissible *if they are due to a redistribution of ingressed H/D*. So, again, I must ask why Bruce Power would believe that "*the redistribution of hydrogen*" is an acceptable excuse for a clear licence non-compliance?

In this intervention I have presented a brief history of events and actions following the discovery of high Heq concentrations in several Bruce pressure tubes in July 2021. And I have shown that, as a consequence of these high Heq measurements, Bruce Power is in non-compliance with regard to both the CSA N285.4 and CSA N285.8 Standards. This situation is, as it should be, of great concern to the CNSC because, as noted in the Event Initial Report CMD 22-M16:

> *Bruce Power does not have a mechanistic understanding of the phenomenon nor validated models as a result of this finding. In other words, their Heq model is invalid because the outputs of the Heq models do not align with the B6S13 measurement of 126 ppm at the inlet end of the PT. These Heq outputs are used as inputs into Fitness for Service Assessments such as leak-before-break (LBB) and fracture protection (FP) assessments. The uncertainty of the Heq inputs impact the LBB and FP assessments. CNSC staff are of the opinion that Bruce Power cannot confidently perform these assessments until the Heq phenomenon is understood and modelled.*

Unfortunately, OPG and Bruce Power are, by their own admission, unable to provide a new and improved model of hydrogen pickup by Zr-2.5%Nb pressure tubes, but acknowledge that the research required to validate a new fracture toughness model of pressure tube aging is "a work in progress" that will not be completed for years to come. And when I say "a work in progress," the record shows that problems stemming from discrepancies between Heq predictions and Heq measurements may be traced back many years, as shown by the examples below.

1. In 2013, the CNSC asked OPG to establish a fracture toughness model "of upper transition and lower shelf behavior" for Heq > 100 ppm. And in July 2014 the CNSC issued a Bid Solicitation for the provision of a "R565.1 Report" on the fracture toughness properties of Zr-2.5Nb pressure tube material with high hydrogen concentration. The contract was for $120,000 over two years and was awarded to Areva Canada in December 2014. The contract included the following description of the research project:

Determine parameters describing the fracture toughness of pressure tube material with high hydrogen concentrations. Develop a methodology or a model to predict fracture toughness of pressure tube

material with elevated hydrogen concentrations (50 ppm and 120 ppm [H]eq). Validate the predictions against experimental results derived from a parallel experimental program commissioned by the CNSC.

2. In June 2015, OPG provided the following "update" on this issue:

Based on R&D work, ... a new fracture toughness model has been developed and is being integrated into the 2015 edition of CSA N285.8 Standard and will become part of the Licence Condition Handbook for Darlington.

However, in Darlington's 2016 Licence Condition Handbook, we read:
CNSC staff accepted (e-Doc 4272552 and 4369355) OPG's approach (e-Doc 4250561) to fitness for service assessment, regarding the application of new fracture toughness models and probabilistic Leak-Before-Break (LBB) assessments for pressure tubes at increased hydrogen content. OPG has planned continuing research & development to further validate and improve the fracture toughness models (e-Doc 4405852).
In the probabilistic core assessments for flaw, Leak-Before-Break and Pressure Tube to Calandria Tube contact, OPG should provide a comparison of the derived 95 % upper bound PT failure frequency with the criterion identified in "Table C.1 of CSA N285.8-15" and "Table 3 of COG-JP-4363-V078-R02". CNSC staff will compare the 95% upper bound of the calculated PT failure frequency with Table 3 of COG-JP-4363-V078 R02, ***until such time*** *as the CSA N285.8 committee completes its review of the application of Table C. l of the CSA N285.8-15.*
My emphasis in red

3. In August 2021, we have OPG's Progress Report to the CNSC on the status of its new fracture toughness model:

[Heq] MODELLING ENHANCEMENTS
[Heq] modelling enhancements including use of 3D Finite Element Analysis considering fuel channel geometries, local temperatures, location-specific [Heq], and material stress states are being pursued. Note that these proactive enhancements were already in progress prior to the B3 and B6S13 findings. OPG, with industry alignment, intends to submit modelling enhancements for CNSC acceptance ***once fully validated****.*
My emphasis in red

4. In an email from the CNSC I received on January 10th, 2023 we read:

The External Advisory Committee (EAC) will consider all of the relevant information that has been gathered to date, including your written intervention CMD-M37.4 and other submissions. The EAC's report, and the Commission's consideration of that report, will be transparent and public, in accordance with the Commission's commitment to transparency. ***We expect the EAC report to be submitted in Spring or early Summer 2023****.*

High Heq concentrations, well in excess of Bruce Power's operating license limits, were first seen in Bruce Unit 6 pressure tubes in July 2021. By way of dealing with this problem the CNSC ordered Bruce Power to conduct a detailed assessment of the safe continued operability of Bruce Units 1, 2, 4, 5, 7 and 8. Bruce Power was also told, given the discrepancy between the current fracture toughness model predictions and inspection results for Units 3 and 6, to assess whether operation of these Units remains within the licensing basis. At the same time, the CNSC stressed the need for Bruce Power to carry out a **root cause analysis** to determine if the measured elevated hydrogen concentration was caused by a new phenomenon specific to these Units and their pressure tubes.
At its September 3rd, 2021, meeting, the CNSC promised there would be: "*a follow-up public Commission meeting on this topic in late Winter 2021 or early Spring 2022*". However, by July 2022, one year after the discovery of high Heq in some Bruce pressure tubes, no public Commission meeting on high Heq issues had taken place. In fact, the promised meeting was eventually held in early November 2022, *one year and 4 months after the high Heq problem was first recognized*. Now, this delay might be considered acceptable, if the November 2022 meeting did in fact provide a root cause analysis of the high Heq concentrations, but, regrettably, this was not the case.

As noted above, the CNSC stated in a January 10th, 2023, email: "*We expect the EAC report to be submitted in Spring or early Summer 2023*"; with such a timeline, the EAC's report would be issued *a full two years on from the first observation of high Heq at Bruce*. This leads me to conclude that the CNSC is in no hurry to find a root cause of the high Heq problem, but is apparently quite content to let CANDU reactors operate with deuterium ingress rates running well above the predictions of existing [Heq] models.

Interestingly, it appears that the Canadian nuclear industry is like-minded in this regard, which may be seen in the timeline proposed to develop a new and improved [Heq] model, as first reported at an industry workshop on elevated hydrogen equivalent ([H]eq) concentration held on March 25th, 2022. In an account of this meeting, published by Bruce Power in July 2022, we find Attachment B which provides "*a path forward with a target schedule and summary of key deliverables to improve hydrogen equivalent concentration predictions in the inlet/outlet rolled joint regions of pressure tubes*". Action Item 5G in Attachment B, described as "The Development of a Comprehensive [Heq] Model", gives the target date for the completion and issuance of a "comprehensive [Heq] Model" as **the 2nd Quarter of 2026**.

This target date means that the Canadian nuclear industry would have taken ***5 years*** to develop a new and improved [Heq] model as a response to the discovery of high Heq concentrations in some Bruce tubes in July 2021. However, it is important to note that the CNSC issued a bid solicitation document, No. 87055-18-0098, in November 2018 for an independent contractor to:

> "*Predict the fracture toughness of pressure tubes with elevated hydrogen concentrations (40 ppm to 160 ppm [H]eq), and validate the modelling predictions against the experimental results provided by CNSC staff*".

And what is most significant about this *Request for Proposal* document is that the CNSC stipulated that the requested work must be completed within ***fifteen months*** of the awarding of the contract, with a final report issued by March 2020.

5. The real situation with regard to the quest for a new and improved fracture toughness model for CANDU pressure tubes is well illustrated by the current status of one of the Standards on which the model is based – namely CSA N285.8. Thus, as described in clause C.5 of the latest update of CSA N285.8, we read, (with my emphasis in red italics):

C.5 Determination of statistically significant changes in pressure tube properties

C.5.1 General

Clause C.5 describes the method for determining whether statistical evidence exists signifying a change in pressure tube material property information.

C.5.2 Evaluation of hydrogen equivalent concentration

This evaluation procedure is under development. The evaluation procedure shall be justified and may be used provided that the procedure has been accepted by the regulatory authority.

C.5.3 Evaluation of fracture toughness

This evaluation procedure is under development. The evaluation procedure shall be justified and may be used provided that the procedure has been accepted by the regulatory authority.

C.5.4 Evaluation of DHC growth rate

This evaluation procedure is under development. The evaluation procedure shall be justified and may be used provided that the procedure has been accepted by the regulatory authority.

C.5.5 Evaluation of threshold stress intensity factor for delayed hydride cracking

This evaluation procedure is under development. The evaluation procedure shall be justified and may be used provided that the procedure has been accepted by the regulatory authority.

