



## **Supplementary Information**

**Written submission from  
Frank R. Greening**

In the Matter of

**Ontario Power Generation Inc.,  
Pickering Nuclear Generating Station**

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Request for a ten-year renewal of its Nuclear Power Reactor Operating Licence for the Pickering Nuclear Generating Station

**Commission Public Hearing – Part 2**

**June 2018**

## **Renseignements supplémentaires**

**Mémoire de  
Frank R. Greening**

À l'égard de

**Ontario Power Generation Inc.,  
centrale nucléaire de Pickering**

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Demande de renouvellement, pour une période de dix ans, de son permis d'exploitation d'un réacteur nucléaire de puissance à la centrale nucléaire de Pickering

**Audience publique de la Commission –  
Partie 2**

**Juin 2018**



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**From:** Frank Greening  
**Sent:** Friday, May 18, 2018 9:21 PM  
**To:** Interventions (CNSC/CCSN)  
**Cc:** Levert, Louise (CNSC/CCSN); Leblanc, Marc (CNSC/CCSN)  
**Subject:** Supplemental Material for my Pickering Intervention  
**Attachments:** AGSPAPER.docx

To: The Senior Tribunal Officer  
Secretariat of the Canadian Nuclear Safety Commission (CNSC)  
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Re: Supplemental material for my submission to the CNSC Public Hearing to consider Ontario Power Generation's (OPG's) application to renew its Nuclear Power Operating Licence for the Pickering Nuclear Generating Stations, A and B, for a period of 10 years, Hearing Number Ref. 2018-H-03

To whom it may concern:

Today, May 18<sup>th</sup>, 2018 I became aware of a submission, CNSC reference No: CMD 18-H6.20, from Paul Sedran of RESD Inc, for the upcoming Pickering relicensing hearing. Mr. Sedran's submission includes Sections 4.2 to 4.5 which discuss the so-called leak-before-break methodology as currently applied to pressure tube failures in CANDU reactors. Mr. Sedran's analysis makes the claim that the first indication of moisture from a through-wall crack in a pressure tube could be detected by dew point measurements of the affected Unit's AGS, (Annulus Gas System), within 1 hour of the start of leakage – See Sedran's Figure 3. In addition Mr. Sedran estimates that a so-called beetle alarm would occur within 2.5 hours.

By way of a supplemental submission to my CMD 18-H6-155, I wish to include this email as well as the attached material, (AGSPAPER.doc), as a refutation of the claims made by Mr. Sedran concerning the efficacy of pressure tube leak detection by AGS dew point measurements. In the attached paper I show that leak detection times in real CANDU AGS are orders of magnitude higher than the times quoted by Mr. Sedran. In addition, I should note that AGS leak detection sensitivity is adversely affected by an AGS purge. It is for this reason that I requested AGS purge frequency data in my original submission.

Sincerely,

Dr. F. R. Greening



# **CANDU Pressure Tube Leak Detection by Annulus Gas Dew Point Measurement: A Critical Review**

**By F. R. Greening**

## **1.0 Introduction**

The use of a CANDU Annulus Gas System (AGS) for the early detection of a pressure tube leak – a reactor operating methodology known as “leak-before-break” – is reviewed and its efficacy analysed. It is shown that AGS moisture injection tests, as used at Bruce, Pickering, and Darlington to demonstrate acceptable leak detection performance were flawed because they were carried out under conditions that did not simulate real pressure tube leaks.

## **2.0 Leak-Before-Break as Applied to CANDU Pressure Tubes**

The concept of leak-before-break, (LBB), is an operational requirement that, in the event of a pressure boundary leak from a through-wall crack during normal reactor operation, 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.

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*, [1]. 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 crack growth rate,  $V$ ,* which is usually expressed as a velocity, in m/s, and is typically in the range  $1 \times 10^{-7}$  to  $2 \times 10^{-6}$  m/s.  $V$  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.  $\sim 2$  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, [2]. Nevertheless, in spite of these variabilities, the approach used at CANUU stations is to average the available data; using this approach *typical* pressure tube LBB behavior is considered to be 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  $\sim 5.3 \times 10^{-7}$  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.  $\sim 2$  hrs).

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<sub>2</sub>O leak rate in kg/hr

$2C$  = crack length in mm

For crack growth at a velocity  $\sim 5 \times 10^{-7}$  m/s, this equation predicts a D<sub>2</sub>O 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<sub>2</sub>O 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<sub>2</sub>O leak detector. 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*. Therefore, it is important to evaluate the reliability, sensitivity, precision and accuracy of annulus gas leak detection *as currently practiced at CANDU stations*. These issues are reviewed in detail in Sections 3.0 to 7.0 of this paper.

### 3.0 What is a CANDU Annulus Gas System (AGS)?

The CANDU power reactor design was introduced to the world on April 11<sup>th</sup> 1962 with the commissioning of the 22 MW<sub>e</sub> Nuclear Power Demonstration (NPD) reactor in Rolphton, Ontario. This basic CANDU design consisted of an array of horizontal fuel channels interpenetrating a moderator tank (or *calandria*) where each channel was separated from the moderator by an annular gap of about 1cm. In the NPD this annular gap was left open to the air which provided the necessary thermal insulation to maintain the moderator at about 80 °C while the pressure tube carried heat transport heavy water (D<sub>2</sub>O) at up to 277 °C.

After about 20 years of operation of the NPD reactor the presence of radial zirconium hydrides in two removed pressure tubes, (G-07 and F-08) caused the fracture toughness of these tubes to decrease to such an extent that it became questionable whether a through-wall crack could be detected and the reactor shut down before the crack had grown to the critical size, (about 50 mm), for unstable rupture, [3]. In the NPD open annulus design, the only option available to the operator in the event of a pressure tube leak was to measure the increase in humidity in the reactor vault from the escaping water vapor. It was estimated that this would take at least 40 hours, which was too long to satisfy leak-before-break safety criteria. Thus the decision was taken to close down the NPD reactor.

Nevertheless, a solution to this problem was developed for the next generation of CANDU reactors. An optimum approach would have been to close off the ends of the pressure/calandria tube annuli and monitor the moisture content of the air in each channel. However, because this would have required each reactor to deploy moisture monitoring for ~ 400 individual annuli, a compromise was adopted whereby the annuli are interconnected into 48 separate “strings”, each made up of between 4 and 12 channels. The flow of gas in each string is controlled by a rotameter and the entire volume of gas in the system, (~12,000 liters), is circulated by a compressor at a flow rate of up to 6 liters/second. This method of interconnecting channel annuli in “strings” is now incorporated into the design of the AGS in all currently operating CANDU reactors.



#### 4.0 How are pressure tube leaks detected by an AGS?

A pressure tube leak results in the release of high pressure heavy water to the AGS. Because an AGS is operated at about 150 °C, and a little above atmospheric pressure, the escaping water immediately “flashes” to steam. The presence of water vapor in an AGS is continuously monitored by two (redundant) dew point meters connected in parallel outside of containment at the outlet end of the AGS. The meters used in most CANDU stations are Parametric Model 600 dew point hygrometers which utilize aluminum oxide moisture sensors.

It is important to recognize that with such an arrangement of dew point meters, only an *average* AGS moisture content is measured and thereby indicates how much water is present in the *total* AGS volume at that time. However, this dew point meter reading does *not* identify the source of a leak. Nevertheless, a steadily *increasing* AGS dew point, observed over several hours, indicates that there is water in-leakage occurring *somewhere* in the system. CANDU reactor operating experience has shown that three types of leaks are possible:

- (i) End shield cooling leaks. These are chronic *light water* leaks usually caused by a flaw in a lattice tube weld.
- (ii) Pressure tube/end fitting rolled joint leaks. These are chronic heavy water leaks caused by improper rolling procedures during pressure tube installation.
- (iii) Pressure tube leaks involving through-wall cracks. As previously noted, these are potentially catastrophic leaks unless they are detected before the pressure tube crack has grown beyond the critical size for unstable rupture.

#### 5.0 AGS Chemistry

What has been discussed so far about a CANDU reactor’s annulus gas system is its physical layout and the various modes of entry of water. However, *the chemistry of a CANDU AGS* is extremely important in assessing pressure tube leaks, especially since the fill gas has been changed several times over the past 30 years. Localized corrosion of metal surfaces, especially caused by AGS chemistry upsets due to air or water ingress, may also adversely impact an AGS response time.

## 5.1 Air Filled AGS

Air was the first gas used to fill a CANDU AGS, as in the NPD reactor commissioned in 1962. And, as we have seen, water vapor entering the NPD AGS was simply allowed to diffuse into containment and escape to the environment via station ventilation stacks. The resulting dilution of water vapor generated in the AGS made leak detection by dew point measurement practically impossible.

## 5.2 Nitrogen Filled AGS

The need to close off open-ended fuel channel annuli led to the Pickering reactor design where AGS “bellows” and “pigtailes” were used to “feed and bleed” the fill gas to and from each channel. In addition, the Pickering AGS used nitrogen as the fill gas instead of air. As it turned out, the choice of nitrogen to fill an AGS was ill-advised; this is because nitrogen has a significant neutron absorption cross section, (1.8 barns), for the conversion of nitrogen-14 to carbon-14 by the  $^{14}\text{N}(n,p)^{14}\text{C}$  reaction. As a result, carbon-14 particulate accumulates in a  $\text{N}_2$ -filled AGS at a rate of about 40 grams per year. When Pickering Units 1 & 2 were opened up for replacement of their pressure tubes in February 1985, carbon-14- rich deposits were discovered throughout the AGS, but were especially heavy on the radiation shielding sleeve and the outboard bearing journal ring, [4].

Also observed in AGS samples collected from Pickering Units 1, 2 & 3 in the period 1984 – 1992 were brown or black deposits containing iron oxide mixed with carbon-14. These deposits were found not only in the locations noted above but also in the AGS pigtailes and bellows. It was subsequently realized that these deposits were causing annulus gas flow blockages that were very detrimental to pressure tube leak detection by AGS dew point measurements.

## 5.3 Carbon Dioxide Filled AGS

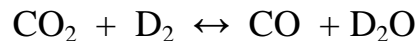
Because of problems with carbon-14 formation, all CANDU reactors had switched from  $\text{N}_2$  to  $\text{CO}_2$  as the fill gas for their AGS by the mid-1980s. However, far from providing an optimum medium for trouble-free pressure tube leak detection by dew point measurements, the use of  $\text{CO}_2$  introduced a new set of problems. The most important of these problems were:

(i) The accumulation of viscous organic deposits in AGS flow rotameters leading to partial or complete blockage of the affected strings, [5].

(ii) The entry of deuterium gas, D<sub>2</sub>, into an AGS and its conversion to D<sub>2</sub>O by reaction with CO<sub>2</sub>, [6].