Clearly, these five clauses show that the key determinants of CSA Standard N285.8 are all "*under development*," and evidence already presented in this intervention suggests that this uncertainty in one of the key Standards governing the fitness for service of pressure tubes *will remain unresolved for years to come*. Nevertheless, the CNSC claims that it provides "*Comprehensive and Effective Regulatory Oversight of pressure tube degradation mechanisms*." And, according to a CNSC presentation from a January 2018 Commission Meeting, this is achieved by setting goals and guidelines for reactor operators which include:

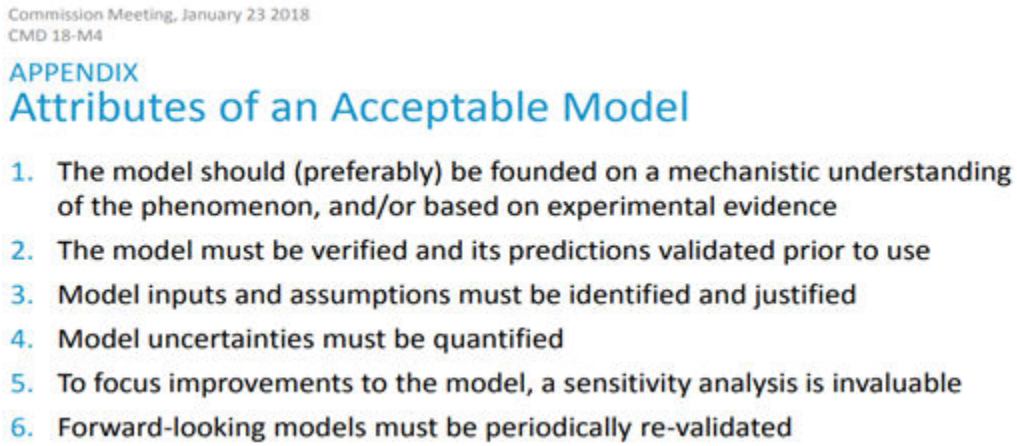
(i) Clear and well-documented expectations and Licence Compliance Plans through REGDOC-2.6.3 and CSA Standard N285.8

(ii) Effective Compliance Verification Criteria as presented in the Licence Condition Handbooks for a particular NGS

(iii) Licensees must have an in-depth understanding of pressure tube degradation phenomena

The Compliance Verification Criteria for Bruce Units may be found in Section 15.3 of Licence Conditions Handbook, LCH-PR-18.01/2028-R002. Through this license condition, Bruce Power is required to submit to the CNSC a fracture toughness model for review and acceptance and no Bruce Unit is authorized to operate above the 120 ppm [Heq] limit set in Clause 8.2 of CSA Standard N285.8. In addition, through a fracture toughness model, Bruce Power must verify that measured Heq changes between inspection periods are bounded by Heq concentrations predicted by an acceptable model.

But what constitutes an acceptable model? This question has been addressed by the CNSC, as shown by the overhead presented below taken from a January 2018 Commission Meeting:



Of the six attributes listed above, No. 2 – "*The model must be verified and its predictions validated prior to use*" – is perhaps the most important, but is also the most problematic. This is because the required predictive modeling involves making estimates of *future* pressure tube performance parameters, such as hydrogen pickup rates and fracture toughness values; however, because we are dealing with *future* expectations, it follows that appropriate experimental data are obviously not yet available from real-world pressure tubes. And since it is not possible to verify or validate a model without the requisite experimental data, one has to ask how *any* fracture toughness model could ever be "*verified and validated prior to use*".

An issue that further complicates the validation of fracture toughness models is the fact that the fracture toughness of a pressure tube depends on many factors, including the [Heq], temperature, neutron fluence, trace impurities, etc., of the tube in question This leads to a large variability in measured values of fracture toughness parameters such as a pressure tube's crack growth resistance, dJ/da, as shown in the Figure below:

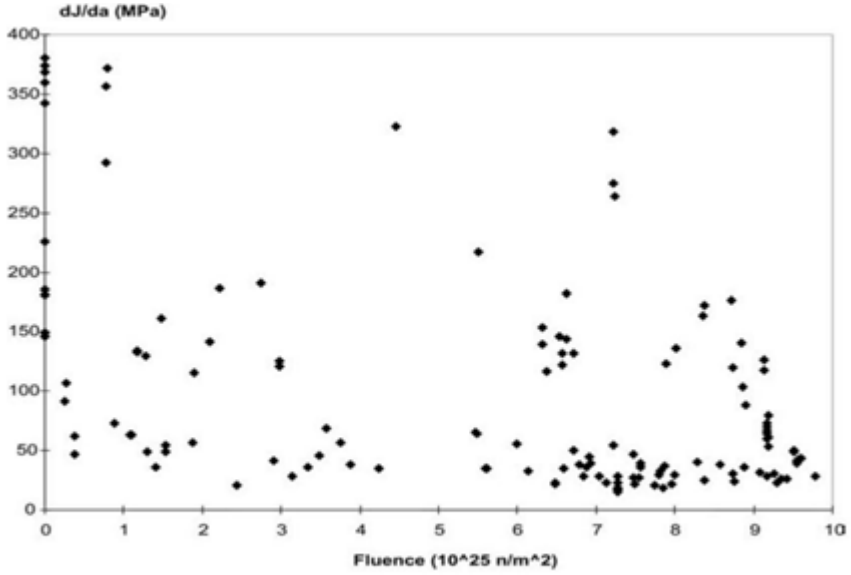


Figure 2.9: The fracture toughness of cold worked Zr-2.5Nb pressure tubes at 240-300 $^{\circ}C$ after service in a CANDU reactor showing large variability

This extreme variability in a tube's fracture toughness – a variability that is also seen in many other pressure tube properties such as H/D pickup rates – means that predictive models themselves are inherently uncertain and can only offer upper bounds, for H/D pickups, or lower bounds, for fracture toughness parameters. However, in addition to the model predictions provided annually by a reactor operator, the operator is also obligated to obtain scrape measurements during each planned outage to determine the level of [Heq] in sampled pressure tubes. These measurements serve as a means of comparison to model predictions and allow a reactor operator to determine if the validity limits of the fracture toughness model are satisfied. It follows that the predictions need only go as far as the next scheduled inspection – a time interval that is typically about 2 to 3 years.

Until the discovery of very high Heq concentrations in several Bruce pressure tubes in July 2021, the Canadian nuclear industry, and the CNSC, stated at many Public Hearings that Canadian reactors are operating in full compliance with the fracture toughness model described in CSA Standard N285.8-2015 Update 1, Clause D.13.2.3.1.2 (a). And such a claim was made by all parties concerned in spite of the fact, as shown above, that the required validation and verification of the 2015 fracture toughness model has ***not*** been accomplished. Of course, such an approach may appear to work as long as pressure tube corrosion and H/D uptake rates remain relatively constant. But this approach is doomed to failure if a tube is subject to the rapid onset of accelerated corrosion, which appears to be the case for Bruce pressure tubes B6S13 and B3F16.

Before completing this intervention, I wish to address an important issue that, (I believe), has been inadequately dealt with by our nuclear operators and regulators alike – namely the link between the concentrations of H/D in a pressure tube and the tube's fracture toughness.

The operational limits on a pressure tube's fracture toughness are specified in Clause 8.3 of CSA Standard N285.8-15 as shown below:

8.3 Evaluation of fracture toughness

The owner/operator shall satisfy the following technical requirements:

a) A representative statistical model for fracture toughness shall be established on the basis of available fracture toughness data, including the surveillance measurements under evaluation. The procedure provided in Clause C.5 may be used for model derivation. The representative model shall be used in the evaluations in Items (b) to (d).

b) The probability of observing a fracture toughness value below the reference lower-bound fracture toughness shall be estimated. The relations in Clause D.13 may be used as the reference lower-bound. The procedure provided in Clause C.5 may be used to estimate the probability.

c) When the estimated probability in Item (b) does not exceed the threshold probability P_{TH} in Table 7, the evaluation requirements for fracture toughness measurements are satisfied.

d) When the estimated probability in Item (b) exceeds the threshold probability P_{TH} in Table 7, the reference lower-bound fracture toughness shall be revised. The probability of observing a fracture toughness value below the revised lower-bound fracture toughness shall not exceed the allowable probability P_A in Table 7. The procedure provided in Clause C.5 may be used for revision. In accordance with Clause 4.5.1.3, the validity of assessments that have been performed previously to satisfy the requirements of Clauses 7.2 to 7.4 shall be evaluated using the revised representative fracture toughness model.

Here we see CSA Standard N285.8-15 imposing the license requirement that a reactor owner/operator shall establish a fracture toughness model "*on the basis of available fracture toughness data, including surveillance measurements under evaluation*." However, this apparently straightforward license requirement is actually quite complex and problematic because, as previously noted in this intervention, the fracture toughness of a pressure tube depends on a number of variables including the temperature, neutron fluence and Heq concentration, the effects of which need to be included in any meaningful evaluation.

Furthermore, in order to replicate a pressure tube's behavior as it approaches end-of-life, pressure tube data for samples with fast neutron fluences up to 2×10^{26} n/m^2, (E>1MeV), and Heq concentrations in excess of 100 mg/kg, are required. Unfortunately, the availability of such data for real-world pressure tube material is very limited.

CSA Standard N285.8-15 also includes a Table 7 which stipulates that a pressure tube's fracture toughness, K_c, should be evaluated "*on a case-by-case basis*". However, Table 7 also introduces the additional parameters V_a and V_r, the DHC axial and radial crack growth rates, respectively, and Clause 8.4 of CSA Standard N285.8 stipulates that the owner/operator shall establish a statistical model based on available delayed hydride cracking growth rate data. Table 7 sets limits on V_a and V_r in the form of threshold probabilities of observing a fracture toughness value below a reference lower-bound fracture toughness.

Interestingly, the methodology used to quantify DHC crack growth rates in CSA Standard N285.8 has varied considerably over time. Thus, for example, the 2005 model considers a Unit's operating temperature only, but the 2015 model also includes the neutron fluence – a parameter that depends on the axial position of a pressure tube. This means that the 2005 model assumes one DHC crack growth rate while, for the 2015 model, the DHC growth rate is different at the inlet and outlet locations. Furthermore, in the case of a probabilistic assessment, the 2005 fracture toughness model does not consider the known hydrogen concentration variations, while the 2015 fracture toughness model is segmented into different calculation regions according to the pressure tube's hydrogen content and temperature range. Thus, it is not surprising the 2005 and 2015 models give different answers to the same question.

CSA Standard N285.8 also runs into problems with regard to the methodology it recommends for a so-called Leak Before Break, (LBB), analysis. An LBB analysis is intended to evaluate the response of an annulus gas system to a leak, and specifies the required operator actions in the event of a pressure tube through-wall crack.

The concept of leak-before-break, (LBB), is an operational requirement that, in the event of a pressure tube leak from a through-wall crack, there will be sufficient time for the leak to be detected and the reactor shut down *before* the crack grows to the critical size for a fast-uncontrolled rupture. Thus, if a pressure tube develops a through-wall crack, it is assumed that there will always be a "window of opportunity" for the reactor operator to detect the leak and safely shut down the Unit. This assumption is the basis for the claim

that CANDU reactors safety systems provide "*defense in depth*," a notion that is often touted by the Canadian nuclear industry – but is such a claim justified?

The practical implementation of LBB is described in Section C.4 of CSA Standard N285.8: *Technical requirements for in-service evaluation of zirconium alloy pressure tubes in CANDU reactors*. Clause C.4.1 of this Standard stipulates that:

> *LBB analysis shall demonstrate that the leak detection capability of the annulus gas system (AGS) provides the operator with sufficient warning time to shut down and depressurize the reactor in a controlled manner.*

Similarly, Clause C.4.2.2.6 of the Standard states that:

> *To perform a LBB analysis, the response of the AGS and operator to indications of leakage from a through-wall DHC crack must be defined….. The time needed to detect, confirm, and locate the leak varies from station to station and depends on the leak rate, and therefore is an important input into the LBB analysis.*

There are four key parameters that need to be evaluated in a LBB analysis:

(i) *The initial crack length at wall penetration*, L_p. This is typically in the range 20 – 30 mm.

(ii) *The location of crack initiation*. Cracks may originate anywhere in the body of a pressure tube, but are most likely to occur in the vicinity of the rolled joint between a pressure tube and its end-fitting.

(iii) *The axial crack growth rate*, V_a, which is usually expressed as a velocity, in m/s, and is typically in the range 1×10^{-7} to 2×10^{-6} m/s. As previously noted in this intervention, V_a depends on the temperature, irradiation history and hydrogen equivalent concentration in the pressure tube at the crack location.

(iv) *The critical crack length*, CCL. This refers to the point in the development of a crack in the *axial* direction of a pressure tube at which a slowly increasing crack, (e.g. ~ 2 mm/hr), accelerates to a fast rupture (e.g. to a crack velocity > 2 mm/s). The CCL of a pressure tube is dependent on the temperature and hoop stress at the crack location but is typically in the range 30 – 80 mm.

A CANDU station's operating procedures require immediate shutdown of a Unit at a confirmed D_2O leakage rate of 0.5 kg/s. It is therefore important to closely monitor D_2O leaks and it follows that a LBB analysis of a pressure tube should include an estimate of potential D_2O leak rates as a function of crack length. One approach that has been used to make such estimates is to collect data from tests on removed pressure tubes and look for a correlation between D_2O leak rate and crack length.

Unfortunately, data from hot tests on removed pressure tubes show considerable scatter – for example, tubes with crack lengths in the range 18 – 30 mm exhibit D_2O leak rates in the range 0.8 to 30 kg/hr. Nevertheless, in spite of these large variabilities, the approach used at CANDU stations is to simply average the available data. Using this approach, *typical* pressure tube LBB behavior, as presented in a station's AGS Design Manual, is as follows:

1. With a reactor at full power, a crack will penetrate a pressure tube wall at a crack length of 27 mm and grow at a velocity ~ 5.3×10^{-7} m/s, equivalent to 1.94 mm/hr.
2. After 0.5 hours, the crack length would extend to 28 mm and the leak rate would be 0.6 kg/hr.
3. After 1.5 hours, the crack length would be 30 mm and the leak rate would have increased to 1.79 kg/hr.
4. The Unit's AGS leak detection capability is expected to recognize such a leak within this time window, (i.e, ~ 2 hrs).

However, this approach is quite different to the methodology used in CSA N285.8 which recommends the following *cubic* leak rate equation for LBB analysis:

$Q = -11.2 + 0.0014(2C)^3$

Q = D_2O leak rate in kg/hr

2C = crack length in mm

For crack growth at a velocity ~ 5×10^{-7} m/s, this equation predicts a D_2O leak rate of 4 kg/hr after 0.5 hr and 15 kg/hr after 1.5 hrs. These values are more than 6 times higher than the AGS Design Manual values noted above. Nevertheless, regardless of the precise leak rate that occurs after a crack extends through the wall of a pressure tube and starts to leak, it is expected that D_2O will enter the AGS at a rate of at least 1 kg/hr within the first hour after leak initiation.

However, it is important to note that neither a station's AGS Design Manual nor the CSA Standard N285.8 have anything to say about the ***methodology*** to be used, or how well it should perform, for D_2O leak detection. Similarly, although Fitness for Service Guidelines for a CANDU power reactor do require the operator to establish a pressure tube leak detection capability that is active at all times during reactor operation, ***no leak detection methodology is specified***.

A theoretical basis for the LBB methodology as currently applied to CANDU pressure tubes is provided by the equation:

$$t = CCL - Lp2Va \quad \text{.....(i)}$$

Where:

t is the time for a crack to propagate to a critical length for fast rupture
CCL is the critical crack length
L_p is the crack length when it penetrates through the pressure tube wall
V_a is the axial crack velocity

Equation (i) has *three* unknown variables – CCL, L_p and V_a – and the assignment of meaningful values to these parameters has proven to be quite difficult because of the tremendous scatter in the available experimental data. Thus, for example, we have the values of CCL quoted by Cheadle et al. in "*Operating Performance of CANDU Pressure Tubes*", AECL Report No. AECL-9939, (April, 1989). The CCL values are for Zr-2.5% Nb exposed to 240 – 300 °C D_2O and fast neutron irradiations up to 8×10^{25} n/m^2 and span the range from 40 to 90 mm. Similarly, the same report provides values for V_a in the temperature range 150 – 280 °C. These data show that V_a is strongly dependent on the pressure tube temperature, but even for a *fixed* temperature such as 250 °C, the values of V_a span the range 1×10^{-7} to 6×10^{-7} m/s, (equal to 0.36 to 2.16 mm/hr).

More recent research has confirmed most of these values for CCL and V_a. For example, consider the report by D. Rogers et al: "*Performance of Pressure Tubes in CANDU Reactors*," CNL Nuclear Review Vol 5, (1), pp 1 – 15, November 2015. Here we read: "*For a test temperature of 250 °C, the CCL ranges from a minimum of ≈ 41 mm to a maximum of >80 mm*". In addition, Rogers et al's 2015 publication includes a plot of a pressure tube's 95% upper bound mean, and lower bound crack growth velocity, as a function of temperature. At 250 °C the data span the range 1×10^{-7} to 4×10^{-7} m/s or 0.36 to 1.44 mm/hr which is similar to Cheadle's 1989 estimate noted above.

The value of L_p, the crack length when it first penetrates the pressure tube wall and D_2O starts leaking into the annulus gas system (AGS), is also subject to great uncertainty. It was initially assumed that the upper bound on L_p would be 4W, where W is the wall thickness of a pressure tube, (which is ≈ 4 mm). However, it was subsequently realized that a pressure tube crack may tunnel so that the length of a crack at wall penetration may be considerably larger than 4W – See report by Moan et al: "*Leak Before Break Experience in CANDU Reactors.*" AECL Report No. AECL-9609, issued April 1988.

Data collected from measurements of pressure tubes removed from Pickering and Bruce has shown that 27 mm is the longest initial crack opening size observed to date. For this reason, the initial crack length, L_p, for pressure tube LBB assessments is often conservatively assumed to be 27 mm. Using these bounding values for the parameters in question we then have:

$t = CCL - Lp2Va$

$t = (40 - 27)/(2 \times 1.44)$ hours

t = 4.5 hours

It is very telling that estimates of the time it takes for a pressure tube crack to reach its critical crack length have varied considerably since the LLB approach to CANDU pressure tube fitness for service assessments was

first introduced in the 1970s. Thus a 1988 review by E.G. Price et al. entitled *Leak Before Break Experience in CANDU Reactors* asserted that "*the time available for operator response is about 100 hours*." Remarkably, just two years later, the same authors reduced this estimate to a mere 18 hours in a paper published in the International Journal of Pressure Vessels and Piping in 1990.

However, by 1995, a Korean Atomic Energy Research Institute report entitled "*Safety Margin Improvement Against Failure of Zr-2.5Nb Pressure Tubes*", stated that the time for operator action in the event of a DHC-induced pressure tube rupture is 11.7 hours. But ten years later, a 2005 report from the same Korean Institute concluded: "*The time for the operator to take action against a LOCA is 1.7 hours*.

More details on the problems associated with the LBB methodology described in CSA Standard CSA N285.8 may be found in the journal article: *CANDU Pressure Tube Leak Detection by Annulus Gas Dew Point Measurement: A Critical Review*. Kerntechnik Volume 82, pages 1 -15, (2017).

Summary of Bruce Power's Position on High [Heq] Issues:

1. On July 5, 2021, Bruce Power reported that measurements obtained from a Unit 6 pressure tube after 271,729 hot hours of operation showed Hydrogen Equivalent Concentrations ([H_{eq}]) above the generic predictions and exceeding the Licence Condition 15.3 [H_{eq}] limit of 120 parts per million (ppm – by weight). Bruce Power reported that pressure tube B6S13 has a [H_{eq}] of 211 ppm at the burnish mark and 212 ppm at the burnish mark plus 10mm. Also, on July 8, 2021, Bruce Power reported that measurements obtained from a Unit 3 pressure tube showed [H_{eq}] above the generic predictions and above the Licence Condition 15.3 [H_{eq}] limit of 120 ppm. For the Unit 3 pressure tube B3F16, Bruce Power indicated a preliminary measurement of 131 ppm [H_{eq}].