The first of these problems – deposit formation in AGS flow rotameters – was observed at Bruce A in the late 1980s. Chemical analysis of samples of deposit from Bruce Unit 3 showed the presence of complex mixtures of long-chain dicarboxylic acids, HOOC-(CH<sub>2</sub>)<sub>n</sub>-COOH, and cyamelide polymers, [NH-CO]<sub>n</sub>, [7]. The presence of nitrogen in these samples was unexpected but consistent with the fact that N<sub>2</sub> is routinely detected in a CO<sub>2</sub>-filled AGS at concentrations up to 1000 vpm. Oxygen addition to the AGS, at concentrations up to 2 % by volume, was introduced in the late 1980s as a way to counter polymer production.

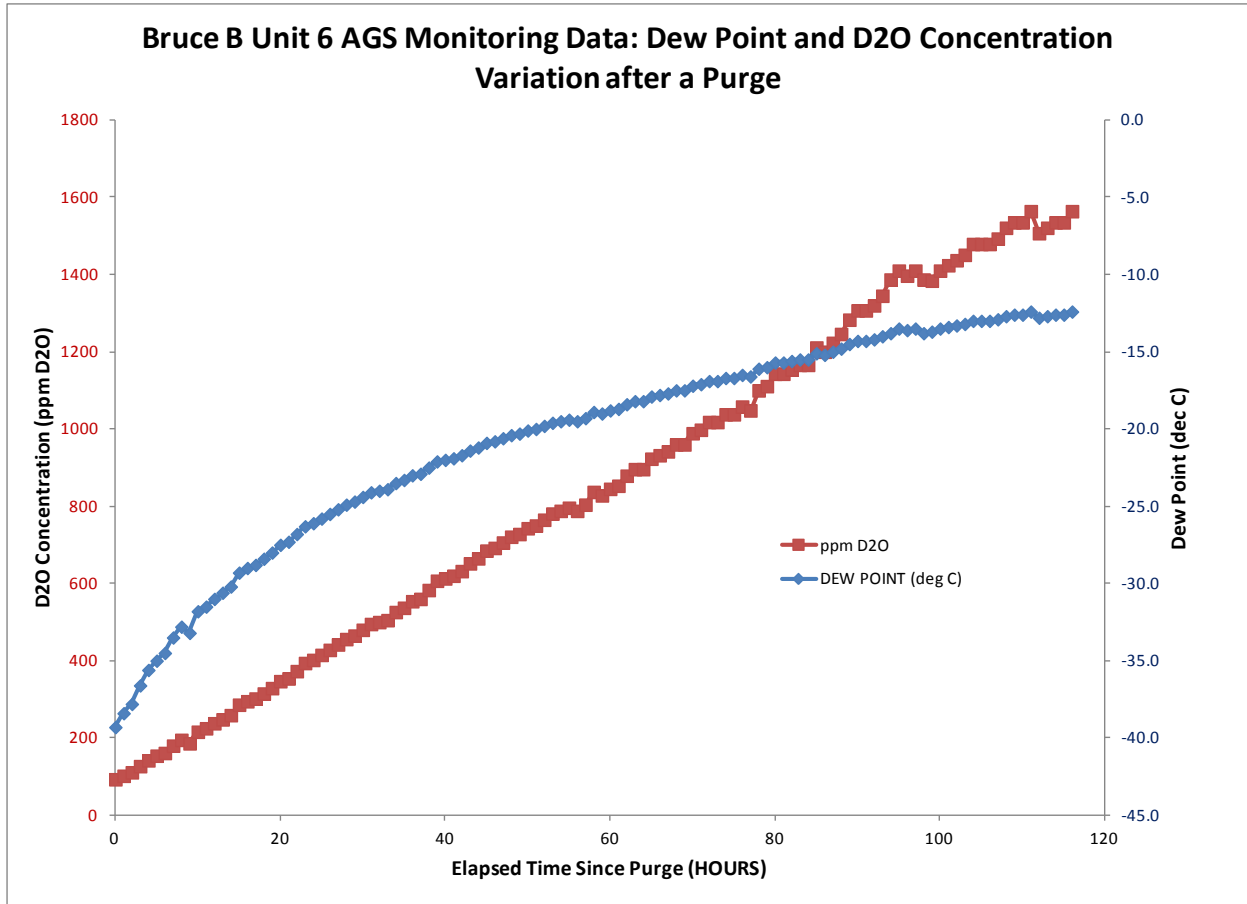
The second problem with the use of CO<sub>2</sub> to fill an AGS is that it undergoes the so-called *reverse water gas shift reaction* in which it acts as an oxidizing agent to deuterium, (that enters an AGS by mechanisms discussed below). This leads to the formation of water, (D<sub>2</sub>O), while the CO<sub>2</sub> is itself reduced to carbon monoxide:



The symbol “↔” indicates that this is a *reversible* reaction that does *not* go to completion under typical annulus gas conditions, namely temperatures ~ 150 °C, and pressures ~ 1 atmosphere. Nevertheless, the water gas equilibrium means that as more D<sub>2</sub> enters an AGS, more D<sub>2</sub>O is formed.

Station data show that D<sub>2</sub> enters a CANDU AGS at variable (Unit dependent) rates, typically between 10 and 50 vpm/hour. It is useful to compare this to the corresponding moisture ingress rates derived from dew point data. Thus, while the dew point varies from – 30 °C to –10 °C, instrument calibration data for the Bruce B Unit 6 AGS show that the D<sub>2</sub>O concentration varied from 100 vpm to 1600 vpm D<sub>2</sub>O over about 120 hours – See Figure 1:

**Figure 1: Dew Point vs. D<sub>2</sub>O Concentration Variation After a Purge**



This data shows that there was an approximately linear rate of increase in the D<sub>2</sub>O concentration in the Bruce Unit 6 AGS equal to about 13 vpm/hour.

D<sub>2</sub> is believed to enter an operating AGS by a combination of three mechanisms:

- (i) Gaseous permeation of H/D through the walls of pressure tubes and/or end fittings
- (ii) Water-induced corrosion of in-core metal surfaces, (Zr, Fe, etc), via the reaction: Metal + Water = Metal Oxide + Hydrogen
- (iii) Degassing of primary heat transport water entering an AGS through leaky rolled joints that connect pressure tubes to steel end fittings

Calculations show that mechanisms (i) and (ii) could contribute a maximum of 0.32 liters/hour, and 0.18 litres/hour of D<sub>2</sub>, respectively. Mechanism (iii) is difficult to quantify but there is reason to believe it could make a significant contribution to D<sub>2</sub> ingress into an AGS. The level of dissolved D<sub>2</sub> in a PHTS is controlled at  $8 \pm 4$  cm<sup>3</sup>/kg of D<sub>2</sub>O for corrosion control and it has been found that variations in this dissolved D<sub>2</sub> correlate with the rate of D<sub>2</sub> ingress into an AGS. From such observations it has been estimated that D<sub>2</sub> ingress by mechanism (iii) occurs at rates between 0.45 and 0.75 liters/hour for PHTS dissolved D<sub>2</sub> concentrations between 4 and 10 cm<sup>3</sup>/ kg D<sub>2</sub>O, respectively.

#### 5.4 Carbon Dioxide with Added Oxygen

By the early 1990s it was considered prudent to add O<sub>2</sub> to a CO<sub>2</sub>-filled AGS at concentrations up to 2 vol % to suppress the build up of D<sub>2</sub> and thereby maintain an oxidizing environment that would prevent deuterium pick up by pressure tubes from their annulus gas side. Initial tests with O<sub>2</sub> addition to an AGS resulted in very high Ar-41 radiation fields due to the neutron activation of trace argon which is usually present in industrial grade O<sub>2</sub>. As a result, high purity oxygen is now specified and noble gas emissions from CANDU AGS undergoing O<sub>2</sub> additions are generally not significantly increased. The O<sub>2</sub> gas supply requirements are as follows:

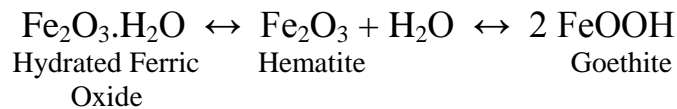
- > 99.98% O<sub>2</sub>
- < 0.1% N<sub>2</sub>
- < 2 µL/L Hydrocarbons
- < 15 µL/L Ar, Kr and Xe combined
- < 50 µL/L H<sub>2</sub>
- < 10 µL/L moisture (Dew point > - 60 °C)

O<sub>2</sub> is added to an AGS in such a way as to maintain the O<sub>2</sub> concentration between 0.5 and 2.0 vol % at all times. After an initial system purge, the AGS is placed in re-circulating mode and the added O<sub>2</sub> rapidly combines with free D<sub>2</sub> to produce D<sub>2</sub>O as intended. However, this means the pre O<sub>2</sub>-addition average dew point rate of rise inevitably *increases* after the introduction of O<sub>2</sub> additions with a resulting *increased* AGS purge frequency.

## 5.5 The Impact of Localized Corrosion on Long-Term AGS Performance

As we have seen, CANDU reactors operate with an AGS containing varying amounts of ingressed water, (100 – 2000 vpm), in a CO<sub>2</sub>/O<sub>2</sub> atmosphere. The presence of water and oxygen at temperatures up to 250 °C creates a relatively corrosive environment for the > 10<sup>7</sup> cm<sup>2</sup> of metal surfaces in an AGS. These surfaces include zirconium alloys (pressure and calandria tubes), stainless steel (lattice tubes and end fittings), hardened tool steel (bearing and journal rings), carbon steel (shielding sleeves) and Inconel (bellows). Of these materials, carbon steel is the most susceptible to corrosion, especially in the presence of water, which leads to the formation of hydrated ferric oxide or “rust”. Thus, reddish-brown colored deposits, exhibiting very high specific activities of Fe-55, (600 mCi/g), have been observed on many AGS surfaces in Pickering and Bruce Units during their refurbishments, [8].

In the temperature range relevant to an AGS, namely 50 to 250 °C, rust deposits are subject to two reversible transformations involving the adsorption or desorption of water:



It is readily calculated that oxidation of a 100 µm layer of the carbon steel components in an AGS would create sufficient goethite to absorb/desorb about 500 g of water; this would be more than enough to explain the sluggish response of Pickering Unit 3 in its AGS moisture injection tests in 1991.

Another potentially serious corrosion problem observed in a number of AGS during moisture injection testing is the formation of NO<sub>2</sub> at concentrations up to 500 vpm. The presence of NO<sub>2</sub> in these tests was attributed to air ingress into the AGS, followed by radiolysis of the N<sub>2</sub>/O<sub>2</sub> mixture to form NO<sub>2</sub> and N<sub>2</sub>O, [9]. In the presence of water, nitric acid, HNO<sub>3</sub>, is rapidly formed by reaction of NO<sub>2</sub> with OH free radicals generated by the radiolytically induced decomposition of water.

During the Bruce Unit 4 AGS moisture injection tests in 1994 anomalous and erratic dew point meter readings were noted immediately after the tests began. Metal surfaces in the dew point meter housings were analyzed after the tests and concentrations of nitrate ion ~ 2 µg/cm<sup>2</sup> were found on all the surfaces examined. It is suspected that this nitrate was in the form of ferrous or ferric nitrate corrosion

product,  $\text{Fe}(\text{NO}_3)_{2/3}$ , which is slowly hydrolysed in the presence of water to goethite,  $\text{FeOOH}$ , the most stable form of hydrated ferric oxide below 100 °C.

The greatest concern with these and similar observations is that in the event of acute air or water ingress into an AGS – a relatively common occurrence – older AGS are going to become increasingly contaminated with the resultant corrosion debris. This debris has two deleterious effects:

- (i) Corrosion debris can absorb/desorb significant amounts of water
- (ii) Corrosion debris can cause annulus gas flow restrictions or blockages

Point (i) further complicates the already difficult thermal-hydraulic analysis of an AGS under extreme conditions. Nevertheless, a simplified model of water ingress into a mature CANDU reactor, exemplified by Darlington Unit 1 in 1996, has been developed, [10]. This model shows that in the first 10 hours after a system is purged, heavy water desorption from the AGS pipe work is ongoing and is the major source of  $\text{D}_2\text{O}$  ingress into the system. This is because water desorption is a slow process; thus, even when purging of an AGS is stopped, water desorption continues unabated until a new equilibrium state is attained.

Point (ii) has been a long-standing issue for CANDU reactors because there appears to be no simple solution to the problem of flow restrictions and blockages in aging AGS. Corrosion debris may come from a number of sources, but the carbon steel shielding sleeves are a major contributor because they are not very corrosion resistant and readily form a loosely adherent iron oxide coating in a moisture laden environment. Furthermore, corrosion product dislodged from a shielding sleeve tends to collect in the very narrow opening, typically only 0.4 mm wide, at the inboard channel bearing - a location that is already a region with a very low gas flow rate.

Indeed, thermal-hydraulic analysis of the Bruce B AGS shows that for a system flow rate of 2.6 l/s the maximum flow through an individual channel is only 0.054 l/s, [11]. Since one channel represents about 30 liters of AGS volume, it takes almost 10 minutes for a channel volume of  $\text{CO}_2$  to be exchanged. However, if corrosion debris collects around a bearing sleeve, the flow will be even more restricted and a sluggish response to any air or water ingress into the AGS is an unavoidable consequence.

## 6.0 Annulus Gas Purging: Theory and Observation

As we have seen, a rising AGS dew point indicates that there is moisture entering the system. A slow rate of rise is considered “normal” because there is always a small chronic in-leakage of moisture into the AGS. However, once too much moisture has entered, the AGS must be purged to restore the Unit’s pressure tube leak detection capability. CANDU AGS are specified to operate over a range of dew points from  $-30\text{ }^{\circ}\text{C}$  to  $-10\text{ }^{\circ}\text{C}$ , so that a dew point reading above  $-10\text{ }^{\circ}\text{C}$  requires an immediate system purge. Station dew point meter calibration data, (See Figure 1 for example), show that  $-30\text{ }^{\circ}\text{C}$  and  $-10\text{ }^{\circ}\text{C}$  dew point readings correspond to  $\text{D}_2\text{O}$  concentrations of 500 vpm and 2,000 vpm, respectively.

In order to evaluate an annulus gas system’s leak detection capability, it is first necessary to consider the “normal” rate of rise of the system’s dew point in the absence of an acute pressure tube leak. This may be predicted from knowledge of the AGS volume (12,000 liters) and the  $\text{D}_2\text{O}$  concentration corresponding to a dew point of  $-10\text{ }^{\circ}\text{C}$ , (2,000 vpm). Then, if we assume the  $\text{D}_2\text{O}$  in-leakage rate is constant over a purge cycle, we may calculate the time between successive AGS purges for different  $\text{D}_2\text{O}$  ingress rates:

**Table 1: Time between Purges for Different  $\text{D}_2\text{O}$  Ingress Rates**

<b><math>\text{D}_2\text{O}</math> In-leakage Rate</b>		<b>Time to Reach Purge Point</b>	
<b>grams/hour</b>	<b>vpm/hour</b>	<b>Hours</b>	<b>Days</b>
0.125	10	200	8.33
0.25	20	100	4.16
0.5	40	50	2.08
1.0	80	25	1.04
2.0	160	12.5	0.52
4.0	320	6.25	0.26

The range of  $\text{D}_2\text{O}$  ingress rates covered in Table 1 approximately encompasses the range of purge times observed in actual CANDU reactors worldwide. New, “leak tight”, CANDUs have achieved purge periods in excess of 8 days immediately after commissioning. However, as a reactor ages, its leakiness tends to increase and AGS purges are generally needed every 5 days in reactors that are more than 10 years old. At the other extreme, a very short interval between AGS purges, as listed in Table 1, would be 6.25 hours. This interval was chosen because it’s close to the



*practical* limit on a system’s purge frequency, although this limit is ultimately set by the finite time required to purge the system.

In theory, at a re-circulating flow rate of 5 litres/s, a wet AGS should take less than an hour to dry to a  $-30\text{ }^{\circ}\text{C}$  dew point. However, because the *maximum purge flow rate* achieved by most CANDU reactors is only about 2 litres/s, 5 hours is the recommended time for the completion of an AGS purge since this allows for approximately *three* AGS volumes to be displaced from the system.

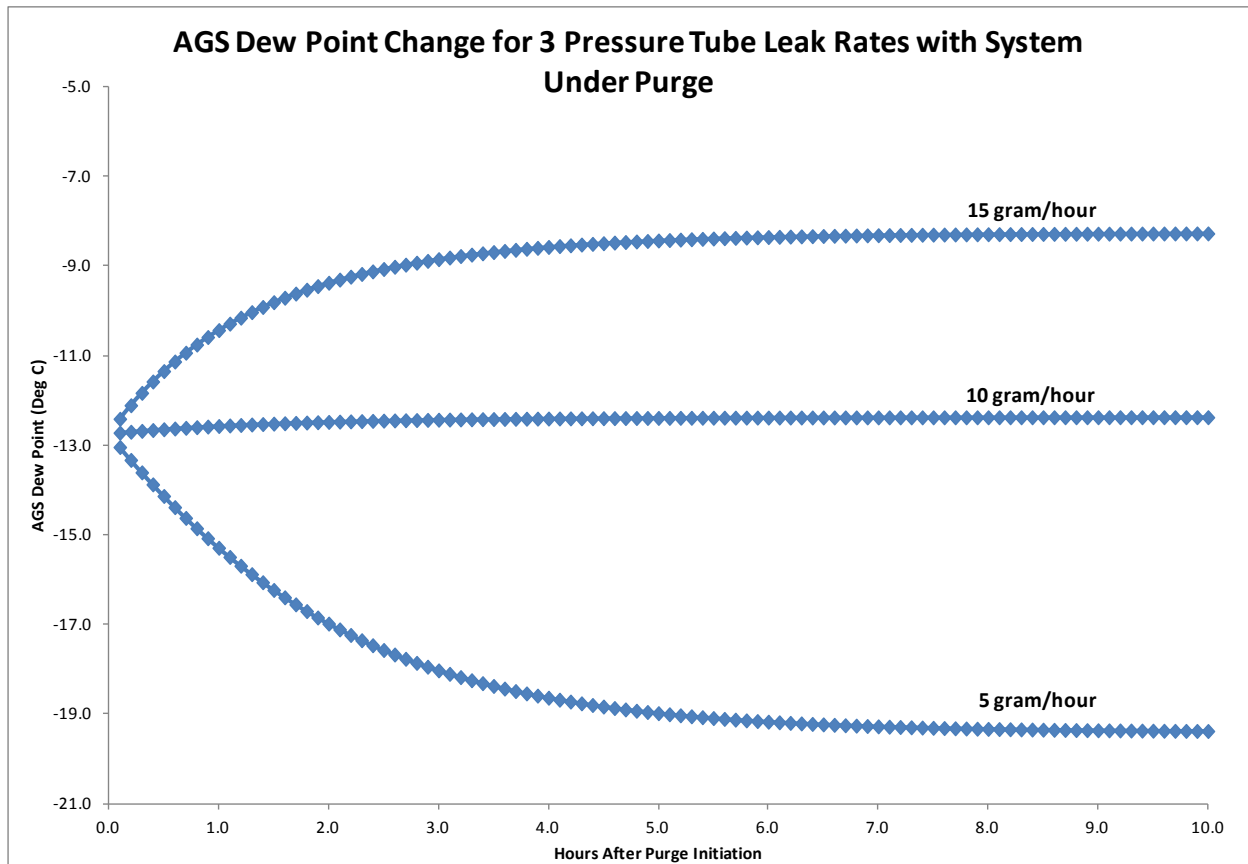
It is important to note that an AGS with an inter-purge period of less than about 6 hours is approaching a state known as “continuous purge” where moisture must be flushed from the system on a continuous basis. This situation prevails when the rate of  $\text{D}_2\text{O}$  ingress into an AGS approaches the rate at which the AGS compressors are capable of purging  $\text{D}_2\text{O}$  from the system.

CANDU station experience shows that operating under continuous AGS purge may indeed achieve the goal of maintaining an AGS dew point below  $-10\text{ }^{\circ}\text{C}$ , but this operating mode comes with the price of significant loss of pressure tube leak detection sensitivity. This is illustrated by Figure 2, below, which shows the results of calculations of the change to the system dew point induced by three different pressure tube leaks: 5 grams/hour, 10 grams/hour and 15 grams/hour. It was assumed that these leaks were into an AGS *in purge mode*. System behavior was modelled for intervals of 10 hours with an AGS in the following initial state:

AGS Volume:	12,000 litres
AGS Purge Flow:	2 litres/s
$\text{D}_2\text{O}$ Concentration:	1500 vpm
System Dew Point:	$-13\text{ }^{\circ}\text{C}$

Figure 2 clearly shows that the two smaller  $\text{D}_2\text{O}$  leaks – namely 5 g/hr and 10 g/hr – fail to induce a measurable increase in the system dew point. It is also noteworthy, that even for the highest  $\text{D}_2\text{O}$  leak rate of 15 g/hr, the rate of rise of the system dew point for a Unit under purge is never more than about  $2\text{ }^{\circ}\text{C/hr}$ . By comparison, for a dry AGS ( $\text{D}_2\text{O}$  concentration  $\sim 200$  vpm) in re-circulating mode, the initial dew point rate of rise for a 15 g/hr leak is greater than  $20\text{ }^{\circ}\text{C/hr}$ .

**Figure 2: Modelling of Dew Point Trends for an AGS under Purge**



## 7.0 Moisture Injection Testing of CANDU AGS

As we have seen, by 1990, CANDU AGS used CO<sub>2</sub> as the fill gas with 2 vol % O<sub>2</sub> added to maintain an oxidizing environment and prevent D<sub>2</sub> pick up at the OD surface of the pressure tubes. At the same time, pressure tube “leak-before-break” methodology requires continuous dew point monitoring of every fuel channel in all operating CANDUs for early detection of moisture ingress into an AGS.