2. On September 10th, 2021 CNSC held a Public Hearing where Bruce Power made the following three comments with reference to the high [Heq] data reported in July 2021:

> **MR. NEWMAN:** *For the record, Gary Newman. Chief Engineer and Senior Vice President of Engineering at Bruce Power:*
>
> **(i)**. *High hydrogen concentrations alone do not impact pressure tube integrity. This is why we evaluate, the combination of both hydrogen concentration and flaws. There are no flaws in the region of interest where the elevated hydrogen concentrations are observed on any Bruce unit. This means there is no driver for crack initiation. The higher hydrogen concentrations at the top of the tube is caused by hydrogen redistribution due to a temperature gradient at outlet ends. This is not an overall increase in the amount of hydrogen in the pressure tube, but rather a redistribution to the region of interest*
>
> **(ii).** *We do not see an incremental change in the rate of deuterium pick-up, and that's reflected in the modelling and bounding nature of the deuterium predictions we're doing in the largest part of the pressure tube. What we are seeing, though, where that model doesn't work and we've had to go to, now, a new model and a two-dimensional treatment is that we're getting a redistribution of H/D. But it's not a change in the corrosion rate or the pick-up rate; it's actually just -- it's just a redistribution of the hydrogen that's already there.*

3. In March 2022 Bruce Power submitted an Initial Event Report, CMD 22-M16, which informed the CNSC that *inlet rolled joint*, (IRJ), punch samples from pressure tube B6S13 showed an elevated [Heq] concentration of 126 ppm localized at ~ 10 mm inboard of the burnish mark (BM). Metallographic examinations on these IRJ punch samples showed a significant radial gradient in hydride concentration decreasing from the pressure tube outer diameter (OD) to inner diameter (ID). The radial gradient was confirmed by direct [H] and [D] measurement of radial sections of B6S13 IRJ. Bruce Power acknowledged that the root cause and impact of this discovery on the fitness for service of Bruce pressure tubes remains undetermined.

5. On October 11th, 2022 Bruce Power applied for an Amendment to its Power Reactor Operating Licence. In its application, Bruce Power is seeking to remove licence condition 15.3, which states that "Before hydrogen equivalent concentrations ([Heq]) exceed 120 ppm, the licensee shall demonstrate that pressure tube fracture toughness will be sufficient for safe operation beyond 120 ppm." Bruce

Power is, instead, proposing a Licence Amendment that all fitness-for-service requirements related to pressure tubes be incorporated into the existing Licence condition 6.1, which states simply that "The licensee shall implement and maintain a fitness for service program."

Discussion:

In July 2021, very high [H_{eq}] levels were measured in pressure tube samples removed from Bruce Units 3 and 6. At a CNSC Public Meeting held on September 3rd, 2021, to discuss this finding, Bruce Power was asked by Commissioner Lacroix for its interpretation of this observation, to which Bruce Power Chief Engineer, Gary Newman, replied:

> "*We're not seeing a change in the rate of hydrogen uptake. What we're seeing is a redistribution (of the hydrogen) to the cooler region at the top of the pressure tube. So, it's not an acceleration but a redistribution*".

Here we see Bruce Power proposing that the very high [H_{eq}] levels measured in a number of pressure tubes from Bruce Units 3 and 6 are simply due to a *redistribution* of ingressed H/D and that this ingress does not represent *acceleration* in the H/D pickup rate. However, in order to validate the assertion that there is no accelerated H/D ingress occurring in Bruce pressure tubes, we need to look for evidence of this in the available data. With this in mind, I have collected recent data reported for Bruce Unit 3 pressure tubes near the 12 o'clock location as shown in Table 5, below. The Table includes values for the initial hydrogen in the pressure tube ingot, [H_{Init}], which is needed to make a small correction to the hydrogen pickup data used in calculating the ratio {[$H]_{Cor}$ /0.5[D]}, where:

$[H]_{Cor} = [H] - [H_{Init}]$

Table 5: Hydrogen and Deuterium Concentrations Near the Outlet Rolled Joints in Selected Pressure Tubes from Bruce Unit 3

Pressure Tube ID (~10 mm from ORJ [Ref 1])	[D] (mg/kg)	[H] (mg/kg)	[H_{Init}] (mg/kg)	Ratio {[$H]_{Cor}$ /0.5[D]}
F16	1340	111	12.6	0.147
L11	790	60	9.3	0.128
G15	853	103	11.3	0.181
K10	1016	74	7.4	0.156
Q16	853	96	10.9	0.233
H06	730	66	15.0	0.329
X09	475	73	14.9	0.245
O20	921	96	12.3	0.182
Q12	850	99	10.2	0.209
N04	337	58	14.7	0.257
O15	303	56	8.7	0.312
O17	122	28	9.0	0.311
O13	443	72	12.5	0.269
P14	191	34	9.0	0.262
Q13	582	87	11.7	0.259
L12	156	27	6.7	0.260
F05	75	25	11.7	0.355
L22	42	24	9.3	–
R10	199	26	5.5	0.206
S13	314	38	6.0	0.204

Ref 1: ORJ = Outlet Rolled Joint

Inspection of the data in Table 5 shows that there is a great deal of variability in the deuterium concentrations in these samples: from a high of 1340 mg/kg for sample F16, to a low of 42 mg/kg in sample L22. In a few cases, such as sample X09, the pressure tube outlet temperatures are known to be somewhat lower, (~ 290 °C), than the temperatures for samples from high power channels such as L12, (~ 297 °C); however, the D pickups for these samples are *the reverse* of what might be expected based in these temperatures alone; thus, the D-pickup is 475 mg/kg for X09, but only 156 mg/kg for L12.

As noted above, Table 5 includes values of the $[H]_{Cor}/\{0.5\times[D]\}$ ratio, where $[H]_{Cor}$ represents the *corrected* hydrogen concentration, derived by subtracting $[H_{Init}]$ from the measured [H], and the factor of 0.5 in the denominator is needed to convert the ratios to *atom ratios*. What is noteworthy about these values is that the atom ratios are relatively constant, averaging 0.24 ± 0.08, even though the tube-to-tube values of $[H_{eq}]$ vary by a factor of more than 20.

The most important question concerning the data in Table 5 relates to the notion of "redistribution" of ingressed H/D. This phenomenon has been proposed by Bruce Power to explain the high H/D levels observed in some B3 and B6 tubes analyzed in July 2021. However, the suggestion that ingressed H/D could *redistribute* within the wall of a pressure tube is not new. Thus, in a COG Report issued in 1998, we read in an Appendix C entitled: "*Influence of Temperature & Concentration Gradients on the Redistribution of Hydrogen Isotopes*":

> *Based on a thermal hydraulic simulation, temperature differences between the 6 and 12 o'clock locations in a tube with 2% diametral creep have been calculated to be 20 °C. Modeling results, assuming a 20 °C temperature difference, show that the deuterium concentration may be up to ~ 17% higher at the top compared to the bottom of the pressure tube.*

The modeling calculation referred to in this COG Report is for a pressure tube after about 12 EFPY of operation which was the longest exposure of pressure tubes in Canadian reactors at that time (1998). If we extrapolate this calculation to currently operating Units, we would expect the diametral creep to have increased from ~ 2% to ~ 6%. The resulting diffusional redistribution of H/D may then be estimated using the following formalism:

Assuming that the diffusion of hydrogen isotopes in zirconium alloys is a thermally activated process, an Arrhenius-type temperature dependence is expected which may be modelled by the following equation:

$$\ln\{[D_{12}]/[D_6]\} = (Q/R) \times \Delta T/(T_6 \times T_{12})$$

where,

$[D_{12}]$ is the deuterium concentration near the pressure tube outlet at 12 o'clock

$[D_6]$ is the deuterium concentration near the pressure tube outlet at 6 o'clock

Q is the activation energy for deuterium diffusion in Zr-2.5Nb = 22.7 kJ $mol^{-1}K^{-1}$

R is the gas constant = 8.314 J $mol^{-1}K^{-1}$

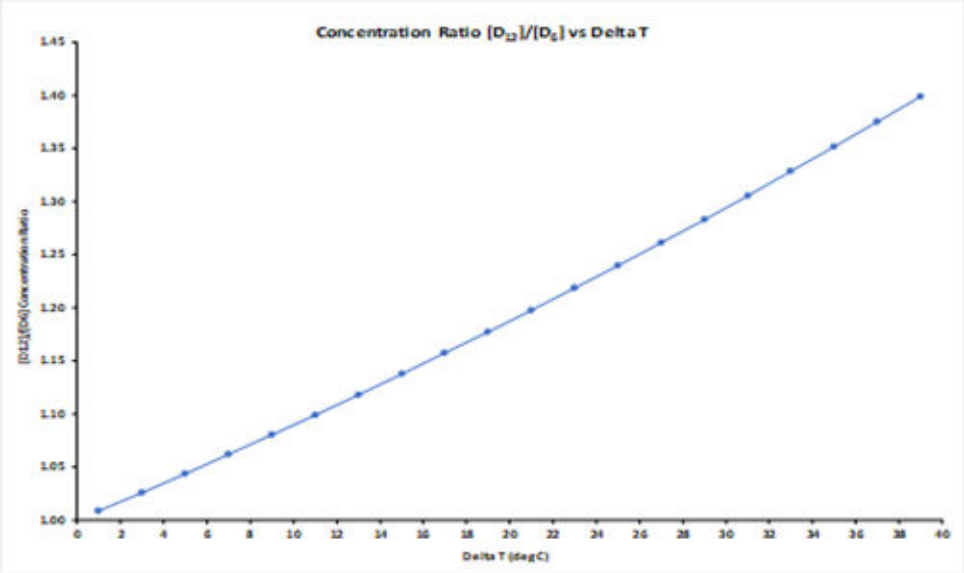
T_6 is the temperature at the bottom, 6 o'clock, position at the outlet of the pressure tube

T_{12} is the temperature at the top, 12 o'clock, position at the outlet of the pressure tube

ΔT is the temperature difference between the 6 o'clock and 12 o'clock locations

Figure10, below, is a graphical representation of this equation plotted for ΔT values up to 40 °C, which may be considered as the highest value of ΔT achievable in mature pressure tubes.