The problems with annulus gas pigtail plugging observed in Pickering A Units in the mid-1980s prompted the AECB – Canada’s nuclear regulator at that time – to request moisture injection testing of new or refurbished CANDU Units to demonstrate their AGS exhibited a satisfactory dew point response. The first of these tests was carried out on Pickering Unit 1, starting in May 1987, [12], followed by Darlington Unit 2 in September 1989, [13], Pickering Unit 3 in July 1991, [14], and Bruce Unit 4 in November 1993, [15].

These tests used essentially the same methodology:

- (i) The AGS to be tested was first purged for several hours to a dew point of  $-30\text{ }^{\circ}\text{C}$  or lower
- (ii)  $\text{D}_2\text{O}$  vapor from a custom-built moisture generator was injected as a  $\text{D}_2\text{O}$ -saturated  $\text{CO}_2$  stream into the inlet of an AGS string
- (iii) Data from station and non-station (calibrated) dew point meters were recorded for periods up to 12 hours

$\text{D}_2\text{O}$  injection rates in the range 2 g/hr to 12 g/hr were used for most tests to simulate pressure tube leaks - the expectation being that for this range of leak rates the dew point would start to rise after a short delay period, (expected to be about 20 minutes), and increase at a predicted rate of  $x\text{ }^{\circ}\text{C/hr}$  for the duration of the test. The predicted values of  $x$  were calculated using a thermal-hydraulic network analysis computer code specific to the design of the AGS under test. The level of agreement between the measured and predicted values of  $x$  was used to gauge “the success” of each test.

Unfortunately, however, the results of many of these tests have revealed a number of unexpected problems:

- Most AGS exhibited a sluggish initial response, well beyond the expected 20 minutes
- Station dew point meters showed large differences in their readings compared to the well-calibrated instruments used to conduct the tests
- In the case of the Bruce Unit 4 tests, the formation of oxides of nitrogen in the AGS led to the production of nitric acid vapor which caused dew point meter drift, followed in some cases by meter failure

A sluggish AGS response was noted in the Darlington Unit 2 tests but was most extreme for Pickering Unit 3. This is illustrated by the fact that a  $\text{D}_2\text{O}$  injection rate of 5 g/hr into the P3 AGS was equivalent to the predicted response for a  $\text{D}_2\text{O}$  injection rate of only 1.25 g/hr. In addition:

- (i) The delay before the dew point started to rise was about 40 minutes compared to a predicted delay of 20 minutes

- (ii) The time to return the P3 AGS to a dry state after each test was more than 4 hours

In order to investigate the cause of the sluggish response of the P3 AGS, helium gas was injected into the system at a measured rate of 270 cm<sup>3</sup>/min and monitored at the system outlet. Helium was first detected at the outlet sampling point after a 20-min delay and reached a concentration ~ 3000 vpm after 2 hrs as expected. This test showed that the loss of D<sub>2</sub>O in the moisture injection tests was not caused by any leaks in the injection line but by water absorption within the AGS.

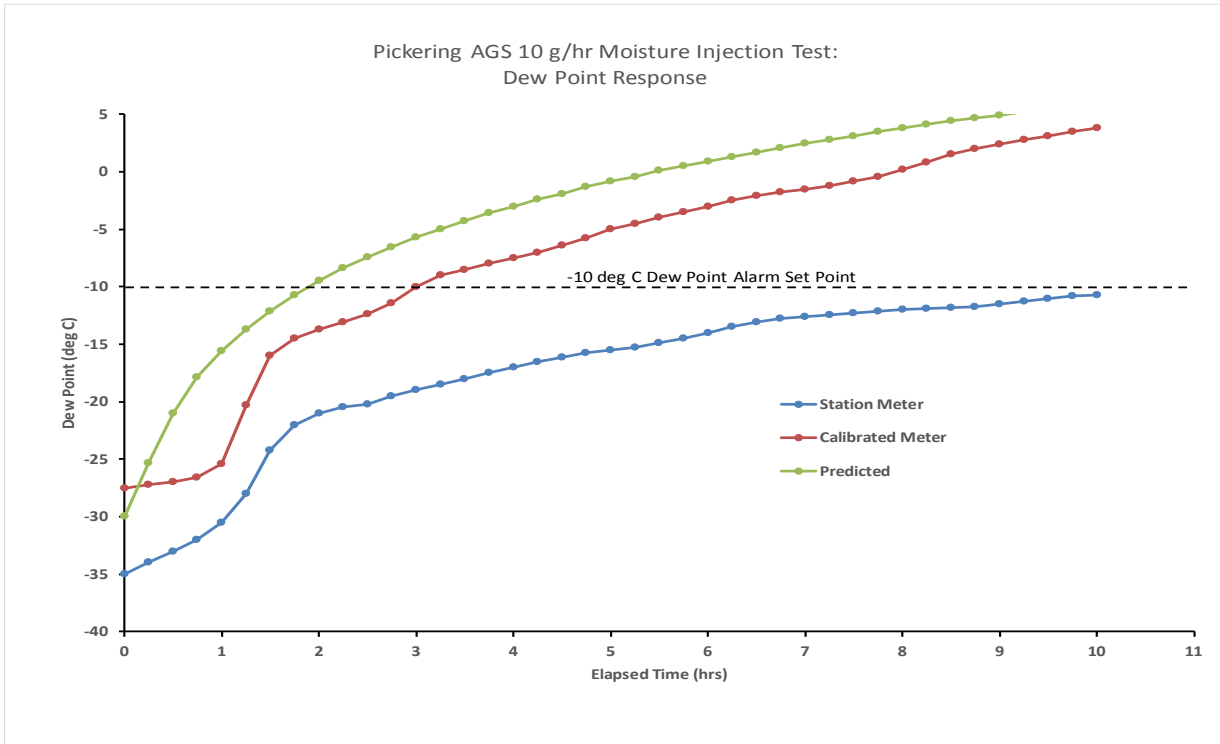
Problems with the calibration of the dew point meters installed at the stations compared to the instruments brought in for the moisture injection tests were also noted during the Pickering and Darlington tests. At Pickering for example, at the start of each test, the station's dew point meters were reading about - 35 °C when two calibrated instruments were reading - 30 °C. However, at the end of the tests, when the station meters were reading - 10 °C, the calibrated meters read 0 °C. At the time, these discrepancies were attributed to H<sub>2</sub>O/D<sub>2</sub>O isotope effects but subsequent investigations have not confirmed this proposal, [10].

Raw data from some of these moisture injection tests at Pickering and Darlington are presented in Figures 3 and 4, below. Each of these figures presents measured dew point readings for up to 10 hours while moisture was being injected into the AGS at a known rate, (10 g/hr for Pickering and 4.5 g/hr for Darlington), and includes three plots as follows:

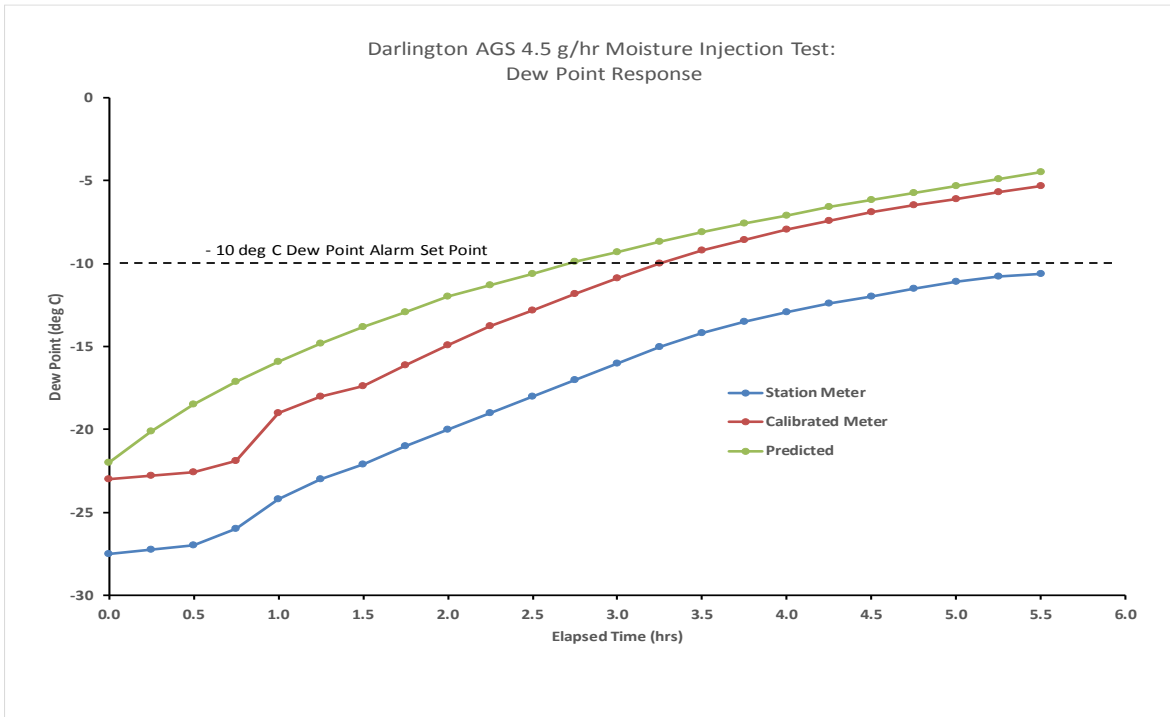
- (i) The average dew point meter readings from the two station Panametrics meters installed at the outlet of the AGS
- (ii) The dew point readings from a well-calibrated meter
- (iii) The predicted dew point readings for an ideal AGS under the known test conditions

The Figures show that there is generally reasonable agreement between the predicted dew point data and the data produced by a well-calibrated dew point meter. On the other hand, the station dew point meters generally read several degrees C low, and showed sluggish response. Indeed, as these Figures show, the station dew point meters failed to reach the -10 °C alarm set point in either of the tests. In addition, (not shown in the Figures), the measured rate of rise of the dew point in each test was about ½ of the predicted rate of rise. Nevertheless, the test results were deemed to be “acceptable” by the regulator.

**Figure 3. Data from a Moisture Injection Test on Pickering Unit 1**



**Figure 4. Data from a Moisture Injection Test on Darlington Unit 1**



## 8.0 Discussion

As the information presented in this paper demonstrates, CANDU AGS have been plagued with operational problems since their first practical realizations in Pickering A's four Units after their fuel channel replacements in the early 1980s. And it is important to recognize that a pressure tube leak-before-break detection capability was *not* included in the earliest CANDU reactors such as the NPD, but was a later design "add-on" introduced in response to the August 1<sup>st</sup> 1983 rupture of Pickering Unit 2 pressure tube G16.

Unfortunately, 30+ years of operating experience have shown that an AGS is *not* a very effective or reliable means of early pressure tube leak detection. In fact, leak detection that relies on AGS dew point monitoring is actually counterproductive because it offers a false sense of protection from a potentially catastrophic pressure tube rupture. It follows that faith in leak-before-break protection by AGS dew point monitoring is unwarranted because the leak detection methodology is flawed.

Nevertheless, it has been claimed that AGS leak detection by dew point monitoring *is* a proven methodology. Thus, in ref [16] we read:

*To avoid fast fracture due to rolled-joint cracking ... there is an Annulus Gas System (AGS), which is a leak detection system that is active at all times. It can detect any leakage from pressure tubes to meet Leak Before Break (LBB) requirements.*

Similarly, in Ref [17], we read that for CANDU pressure tubes:

*The time available to detect a leak and take action is 18 hours, (which) is much greater than the time required to detect (a pressure tube) crack.*

Unfortunately, a predicted value of 18 hours is contradicted by estimates made by other authors. Thus, in Ref [18] we read:

*The important question that the reactor operators need answering is "How much time is available to detect the leak and to take action before the crack becomes unstable?"*

The authors of Ref [18] estimate this time to be 35 hours, which they claim is "clearly adequate for operator action".

Nevertheless, in Ref [19] we have yet another estimate of the time to detect a pressure tube leak in a CANDU reactor:

*The LBB time of 8.8 hrs for the rolled joint area is about two times longer than the 4.3 hrs for other (pressure tube) areas. Considering 2.6 hrs to detect the initial leaking, the minimum action time recommended for the operator is 1.7 hrs.*

The wide range of leak detection times predicted by these various authors clearly reveals there is a high level of uncertainty and/or disagreement as to what is an appropriate value for this important parameter. Nevertheless, CANDU regulators appear to be satisfied with the current pressure tube leak detection methodology because it is a *fait accompli* that moisture injection testing has been able to verify the adequacy of a Unit's leak detection capability.

Unfortunately, however, moisture injection tests are not carried out under conditions that would be present in an AGS at any time during an actual pressure tube leak:

- (i) AGS testing has only been carried out on newly commissioned or refurbished Units when an AGS is likely to be clean and leak tight. However, in older Units, water absorbing corrosion product builds up in the AGS leading to an increasingly sluggish dew point response.
- (ii) Before each test, the AGS is purged to a dew point of  $-30\text{ }^{\circ}\text{C}$ , or lower. However, as an AGS ages, purges are required more frequently and the *average* AGS dew point steadily increases. Thus, as time goes by, a pressure tube leak is more and more likely to occur in an AGS that is already wet, and in extreme cases (see below), an AGS may be so leaky that it cannot be purged to a dew point of  $-30\text{ }^{\circ}\text{C}$ .
- (iii) AGS tests are generally carried out using two modes of moisture injection: external and internal. The external injection test results can be misleading because the injected moisture reaches the dew point meters via the shortest annuli strings. Conversely, internal injection involves moisture injection into only one pre-selected string and it is only the response of this string that is actually being tested.

- (iv) Most AGS exhibit some level of CO<sub>2</sub> out-leakage from system components such as end fitting attachment rings; this requires continuous CO<sub>2</sub> makeup to the AGS, which in extreme cases has been as high as 80 liters/min. On the other hand, air ingress into an AGS is also a regular occurrence. Ingressed air undergoes radiolysis leading to the formation of highly corrosive NO<sub>2</sub> gas. This species is known to be very deleterious to a dew point meter's sensor and results in instrument reading "drift" and a slow loss of sensitivity.
- (v) The amount of moisture that can be measured in an operating AGS by *in situ* dew point measurements is actually quite limited because condensation of water on cooler surfaces in an AGS circuit occurs for water injection rates above 26 g/hour – (See item 4 below). However, as shown in Section 6.0, leaky AGS exhibit "background" water ingress rates as high as 4 g/hour. Thus, pressure tube leaks that are detectable by dew point measurements span a very narrow range of water ingress rates – namely, between 4 and 26 g/hour.

In order to illustrate the magnitude and significance of these problems it is useful to consider some examples of *real* CANDU AGS performance under four distinct operating conditions, namely: (i) Newly commissioned; (ii) Mature; (iii) Extreme (iv) In the event of a pressure tube leak. Examples of AGS operation under each of these conditions are discussed below:

### **1. Newly Commissioned AGS Performance**

Immediately after the commissioning of a new or refurbished AGS, the system should be "leak tight" and, after an initial purge, be very dry and show a negligible buildup of moisture. There are however, *two* practical limits on AGS performance:

- The AGS CO<sub>2</sub> supply is specified to have a dew point better than – 60 °C, so this level of post-purge dryness is theoretically attainable in a totally leak tight system.
- The time between purges in a completely leak tight AGS is ultimately limited by the unavoidable permeation of D<sub>2</sub> gas through the fuel channel end fittings as described in Section 4.3 of this report. About 70 % of this D<sub>2</sub> is converted to D<sub>2</sub>O by the reverse water gas shift reaction – an amount that increases to 100 % conversion if a few percent of O<sub>2</sub> is added to the CO<sub>2</sub>. For the permeation rates observed in CANDU AGS, the theoretical



minimum D<sub>2</sub>O ingress rate is about 0.1 g/hour which implies an upper limit of about 10 days between AGS purges.

In practice, new or newly refurbished reactors have approached, but never achieved this level of performance. However, in the early 1990s, when the first generation of CANDU 6 reactors was 10 years old, Point Lepreau and Gentilly-2 had purge periods in the range of 8 to 9 days. Similarly, Bruce B and Darlington Units achieved purge periods in the range of 6 to 8 days in their first decade of operation. Nevertheless, as shown below, this level of performance has proven to be unsustainable in older Units.

## 2. Mature AGS Performance

We shall refer to a CANDU Unit that has operated more than 20 years, or let's say beyond 140,000 EFPH, as a *mature* CANDU Unit. Bruce Unit 3 in 2004 would be a good example with 148,000 EFPH of operation at that time. Figure 5, below, provides some Bruce Unit 3 AGS purge rate data. The data show this Unit had an average purge period of 4.5 days and an average dew point ~ -15 °C.

**Figure 5: Plot Showing Purge Frequency of a Mature CANDU Unit**

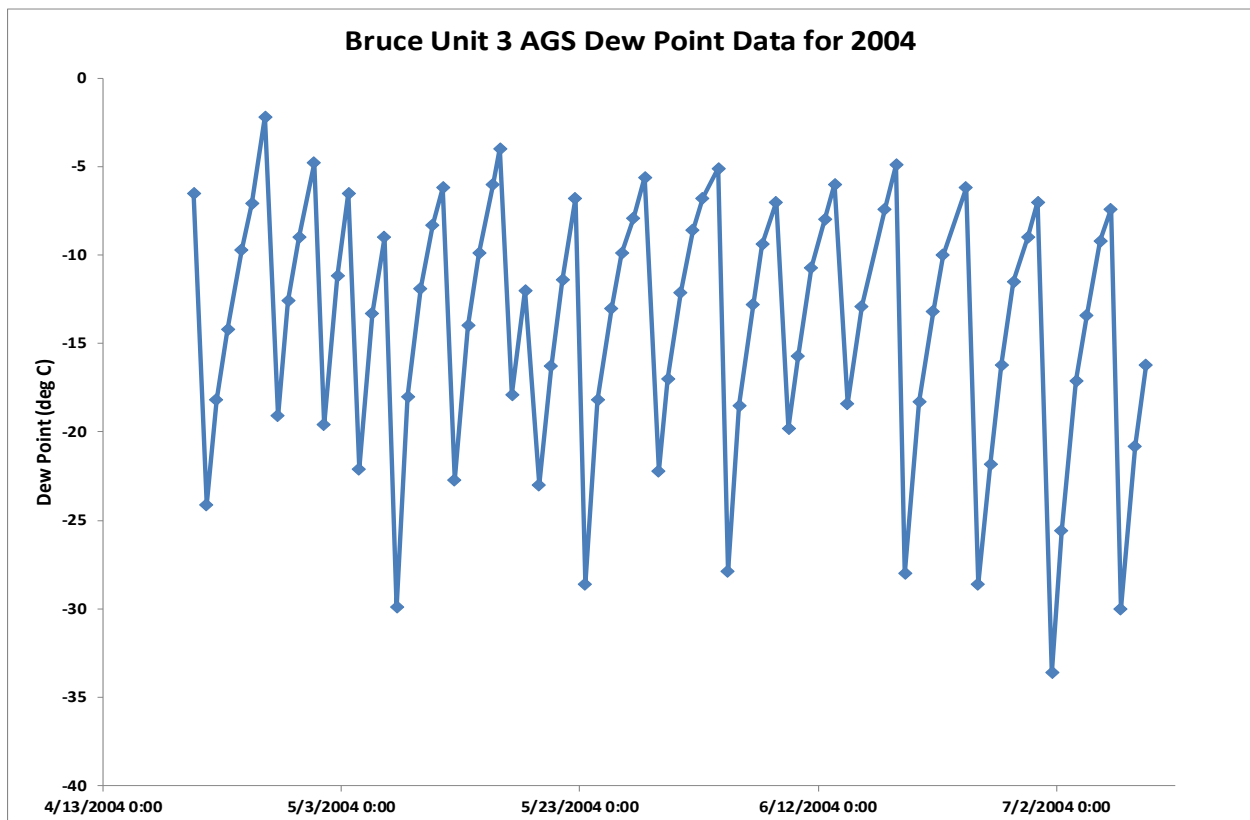
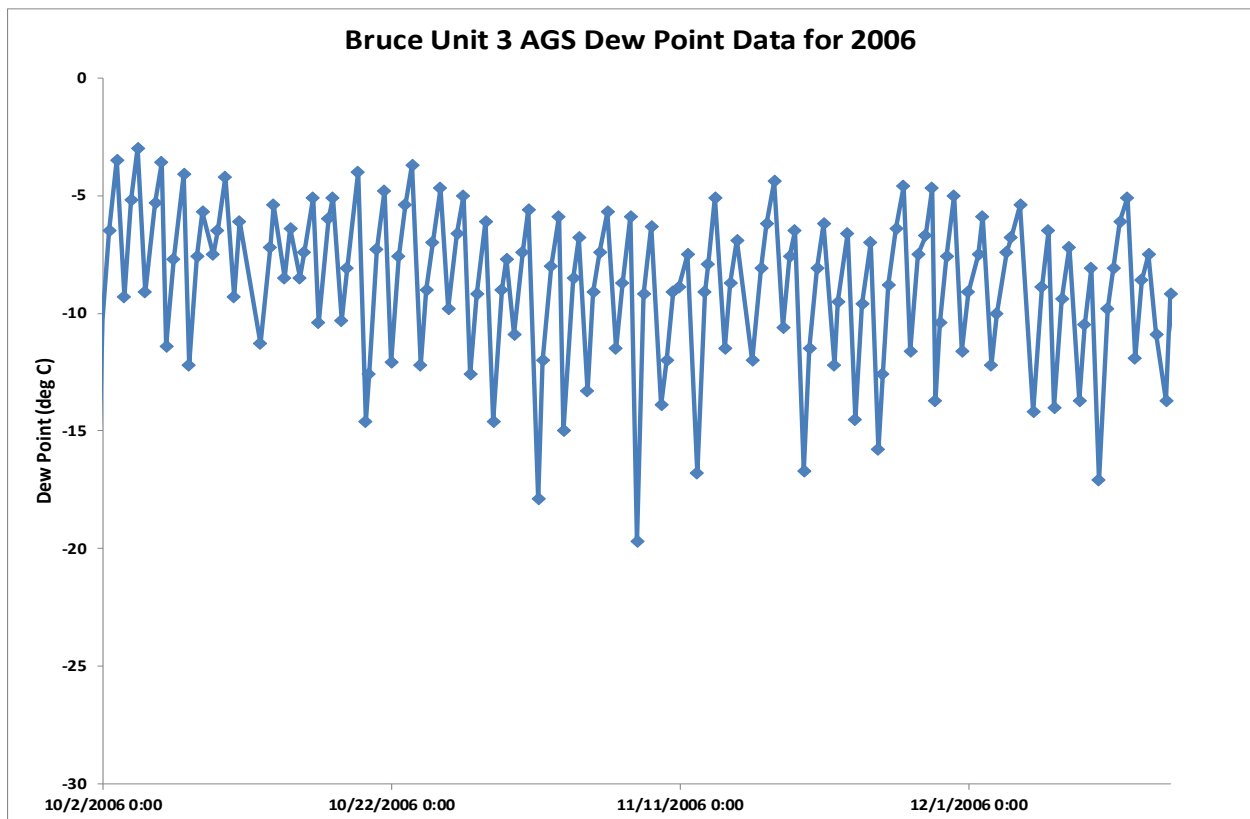