Figure 10: Deuterium Concentration Ratio $[D_{12}]/[D_6]$ vs. $\Delta T = (T_6 - T_{12})$

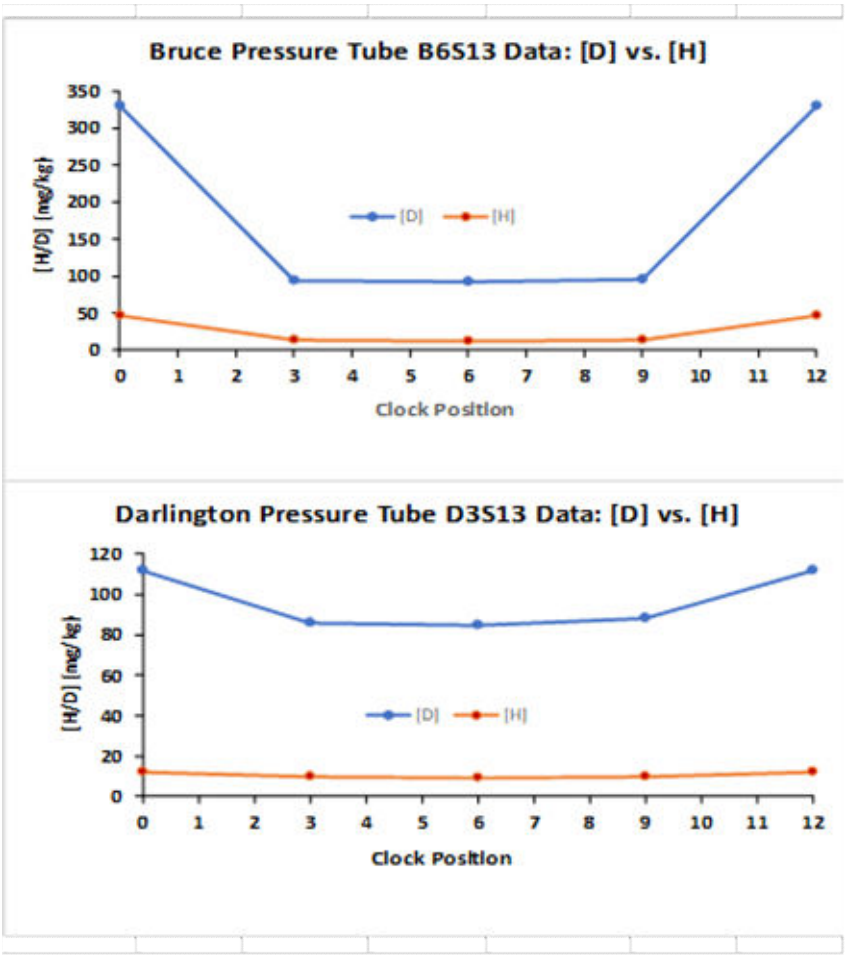


This formalism was previously used to analyze [H] and [D] concentration data in the P3LSFCR sample set at different clock positions and locations approximately 150 mm from the pressure tube outlets – see for example, OHRD 1994 Report A-NFC-94-115-K. For this particular case the observed $[D_{12}]/[D_6]$ ratios were in the range 1.18 ± 0.06 corresponding to a ΔT of 20 °C which is in good agreement with predictions of ΔT for a CANDU reactor after 12 EFPY of Unit operation.

Similar H/D data for a Darlington pressure tube, (D3S13), after 25 EFPY was recently reported by OPG – See the July 2021 OPG Memo: NK38-CORR-31100-0934854. For this pressure tube, data are reported at distances between 8- and 120-mm inboard of the outlet rolled joint and at several different circumferential (clock) positions. The observed $[D_{12}]/[D_6]$ ratios for these samples are in the range 1.25 ± 0.05 corresponding to a ΔT of about 25 °C. Once again, these values are in good agreement with predictions of ΔT for a CANDU reactor after 25 EFPY of Unit operation.

Turning now to [H/D] data for a Bruce pressure tube, namely B6S13, (See OPG Memo: NK38-CORR-31100-0934854), we find the observed $[D_{12}]/[D_6]$ ratio is about 3 which gives a physically unrealistically high value > 60 °C for the corresponding ΔT. This indicates that an additional, (non-thermally activated), process is involved in the H/D ingress into the B6S13 pressure tube. This conclusion is supported by the behavior of H (light hydrogen) in the B6S13 sample which is quite different to the behavior of H in the D3S13 sample, as shown in Figure 11, below. This, once again, is a good indication of an additional source of H/D for the B6S13 sample.

Figure 11: [D] and [H] for Pressure Tubes at Different Clock Positions:
Data for Bruce and Darlington near the Outlet Rolled Joints



The data in Figure 11 are taken from measurements using hot vacuum extraction spectrometry reported in March 2021 by B. Payne at the Canadian Nuclear Laboratories – See Report # COG-19-1034. These results are also discussed in a CNL memo, (ID No: RCC-21-018), dated September 17th 2021, entitled: *Concentrating Hydrogen Isotopes at the Top of Tube at the Outlet rolled Joint Region*, where we read:

> *Hydrogen isotopes in the rolled joint region of the pressure tube originate from three sources: hydrogen initially present in the tube from fabrication, deuterium (including minor amounts of hydrogen) entering the tube during operation from corrosion reactions between the tube and coolant, and deuterium (including minor amounts of hydrogen) entering the tube from the stainless-steel end fitting during operation from corrosion reactions between the end fitting and coolant. The operational sources are evaluated during surveillance examinations and ingress through the end fitting has been observed to be the greater contributor to hydrogen isotope concentrations in this region. For the case of B6S13, the ingress through the inside surface of the pressure tube is in line with other surveillance tubes and scrape campaigns, while the ingress through the rolled joint is marginally greater but well within prior experience projected forward. There is no need, or obvious evidence, for an additional and unknown source of hydrogen to account for the current observations.*

There are two statements that I would question in this CNL assessment of the root cause of the high [H_{eq}] observed in pressure tube B6S13:

(i) The statement that there are only *three* sources of the hydrogen observed in the vicinity of the outlet rolled joint region of a pressure tube

(ii) The statement that "*there is no need, or obvious evidence, for an additional and unknown source of hydrogen*"

Both of these statements ignore the existence of H/D entry into a pressure tube from the *annulus gas system*. Certainly, the three operational sources of H/D identified in this CNL memo are generally considered to be the *major* sources of H/D in CANDU pressure tubes; however, H/D entry from the AGS cannot be ruled out *a priori*. Indeed, there are *two* recent examples of pressure tube failures from high H/D pickup – namely, KAPS-2 on July 1, 2015 and KAPS-1 on March 11, 2016 – that were attributed to H/D entry from the AGS of these Units. Specifically, for item (i) above, it is important to consider potential H/D pickup contributions of *light hydrogen*, H – as opposed to heavy hydrogen, D – to the total hydrogen pickups observed in Bruce pressure tubes. The CNL memo identifies two such sources:

- Hydrogen initially present in the tube from fabrication
- Hydrogen present in the primary heat transport water

Table 5 of this report includes values for the initial hydrogen, H_{Init}, for twenty Bruce Unit 3 pressure tubes, from which an average value for $[H_{Init}]$ of 10 ± 5 mg/kg may be calculated. If we conservatively take an upper limit of 15 mg/kg for $[H_{init}]$, this represents the *minimum* value for [H] after in-reactor exposure of these tubes. It follows that any [H] value *greater* than 15 mg/kg is due to entry from external sources of hydrogen such as the heat transport water. However, as previously noted, heat transport D_2O is specified to have an isotopic purity of at least 99.3 % deuterium, so that the light hydrogen content is only 0.7%.

Using these data, it is possible to estimate a *theoretical hydrogen pickup*, $[H_{calc}]$ for each of the twenty measured deuterium pickups referred to above. Thus, starting with the initial hydrogen contents, $[H_{Init}]$, noted in Table 5, we add a contribution from hydrogen that is assumed to have entered the pressure tube as a fixed, (small), percentage of the measured ingressed deuterium. To allow for kinetic isotope effects in the corrosion reactions leading to such hydrogen ingress, (which depends on the square root of the ratio of the atomic mass of deuterium to the atomic mass of hydrogen), this percentage hydrogen ingress is assumed to be equal to √2 × 0.7%, or 1.0% of [D]. Table 6, below, shows the observed [D] and [H] data for twenty Bruce 3 pressure tubes, as well as the calculated hydrogen concentrations, $[H_{calc}]$, expressed as a percentage of the observed [H].