Figure 5 is only one example of mature AGS performance, but as a general rule: an AGS becomes leakier as it ages. This is shown in Figure 6, which is an example of the 2006 performance of Bruce Unit 3, and should be compared to the 2004 performance of this Unit, shown in Figure 5.

**Figure 6: Plot Showing the Purge Frequency of a Very Mature AGS**



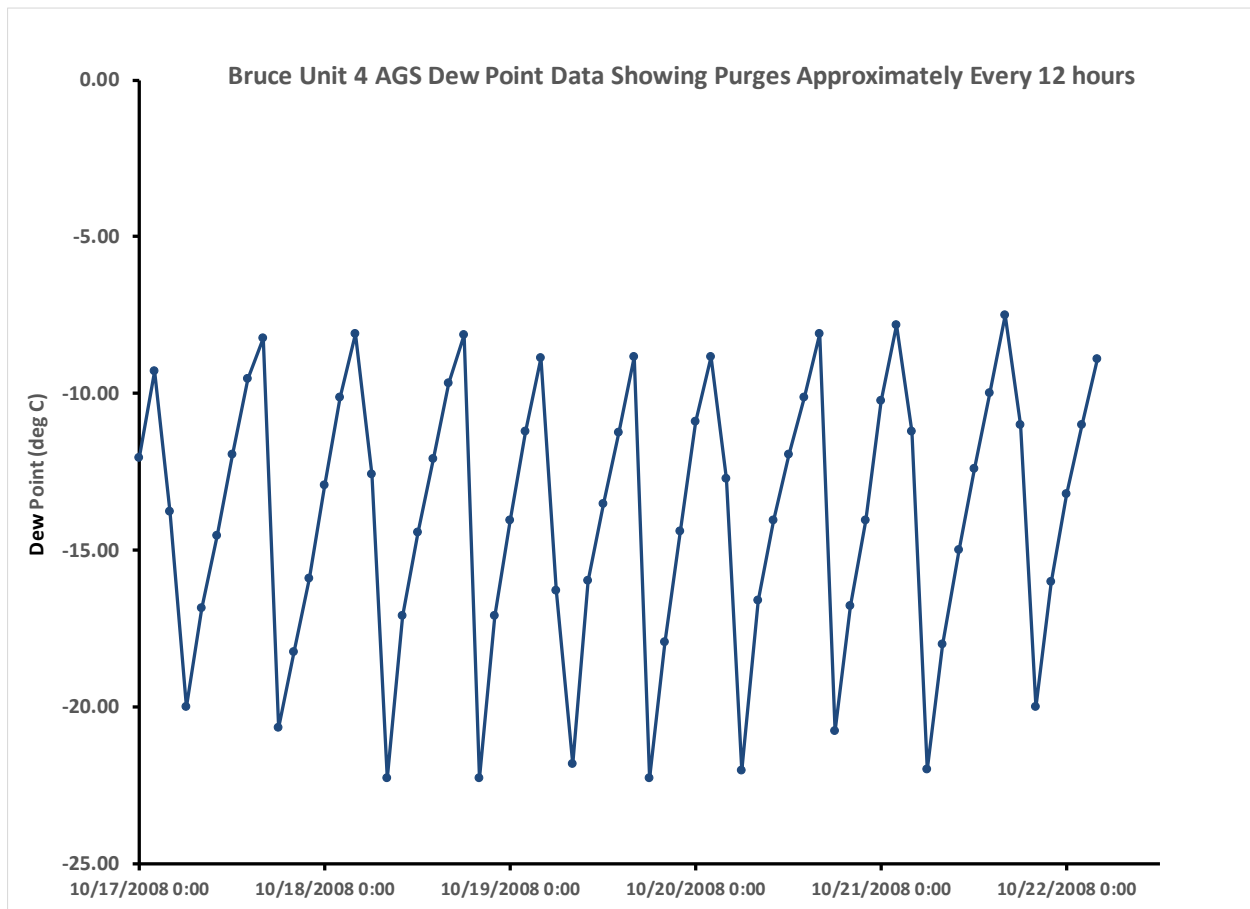
The data in Figures 5 and 6 show that, over a two-year interval, the Bruce Unit 3 AGS performance deteriorated as follows: Its 2006 average purge period was less than 2 days, compared to 4.5 days in 2004, and its *average* dew point was  $-11\text{ }^{\circ}\text{C}$ , compared to  $-15\text{ }^{\circ}\text{C}$  in 2004.

### 3. Extreme AGS Performance

The examples of AGS performance shown above for Bruce Unit 3 are typical of mature, as well as very mature CANDU Units – namely, purge periods in the range 5 to 2 days. Nevertheless, there are cases where an AGS has been operated with

moisture levels that required inter-purge periods of less than 1 day. Thus, Figure 7, below, provides an example of an AGS, (Bruce Unit 4 in October 2008), with an inter-purge period of only 12 hours, [20].

**Figure 7: An Example of an AGS with a High Purge Frequency**

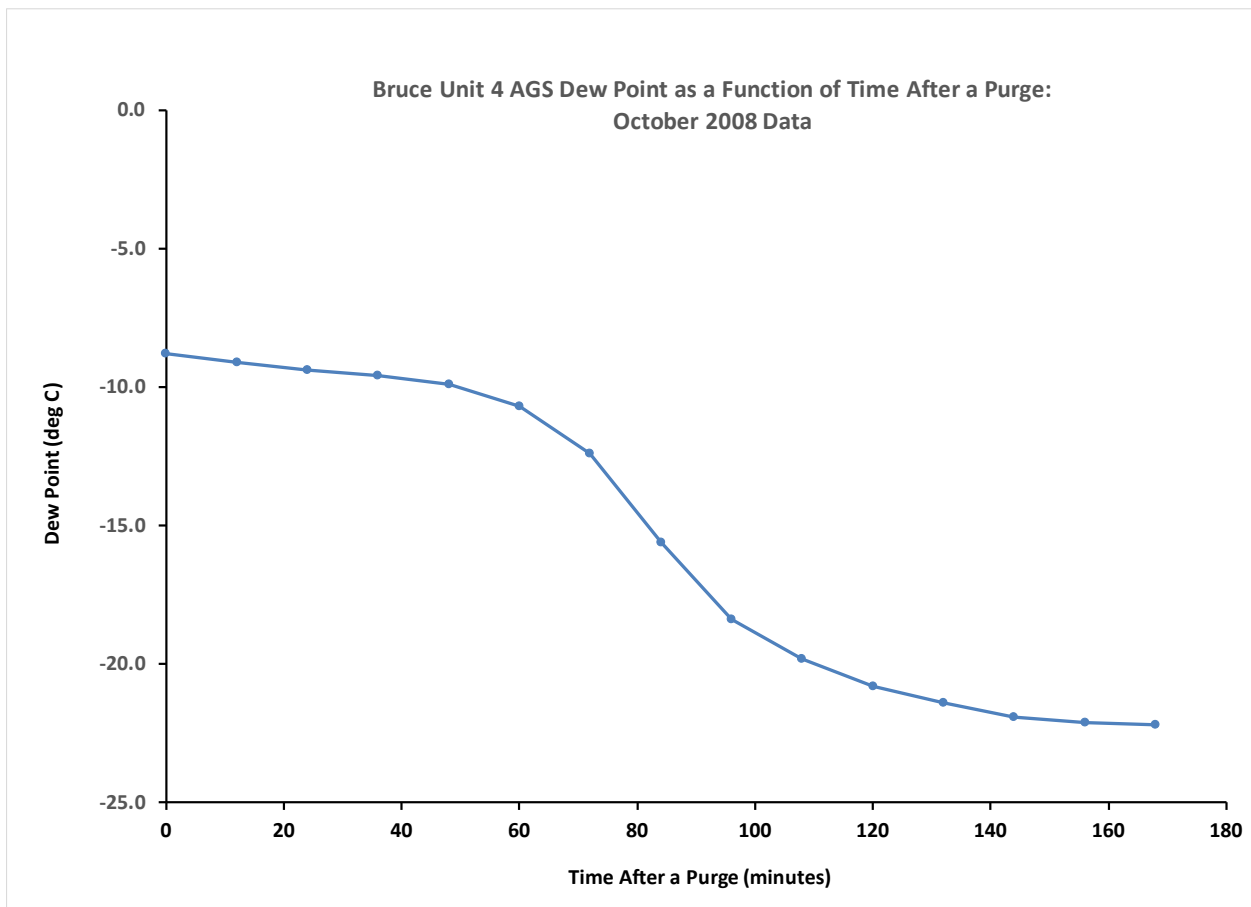


It is important to note that the data points plotted in Figure 7 are for intervals of only 2 hours, (compared to the data points in Figure 5, which are 24 hours apart). These and similar data show that in October 2008 the Bruce Unit 4 AGS spent over 30% of its operating time at dew point readings *above* (i.e. *wetter than*) the specified purge point of  $-10\text{ }^{\circ}\text{C}$ . Indeed, if this Bruce Unit was purged when the dew point reached  $-10\text{ }^{\circ}\text{C}$ , as specified, it would have required purging every 9 hours.