Table 6: Hydrogen and Deuterium Concentrations Near Bruce Unit 3 Outlet Rolled Joints: Comparison of Data at Three Locations Including [H] Predictions as a % of Observations

Bruce Unit 3 Pressure Tube ID	Data for Samples taken at 10 mm			Data for Samples taken at 55 mm			Data for Samples taken at 112 mm		
	[D] (mg/kg)	[H] (mg/kg)	$[H]_{calc}$ as % of Obs	[D] (mg/kg)	[H] (mg/kg)	$[H]_{calc}$ as % of Obs	[D] (mg/kg)	[H] (mg/kg)	$[H]_{calc}$ as % of Obs
F16	1340	111	23.4	670	67	28.8	241	31	48.4
L11	790	60	28.7	350	29	44.1	99	12	85.8
G15	1016	103	20.8	531	63	26.4	178	30	43.6
K10	853	74	21.5	330	35	30.6	120	26	33.1
Q16	730	96	19.0	314	51	27.5	130	32	38.1
H06	310	66	27.4	171	42	39.8	104	41	39.1
X09	475	73	26.9	186	40	41.9	107	24	66.5
O20	921	96	22.4	451	53	31.7	218	35	41.4
Q12	850	99	18.9	400	52	27.3	142	29	40.1
N04	337	58	31.2	87	22	70.8	100	28	56.1
O15	303	56	20.9	151	31	32.9	94	24	40.2
O17	122	28	36.5	103	26	38.6	89	20	49.5
O13	443	72	23.5	216	50	29.3	108	26	52.2

P14	191	34	32.1	125	24	42.7	86	22	44.8
Q13	582	87	20.1	341	61	24.8	139	29	45.1
L12	156	27	30.6	99	23	33.4	83	19	39.6
F05	75	25	49.8	82	23	54.4	69	27	45.9
L22	42	24	40.5	42	21	46.3	43	18	54.1
R10	199	26	28.2	122	20	33.6	93	16	40.2
S13	314	38	24.1	133	19	38.6	98	17	41.1
Averages	–	–	27.4	–	–	37.2	–	–	47.2

Table 6 shows that for these B3 pressure tubes, the observed light hydrogen concentrations are significantly *higher* than the predicted concentrations. In particular, and taking *average* values, the measured light hydrogen concentrations, [H], are 3.7, 2.7 and 2.2 times higher than the calculated values at the 10 mm, 55 mm and 112 mm locations, respectively.

First, however, it is important to note that the theoretical [H] values in Table 8 are calculated based on the following assumptions:

(i) The heat transport D_2O is the sole source of both hydrogen and deuterium pickup.

(ii) The heat transport D_2O in Bruce Unit 3 contains ~ 1% H_2O

(iii) The calculated [H] values are corrected for contributions from the initial hydrogen in the Zr-2.5Nb ingot used to fabricate the pressure tube.

The fact that the observed concentrations are up to three times *higher* than the calculated concentrations is clear evidence that B3 pressure tube hydrogen isotope pickups near their outlet rolled joints do not conform with the proposal that H/D entry into these pressure tubes comes solely from heat transport D_2O.

Furthermore, this excess light hydrogen needs to be explained because it contradicts the claim, made in the CNL memo, (ID No: RCC-21-018), dated September 17th 2021, that "*there is no need, or obvious evidence, for an additional and unknown source of hydrogen*."

However, it is acknowledged that there are examples of pressure tubes in other CANDU reactors that *are* consistent with H/D entry solely from the heat transport system. Thus, consider the H/D data reported in the July 2021 OPG Memo: NK38-CORR-31100-0934853 for the Darlington pressure tube D3S13, as shown in Table 7, below. The calculated [H] values in the Table are derived assuming an average $[H_{Init}]$ of 10.5 mg/kg for the three samples of interest.

Table 7: H and D Concentration Data Near a Darlington Unit 3 Outlet Rolled Joint: Comparison of Data at Three Locations Including [H] Predictions as a % of Observations

Darlington Unit 3 Pressure Tube ID	Data for Samples taken at 8 mm			Data for Samples taken at 64 mm			Data for Samples taken at 79 mm		
	[D] (mg/kg)	[H] (mg/kg)	$[H]_{calc}$ as % of Obs	[D] (mg/kg)	[H] (mg/kg)	$[H]_{calc}$ as % of Obs	[D] (mg/kg)	[H] (mg/kg)	$[H]_{calc}$ as % of Obs
D3S13	109	12	96.6	112	12	96.8	122	12	97.7

In contrast to the corresponding data for Bruce Unit 3, (See Table 6, above), these Darlington Unit 3 samples show calculated [H] values that are within a few percent of the observed [H] values. Thus, in the case of the D3S13 pressure tube at least, the observed H/D entry at the outlet rolled joint is consistent with the heat transport D_2O being the sole source of ingressed hydrogen.

As previously noted, a number of Canadian nuclear industry experts have suggested that the root cause of the high $[H_{eq}]$ observed near the outlets of some Bruce Unit 3 & 6 pressure tubes is the *redistribution* of ingressed

[H_{eq}] induced by diffusion of H/D in the temperature gradient at this location; with the top of the pressure tube being *cooler* than the bottom by about 25 °C, so that ingressed hydrogen migrates to, and accumulates at the cooler top of the tube.

However, if this is in fact true, evidence for H/D thermal diffusion in a circumferential temperature gradient should be observed at the outlets of *all* mature CANDU pressure tubes, which is certainly <u>*not*</u> the case for the D3S13 example noted above. Furthermore, proponents of the diffusional redistribution of H/D as the sole cause of the high [H_{eq}] observed at the 12 o'clock position near a pressure tube outlet, should explain how at least *five* Bruce Unit 3 pressure tubes could pick up close to 100 mg/kg of *light* hydrogen at this location.

The corresponding heavy hydrogen pickups were approximately 1000 mg/kg, (See Table 5). This implies a H/D *atom ratio* of about 0.2. And one has to ask: What is the *source* of this light hydrogen? Surely the answer must be: the light hydrogen is coming from the AGS of these Units. So, we need to consider evidence for the presence of light hydrogen in operating AGS.

Unfortunately, there are only a few published data on light hydrogen concentrations, (H_2 or H_2O), in CANDU AGS; however, as shown below, what little data there is, shows three significant trends:

(i) Measured H/D atom ratios are typically in the range 0.1 to 0.25

(ii) H/D ratios are highest immediately after an AGS purge and decrease over time

(iii) H/D ratios tend to be higher in the AGS of older Units

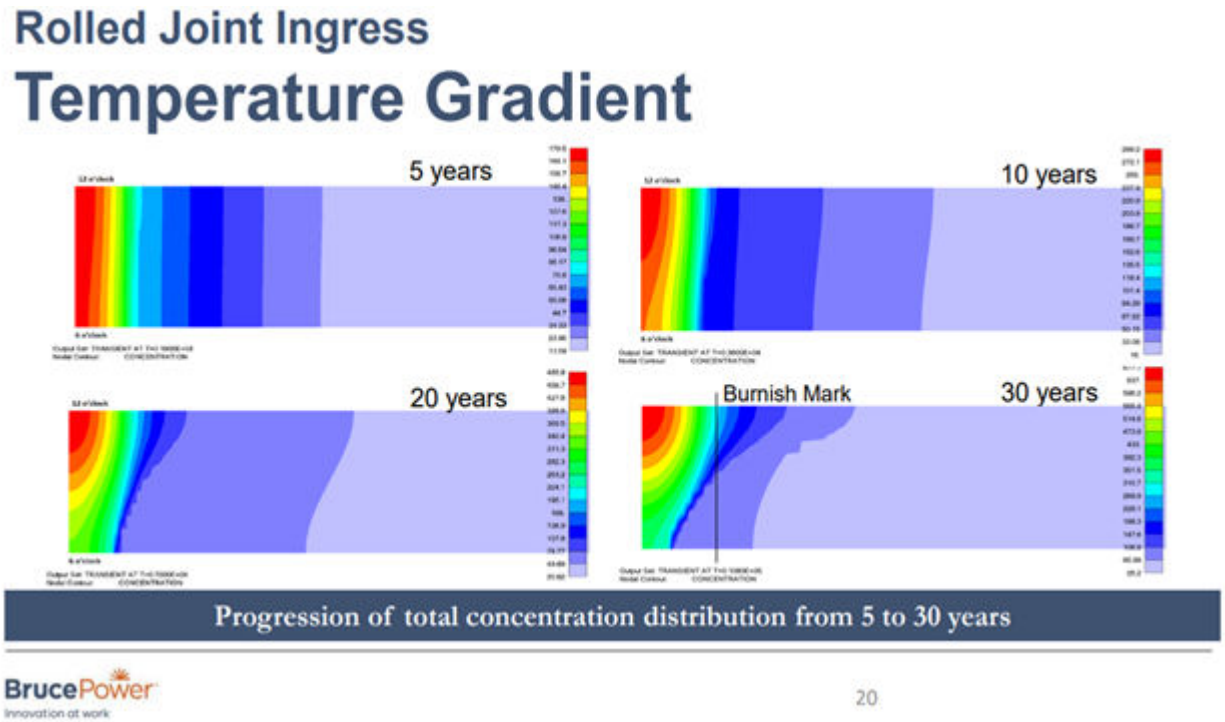
Available evidence suggests that the largest contributor to light water in an AGS is the CO_2 gas supply itself. This is because, at its specified dewpoint of – 45 °C, CO_2 that is deemed to be "dry", nevertheless contains about 70 vpm H_2O.

Studies caried out by myself in the period 1990 to 1998 have shown that light hydrogen, in the form of absorbed water, (H_2O), is always present on the pipework of an AGS and is only *partially desorbed* after system exposures of over 3 hours to dry CO_2, as in a typical AGS purge. (See OHRD Report A-NFC-96-114-P and COG Report COG-96-308).

Such incomplete purges leave significant amounts of light water, H_2O, in the system; and this becomes the initial condition of an AGS at the start of its inter-purge period which usually lasts between 5 and 15 days depending on the Unit. During this time period, water – now mostly as D_2O – builds up in the system via the reverse water gas shift reaction, ($D_2 + CO_2 \rightarrow D_2O + CO$), which is in line with trend (ii) noted above.