Conversely, Figure 7 also shows that in October 2008 the Bruce Unit 4 AGS was never purged to its specified lower dew point limit of  $-30\text{ }^{\circ}\text{C}$  but was returned to

recirculating mode when the dew point had dropped to only about  $-22\text{ }^{\circ}\text{C}$ . The fact is, purging an AGS can only bring the moisture level down to a dew point where the rate of removal of  $\text{D}_2\text{O}$  is equal to the rate of  $\text{D}_2\text{O}$  ingress. This represents an AGS *equilibrium state* which, in the case of Bruce Unit 4 in 2008, corresponded to a dew point of about  $-22\text{ }^{\circ}\text{C}$ . This dew point was attained after about 2 hours of purging and an additional hour of purging was unable to dry the Unit 4 AGS significantly closer to its  $-30\text{ }^{\circ}\text{C}$  target dew point, as shown in Figure 8, below

**Figure 8: AGS Dew Point as a Function of Time After Purge Initiation**

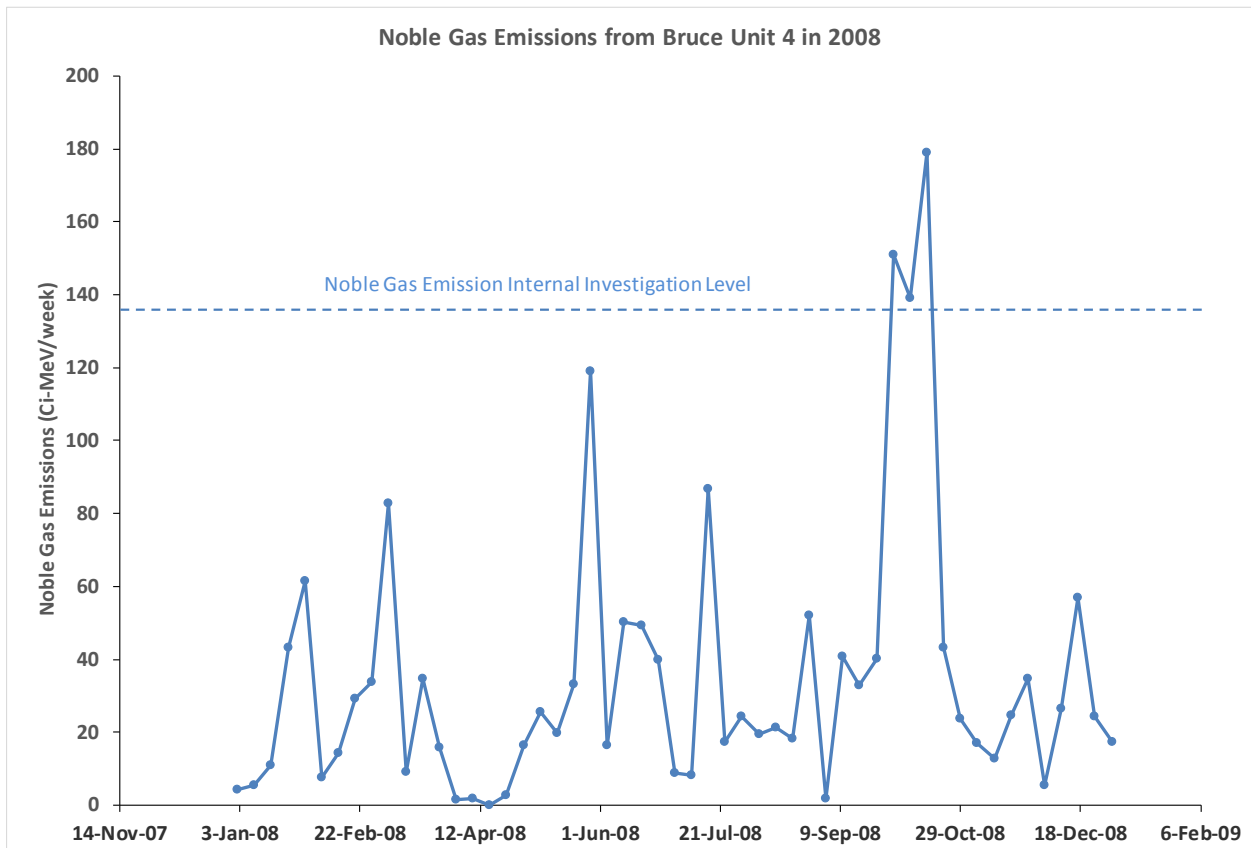


There is an additional concern when an AGS is operated for an extended period of time with a high purge frequency and that is the fact that AGS purging is often associated with noble gas emissions and the release of Ar-41 to the environment. Air contains 78 %  $\text{N}_2$ , 21 %  $\text{O}_2$  and 0.93 % Ar. Natural argon is 99 % Ar-40 which is rapidly converted to radioactive Ar-41 under neutron irradiation. There are a number of circumstances that allow the ingress of air into an AGS including:

inadequate flushing of gas supply lines; insufficient CO<sub>2</sub> make-up gas; compressor malfunction or failure; PHTS D<sub>2</sub>O degassing when dissolved Ar-41 is high (e. g. > 100 μCi/liter). Neutron activation calculations show that the presence of only 200 vpm of argon in an AGS prior to purging leads to a maximum theoretical release of about 140 Ci-MeV of Ar-41.

The 2008 airborne noble gas emissions from Bruce Unit 4 are plotted in Figure 9 below, and show that the station's internal investigation level for noble gas emissions, (138 Ci-MeV), was exceeded in October during the previously noted period of high AGS purge frequency. Perhaps the only positive outcome stemming from the high purge frequency of Bruce Unit 4 in October 2008 is that the source of the D<sub>2</sub>O ingress was eventually identified, (a number of leaking pressure tube end fitting rolled joints,) and fixed in November 2008.

**Figure 9: Noble Gas Emissions Associated with the Bruce Unit 4 High Purge Frequency in October 2008**



There is an additional issue with regard to radioactive species in an operating AGS, and that is the buildup of other noble gas radionuclides besides Ar-41, albeit at much lower concentrations. The most important of these noble gas species are the uranium fission products Kr-85m, Kr-87, Kr-88, Xe-133, Xe-135 and Xe-138. All of these radionuclides have been detected by gamma spectrometry of AGS grab samples collected at high and low dew points, as shown by the CANDU 6 data in Table 2, below:

**Table 2: CANDU 6 AGS noble gas (excluding Ar-41) concentration data**

AGS Dew Point	Noble Gas Radionuclide Concentration ( $\mu\text{Ci/litre of gas}$ )					
	Kr-85m	Kr-87	Kr-88	Xe-133	Xe-135	Xe-138
-35 °C (Dry)	0.58	1.19	1.06	0.72	0.72	4.26
-15 °C (Wet)	0.74	1.59	1.51	2.3	0.80	7.64

There are two potential sources of these radionuclides:

- (i) Degassing of PHTS D<sub>2</sub>O that has entered the AGS through a chronic pressure tube leak, e.g. a rolled joint leak
- (ii) Fission product release by *in situ* irradiation of surficial uranium on pipework within the AGS

Item (i) is a possible source of annulus gas radioactivity because noble gas radionuclides are always present in a reactor's primary heat transport D<sub>2</sub>O at concentrations up to 100  $\mu\text{Ci/kg D}_2\text{O}$  from irradiation of so-called "tramp" uranium. Similarly, item (ii) must also be considered because mass spectrometric analysis of pressure tube OD surfaces shows the presence of uranium at concentrations in the range 0.1 to 0.5  $\mu\text{g/cm}^2$  and noble gas releases from this source should therefore be considered. Nevertheless, if the first of these sources predominates over the second, it offers a novel means of detecting D<sub>2</sub>O ingress into an AGS that could supplement or even replace leak detection by dew point monitoring. The analysis of tritium, (by liquid scintillation counting), in AGS grab samples or collected in cold-fingers could further enhance D<sub>2</sub>O leak detection.

#### 4. AGS Performance during Moisture Injection Tests compared to its Predicted Behavior in the event of a Pressure Tube Leak

As discussed in Section 2.0 of this report, a real life pressure tube leak is predicted to result in D<sub>2</sub>O entry into an AGS at a rate of at least 1 kg/hr. By comparison, moisture injection testing of AGS at Pickering, Bruce, Darlington and Wolsong have been carried out at moisture injection rates less than 15 g/hr. The main reason for using such low moisture injection rates during AGS testing is to avoid water condensation which would provide a dew point reading that would not be commensurate with the amount of D<sub>2</sub>O in the system.

The maximum amount of moisture that can be injected into an AGS without causing condensation depends on the CO<sub>2</sub> flow rate and more importantly on the lowest temperature in an AGS circuit, (which is typically about 60 °C). The coolest region of an AGS is along the end shield at the reactor face and D<sub>2</sub>O vapor from a leak, simulated or otherwise, has to pass through at least one end shield section on its way to a dew point meter.

The leak rate which just saturates the CO<sub>2</sub> in a leaking channel (at the end shield temperature) is referred to as the “maximum discernable” leak rate. With two AGS compressors operating, (giving a total 5.7 l/s flow rate), this is about 26 g/hr. For leaks greater than 26 g/hr moisture will condense in the 3/8 in. outlet tubing and drain into a D<sub>2</sub>O recovery system which includes liquid water detectors known as “beetles”. Each beetle consists of a pair of sealed and insulated electrodes that announce when liquid water spans the gap between the electrodes and an alarm signal is automatically sent to the control room.

In the event of a pressure tube leak greater than 26 g/hr, water will accumulate in each lattice tube between the leak and the nearest beetle, and a leak occurring in the *upstream* end of a 12 annuli string will result in the longest delay time to a beetle alarm annunciation. Furthermore, it may be calculated that each channel is capable of collecting about 1 kilogram of D<sub>2</sub>O in out-of-core locations, e.g. the annulus gas bellows, so that in the case of a 12 annuli string, up to 12 kg of D<sub>2</sub>O can accumulate in an AGS *before a beetle alarms*. For a through-wall crack in a pressure tube, leak rates of at least 1 kg/hr are predicted and a beetles’ response time (at an AGS flow rate of 5.7 l/s) would be as much as 12 hours, or even longer at lower AGS flow rates.