Issues of Concern:

1. For my first issue I wish to address some of the material presented by Bruce Power on September 10th, 2021, at CNSC Public Hearing CMD 21-H11.2A. Specifically, I am especially interested in Slide No. 20 from this Hearing – as shown below:



Slide No. 20 is entitled: "*Temperature Gradient*" – suggesting it could provide some insight into the alleged hydrogen redistribution phenomenon; but most remarkably, *no temperature gradient data whatsoever are provided by Slide 20*. Instead, we are presented with hydrogen concentration profiles calculated for a generic pressure tube outlet rolled joint after 5, 10, 20 and 30-years of Unit operation, *with no indication as to how these concentration profiles were determined*. Worse yet, *the magnitude of the alleged temperature gradient is not even mentioned in Slide 20*, and neither is any information provided as to the values of the diffusion coefficients and H/D ingress rates that must have been used to calculate these concentration profiles.

However, regardless of the paucity of useful information provided by Slide 20, we know that Bruce Power's so-called "predictive model" for [Heq] in operating pressure tubes has been *seriously in error* for many years. By way of acknowledging this problem, Bruce Power agreed back in July 2021 to undertake the "*development of a predictive model accounting for elevated [Heq] and circumferential variation of [Heq] observed in the outlet rolled joint region.* – See Bruce Power's letter to the CNSC File No. BP-CORR-00531-01884.

Now, I assume that Slide 20 represents [Heq] values derived from Bruce Power's promised new and improved "predictive model" for [Heq] in its operating pressure tubes. But, for such a model to be scientifically valid, and not merely a curve fitting exercise, it should clearly identify its starting assumptions, input parameters and computational methodology – which is something that has never been provided by Bruce Power. From this observation I believe that Bruce Power's current attempt to predict H/D pickup at pressure tube rolled joints – as exemplified by Slide 20 – is totally unacceptable because it is entirely lacking in scientific rigor, and adds nothing to our understanding of this high H/D pickup phenomenon. But I have to ask if the CNSC would agree with this assessment, or please show me where I am in error.

2. My second issue concerns statements made by CNSC staff member Blaire Carroll at the CNSC Public Meeting held on September 3rd, 2021, (See, CMD 21-M39/21-M37/21-M37.A), as follows, (with my emphasis in red):

MR. CARROLL:

> *For the record, my name is Blaire Carroll. I'm a technical specialist with the Operational Engineering Assessment Division at the CNSC. Dr. Viktorov has provided overall a very good answer from the CNSC staff perspective. We do understand--or we don't understand the root cause at this point. And with regards to some of the modelling that Bruce Power has provided in its presentation, that has not been formally submitted to CNSC staff yet, so we have not*

completed a technical review of that. In theory, it would be the thermal gradients that would move the hydrogen to the top of the pressure tube because that's where the tube is coldest, and the hydrogen tends to migrate to the colder temperature locations in the tube. That would explain why the concentration is highest at the top. But it doesn't explain the magnitude of the concentration that's been seen, and that's an area where CNSC staff is expecting licensees to do more work to try to determine the cause of the elevated values.

This statement by CNSC Staff Member Carroll suggests that the CNSC is not satisfied with Bruce Power's hydrogen diffusion explanation of the high [Heq] observed in some of its pressure tubes. I therefore have to ask, one year on from this statement by a CNSC staff member, if Bruce Power has "*done more work*" and provided the CNSC with an acceptable and experimentally verified root cause of these elevated [Heq] values?

Issue 3:

I believe one of the most important issues concerning Heq pick up is the need for a *root cause* of the high [Heq] first reported in July 2021 for several pressure tubes operating in Bruce Units 3 & 6. Indeed, it is very troubling that it is now over three years since this problem with Bruce pressure tubes was first discovered and, remarkably, the Canadian nuclear industry, and its paid supporters in academia, have only offered a very sketchy qualitative, not quantitative, theory as to the cause of Bruce Units 3 & 6 high [Heq}that is totally lacking in supporting evidence.

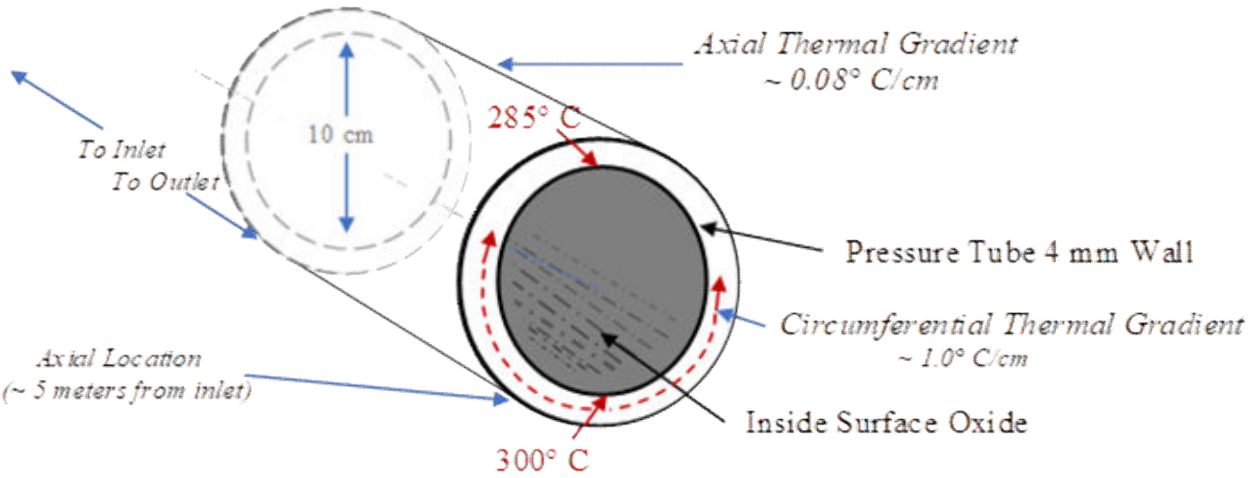
The basis of the nuclear industry's current theory is twofold:

(i) Deuterium, produced by zirconium corrosion in the PHTS, enters a pressure tube at the highest rate at the hottest region of a tube which is at, or near to, the lower (6 o'clock) outlet rolled joint

(ii) The ingressed deuterium subsequently migrates to the cooler regions of the pressure tube located at the top (12 o'clock position) of the tube

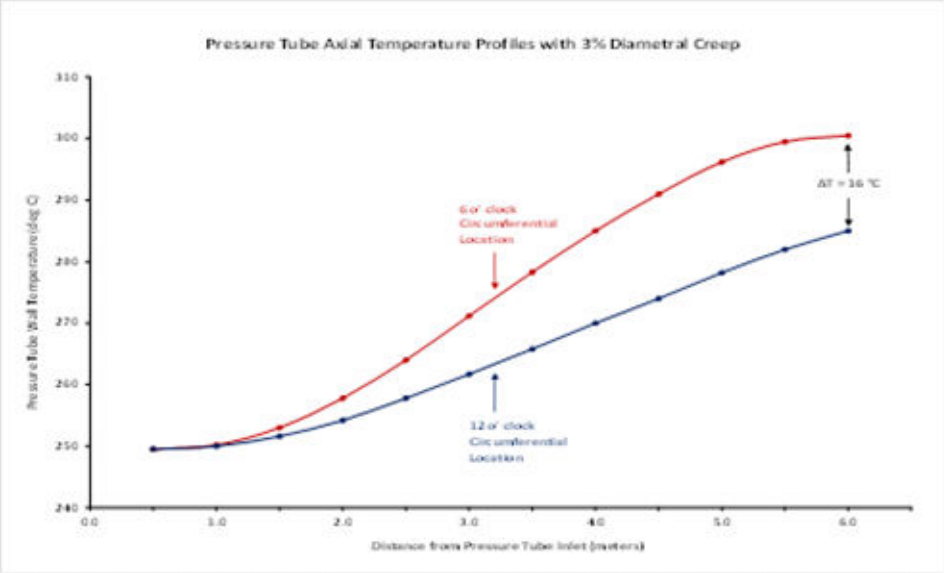
At power, pressure tubes exhibit *two* types of thermal gradients: axial and circumferential, as shown in Figure 1 below. The axial gradient stems from the difference in the inlet and outlet temperatures of the D_2O coolant and equals approximately 0.08 °C/cm along the length of a pressure tube. The circumferential thermal gradient is caused by the off-axis location of the fuel bundles within a fuel channel, especially when the pressure tube wall is subject to neutron-induced diametral creep. As discussed below, the temperature gradient around the circumference of a pressure tube near its outlet end is usually assumed to be about 1.0 °C/cm.

Figure 1: Schematic Illustration of a Pressure Tube near its Outlet End



Thus, for example, D. Rogers et al. at CNL published a plot of the calculated temperature profiles at the 6 o'clock and 12 o'clock circumferential locations for a typical mature pressure tube as shown in Figure 2, below, (See CNL Nuclear Review Vol 5, Number 1, June 2016).

Figure 2: CNL Calculated Temperature Profiles for a Mature Pressure Tube



These plots predict a ΔT – defined as the temperature difference between the bottom and the top of a pressure tube at its outlet end – of about 16 °C. Unfortunately, the methodology used by the CNL to determine these temperature profiles is not provided by the CNL report noted above, from which Figure 2 is derived. Nevertheless, it is possible to predict the basic features of these profiles based on the fact that there are two main contributing factors that determine ΔT:

(i) A contribution, ΔT_6, from *the localized heating of a pressure tube* in the vicinity of the 6 o'clock location, caused by the direct physical contact of fuel bundles with the pressure tube wall at the bottom of a tube

(ii) A contribution, ΔT_{12}, from *the localized cooling of a pressure tube* in the vicinity of its 12 o'clock location, caused by coolant flow bypass due to pressure tube diametral expansion from neutron induced creep

It is assumed that:

$\Delta T(x) = \Delta T_6(x) + \Delta T_{12}(x)$

Where x is the axial distance (in meters) from the pressure tube inlet, (x = 0), to outlet, (x = 6).

1. **Determination of ΔT_6:**

ΔT_6 depends on the bundle power, usually expressed in kW, at a given axial location. I have used bundle power data from a Bruce B Safety Report for a high power, (7.5 MW), channel to derive a dimensionless axial power profile, P(x), over the full 6-meter reactor core, with the data points normalized to the maximum channel power of 1035 kW at the 3-meter axial location. It is assumed that ΔT_6 may be approximated by the simple relationship:

$\Delta T_6(x) = C_1 \times P(x)$

Where C_1 is a proportionality constant, (in units of °C), between the excess pressure tube wall temperature and the local fuel bundle output power – a conversion factor to be determined.

Using published data derived from CANDU reactor thermal hydraulic codes such as ASSERT-PV, C_1 is estimated to be 10 °C.

2. **Determination of ΔT_{12} :**

ΔT_{12} depends on the amount of pressure tube diametral expansion induced by neutron induced creep. CNL has published plots of the amount of diametral expansion, D(x), along the axial length of a pressure tube, (See R. B. Adamson et al. in Journal of Nuclear Materials Vol. 521, 167 – 244, (2019). It is assumed that ΔT_{12} may be approximated by the simple relationship:

$\Delta T_{12}(x) = C_2 \times D(x)$

Where C_2 is a proportionality constant, (in units of °C), between the amount of pressure tube wall temperature cooling and the neutron-induced pressure tube diametral expansion normalized to its maximum value which is well in-board of the mid-core, 3-meter location. For this intervention I have used a value of 8 °C for C_2.

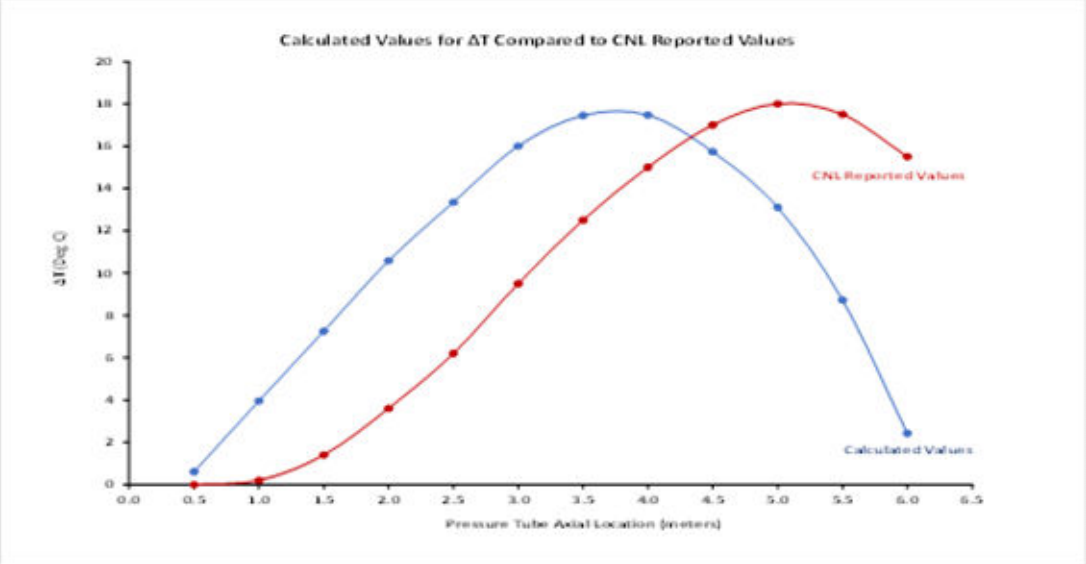
Combining these parameter values with axial power profile and the diametral expansion data noted above we are able to calculate ΔT(x) for a set of axial locations from x = 0 to x = 6.0 meters, as shown in Table 1, below, together with ΔT values taken from CNL's 2016 report.

Table 1: Calculated ΔTs at Different Axial Locations vs. CNL's Published Data

Axial Location, x (meters)	ΔT_6 (Deg C)	ΔT_{12} (Deg C)	$\Delta T_6 + \Delta T_{12}$ (Deg C)	CNL ΔT (Deg C)
0.0	0.0	0.0	0.0	0.0
0.5	0.6	0.0	0.6	0.0
1.0	2.4	1.5	4.0	0.2
1.5	4.4	2.9	7.3	1.4
2.0	6.4	4.2	10.6	3.6
2.5	8.0	5.4	13.4	6.2
3.0	9.5	6.6	16.0	9.5
3.5	10.0	7.5	17.5	12.5
4.0	9.5	8.0	17.5	15.0
4.5	8.0	7.8	15.7	17.0
5.0	6.4	6.7	13.1	18.0
5.5	4.5	4.2	8.7	17.5
6.0	2.4	0.0	2.4	15.5

It is instructive to plot the data in columns 4 and 5 of Table 1 to directly compare the ΔT values determined by the methodology described above, with the results reported by CNL, as shown in Figure 3 below.

Figure 3: Calculated vs. CNL Reported Values for a Pressure Tube 6 to 12 o'clock ΔT



The most striking feature of my calculated values of ΔT compared to CNL's values, as seen in Figure 3, is the large divergence between the data sets at the pressure tube outlet region near the 5-to-6-meter axial location. In this outlet region of the reactor core the neutron flux drops off rapidly with increasing x, as the bundle power declines by about 200 kW/meter from its mid-core output of about 1000 kW. Because diametral creep is a function of the fast neutron fluence, the pressure tube diametral expansion also falls to a minimum value of about 15% of its peak value at the 6-meter location. This trend is clearly seen in my calculated ΔTs, but is barely evident in CNL's calculated values, which is difficult to explain.

I believe that my calculated ΔTs are much closer to reality than CNL's and as a consequence it appears that the ΔTs for the Bruce Unit 3 and 6 pressure tubes with elevated [Heq] near their respective outlet rolled joints are most probably less than 5 °C – a value that is incompatible with the observed $[D_{12}]/[D_6]$ ratio of about 3, which requires a physically unrealistic value > 50 °C for the corresponding ΔT, (See Figure 10 of this intervention).

I have recently re-calculated EoL pressure tube ΔTs using a different formalism of the effects of pressure tube diametral expansion from neutron induced creep. In this alternative approach the pressure tube expansion is calculated from the change in the cross-sectional *area* of the tube to more accurately quantify the flow bypass. The resulting values of ΔT are plotted as compared to the CNL calculated values in Figure 4.

Figure 4: Calculated vs. CNL Reported Values for a Pressure Tube 6 to 12 o'clock ΔT
Alternative Calculation Method

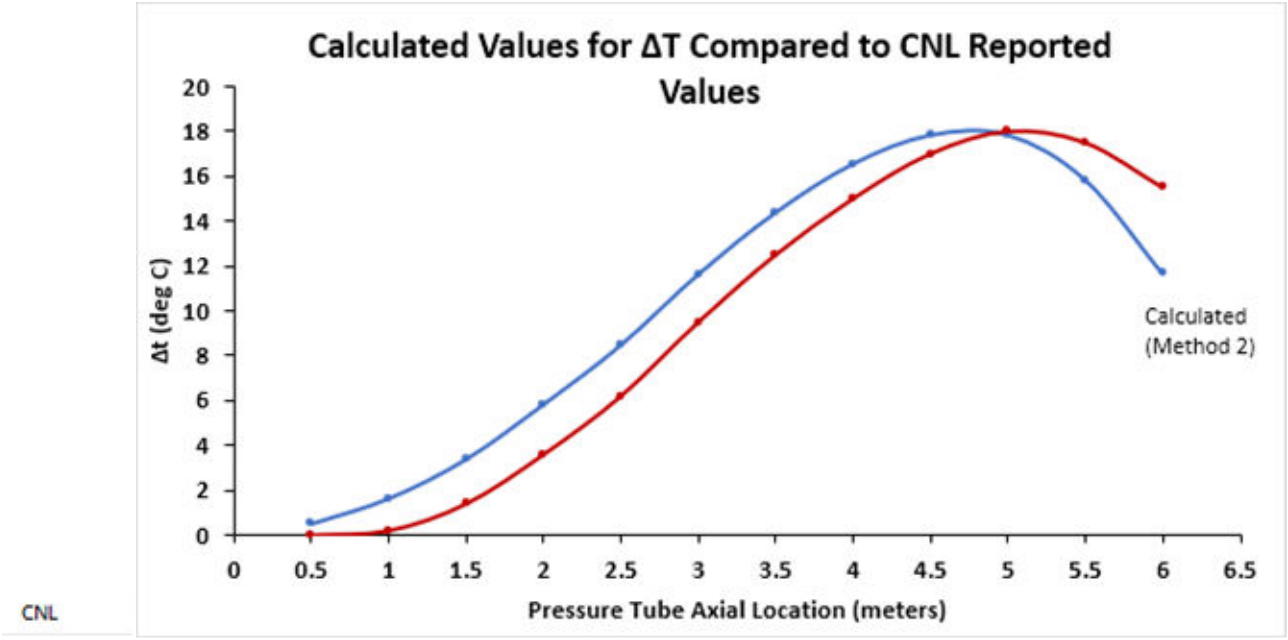


Figure 4 shows better agreement with the CNL values but still shows lower, and more realistic ΔTs near the pressure tube outlet than the CNL values.

A plot of the predicted maximum diametral expansion data for Bruce Unit 7 pressure tubes shows that this Unit will have already exceeded the 108.6 mm diameter limit at 300,000 EFPH. One therefore has to ask if Bruce Power can guarantee that Bruce Unit 7 will not be subject to fuel sheath dry-out if operated beyond 300,000 EFPH?

Sincerely,
Dr. F. R. Greening