Also in the event of a rapid rate of rise of an AGS dew point, the station's AGS Design Manual instructs the operator to confirm the presence of a suspected pressure tube leak through an AGS beetle alarm annunciation, see for example Ref [11]. However, an AGS beetle may take up to 12 hours to annunciate after the first indications of D<sub>2</sub>O ingress into an AGS, leading to two questions: How long does it take for a pressure tube crack to grow to the critical size for unstable (fast) rupture? And, would there be enough time to safely shut down the reactor? The answers to these questions depend on an evaluation of two factors:

1. The crack velocity: CSA Standard N285.8 provides an equation for the axial crack growth velocity,  $V_o$  for irradiated Zr-2.5Nb pressure tubes as a function of temperature and neutron fluence. The equation shows that  $V_o$  increases as the temperature and neutron fluence increase. An upper bound on  $V_o$  at the hot (outlet) end of a pressure tube irradiated to a fluence  $> 10^{26}$  n/m<sup>2</sup> is  $\sim 2 \times 10^{-6}$  m/s.
2. The critical crack length, CCL: Unfortunately, very few data exist on CCLs for irradiated CANDU pressure tubes, although a working value  $\sim 80$  mm is suggested in the Bruce B AGS Design Manual, [11].

Combining these values for  $V_o$  and CCL, we arrive at a value of 11 hours for the time to unstable rupture, which is 1 hour *less* than the 12-hour interval estimated for an AGS beetle alarm. However, it is important to remember that this 12-hour alarm time is based on *two* AGS compressors operating with a total flow rate of 5.7 l/s, which is not always the case. Thus, in 2011 Darlington NGS was operating a Unit at an AGS flow rate of only 1.64 l/s, and the leak detection time was estimated to be over 20 hours. In commenting on this situation, the CNSC stated that a 1.64 l/s flow rate was unacceptable and should be increased to at least 2.5 l/s to provide an adequate leak detection capability for this Unit.

It is important to add that some of the ASG moisture injection tests discussed in this report did, in fact, include moisture injections at high D<sub>2</sub>O delivery rates – e.g. rates  $> 2$  kg/hr. These high injection rates were designed to test the response of AGS beetles to a simulated pressure tube leak and resulted in beetle alarms within 1 hour of the initiation of moisture injection. As such the tests were considered to be a satisfactory demonstration that the beetles would respond as expected when a large ( $> 2$  kg/hr) leak developed. However, as discussed below, these test results are actually of no value in assessing how an AGS would respond in the event of a *real* pressure tube leak because of the mode of moisture injection used in the tests.



Thus, in the 1987 Pickering tests, D<sub>2</sub>O was injected as a liquid into an AGS outlet header at a rate of about 2 kg/hr while the Unit was hot. The D<sub>2</sub>O was vaporized in the ~150 °C CO<sub>2</sub> as it passed through the reactor core and was subsequently condensed in the cooler lines leading to the AGS beetles. However, the problem with this test procedure is that the injected D<sub>2</sub>O was vaporized into *all* 390 channels of the AGS which meant that the associated pressure buildup was shared over the entire AGS volume of about 12,000 liters.

Thus, consider the example of 2 kg of D<sub>2</sub>O injected into an AGS over a period of 1 hour. Upon vaporization, this mass of D<sub>2</sub>O would generate 3470 litres of gas at 1 atmosphere pressure and 150 °C, or about 29 % of the total AGS volume. It follows that, in the absence of leaks, ingress of 2 kg of D<sub>2</sub>O would increase the AGS pressure by about 29 %. An AGS can handle such an increase in pressure over an hour or so, through pressure relief valves and rupture disks.

In the case of a *real* pressure tube leak with the same D<sub>2</sub>O ingress rate of ~ 2 kg/hr, we have an entirely different situation because D<sub>2</sub>O now enters the AGS via *one* channel which has an effective volume of only 30 litres. Furthermore, the gas flow rate through a channel is as low as 0.054 l/s and the ends of each channel annulus have an effective cross-sectional area of only ~ 2.5 cm<sup>2</sup> due to the tight-fitting bearing journals and sleeves at the end of each channel. In addition, after exiting a channel, the annulus gas CO<sub>2</sub> passes through a second narrow bearing before entering a metal bellows assembly. Gas exits these bellows through a 0.2-inch ID stainless steel tube, (known as a pigtail), which also provides the approximately 1-meter long interconnecting tubing to the next AGS channel. The question then arises: how will this tight-fitting arrangement of concentric tubing behave in the event of a pressure tube leak?

To answer this question, consider a single channel annulus in which a pressure tube through-wall crack starts to leak with a D<sub>2</sub>O ingress rate of 20 g/min. The in-leaking D<sub>2</sub>O would flash to steam and, assuming no change in the AGS temperature, (which starts at ~150 °C), the volumetric ingress rate would be 34.7 litres/min. However, because the heat transport D<sub>2</sub>O would be entering the AGS at a temperature of at least 260 °C, the volumetric ingress rate would be closer to 40 litres/min. Furthermore, as the channel temperature increases, the end fitting body and adjacent bearing journal would expand outward and reduce the bearing clearances.

Measurements of Darlington end fitting bearing discharges under simulated pressure tube leak conditions have shown that at reactor operating temperatures, (~260 °C), the discharge of water through a channel bearing assembly is reduced by a factor of about two compared to the cold (room temperature) rate, [21]. This flow rate anomaly illustrates the complexities involved in predicting how an AGS dew point will respond to a real pressure tube leak.

## 9.0 Conclusions

This paper reviews the attributes and performance characteristics of pressure tube leak detection systems employed in the current generation of CANDU reactors. At the present time, all CANDU pressure tube leak detection systems are based on dew point measurements at the outlet end of the annulus gas system, (AGS). The many issues raised reveal generic problems with pressure tube leak detection – even in newly refurbished Units – due to a combination of annulus gas flow anomalies, localized water condensation and limitations in dew point meter performance. In addition, many of these problems only get worse as a reactor ages and the leakiness and purge frequency of an AGS increases.

On this particular point, the CNSC recently noted that the tendency for an AGS flow rate to *decrease* as a Unit ages, with a corresponding *increase* in the time to detect a leak, must be countered by placing more restrictive limits on Unit operation. This is necessary because older Units are more prone to pressure tube leaks making early leak detection of paramount importance in ensuring that safety margins are not being eroded in older CANDU Units.

This paper also draws attention to a number of important generic issues concerning AGS dew point measurement and AGS moisture injection testing:

(i) Dew point meters work well in relatively dry environments when the moisture content is less than 1000 vpm, (dew points below – 10 °C). However, dew point meters fail to function at water concentrations greater than about 10,000 vpm, (dew point readings above 10 °C). At this moisture level the AGS is in a state of water vapor saturation that leads to liquid condensation. This moisture level would be reached within 0.5 hour by a D<sub>2</sub>O leak of 150 g/hr, and is close to the predicted early leak rate from a 30 mm pressure tube through-wall crack.

(ii) Annulus gas dew point measurements are meaningful only over a very limited range of D<sub>2</sub>O leak rates – between about 4 and 26 g/hr. As noted above, at leak rates greater than 26 g/hr, water will condense on the cooler surfaces in an AGS

circuit and the dew point readings will be essentially “off scale” and of no further use to the station operator.

(iii) The currently accepted method for certifying a CANDU Unit’s pressure tube leak detection capability – namely, an AGS moisture injection test with a D<sub>2</sub>O simulated leak rate of about 10 g/hr – totally fails to represent conditions that are likely to be present in an AGS in the event of a *real* pressure tube leak.

CANDU reactor licensees operate under CSA standard N285.8 which requires a leak-before-break assessment of pressure tubes in the event of delayed hydride cracking. Furthermore, this standard states that LBB assessments must demonstrate that the pressure tube leak detection system provides the operator with sufficient time to shut the reactor down before a crack grows to its critical crack length.

Nevertheless, standard N285.8 fails to set any requirements regarding the performance and inspection/testing frequencies of an AGS “leak detector”. On the contrary, CSA N285.8 simply assumes that a pressure tube leak will be detected in a “timely manner”, presumably under all operating conditions and regardless of the years of service of the AGS.

However, this ignores the reality that an AGS develops a number of undesirable characteristics as it ages including loss of dew point sensor sensitivity, reduced gas flow rates accompanied by increased leak detection times, increased water absorption from the accumulation of corrosion product, etc. In addition, the limited dynamic range of a station’s dew point meters means they “go blind” at water injection rates above about 26 g/hr. By comparison, D<sub>2</sub>O leak rates at the start of a pressure tube through-wall crack are predicted to be at least 1 kg/hr.

It is concluded that pressure tube leak detection based on dew point measurement is not an effective or reliable means of satisfying the Leak Before Break requirement for safe operation of a CANDU reactor. In this regard, enhancing the role of the AGS beetles, especially in confirming high rates of water ingress, has some merit but it would be necessary to ensure that the beetles’ unavailability meets an acceptable target, (such as < 0.01). Serious consideration should also be given to abandoning dew point measurements altogether and using alternative leak detection techniques such as liquid scintillation counting and gamma spectrometry of gas and/or cold-finger samples.

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### **Abstract**

In the event of a pressure tube leak from a small through-wall crack during CANDU reactor operations, there is a regulatory requirement – referred to as Leak Before Break (LBB) – for the licensee to demonstrate that 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 fast-uncontrolled rupture. In all currently operating CANDU reactors, worldwide, this LBB requirement is met via continuous dew point measurements of the CO<sub>2</sub> gas circulating in the reactor's Annulus Gas System (AGS). In this paper the historical development and current status of this leak detection capability is reviewed and the use of moisture injection tests as a verification procedure is critiqued. It is concluded that these tests do not represent AGS conditions that are to be expected in the event of a real pressure tube leak.

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## Additional Material for the Submission by F. R. Greening

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

$$t = \frac{CCL - L_p}{2V_o} \dots\dots\dots(i)$$

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

$V_o$  is the crack velocity

Equation (i) has *three* unknown variables – CCL,  $L_p$  and  $V_o$  – 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<sub>2</sub>O and neutron irradiations up to  $8 \times 10^{25}$  n/m<sup>2</sup> and span the range from 40 to 90 mm. Similarly, the same report provides values for  $V_o$  in the temperature range 150 – 280 °C. These data show that  $V_o$  is strongly dependent on the pressure tube temperature, but even for a *fixed* temperature such as 250 °C, the values of  $V_o$  span the range  $1 \times 10^{-7}$  to  $6 \times 10^{-7}$  m/s, (equal to 0.36 to 2.16 mm/hr).

More recent research has confirmed most of these values for CCL and  $V_o$ . 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  $\approx 41$  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  $1 \times 10^{-7}$  to  $4 \times 10^{-7}$  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  $\approx 4$  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, or about  $7W$ , 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:

$$t = \frac{CCL - L_p}{2V_o}$$

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

$$t = 4.5 \text{ hours}$$

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.* So, I would ask the CNSC: Are *any* of these estimates science-based?

As described in CSA N285.8, once a pressure tube crack starts leaking it is mandated that, as a first step in a leak-before-break (LBB) assessment, the leak rate for a given crack length should be determined. To this end, leak rate data from the Chalk River Active Crack Leakage Experiment (CRACLE) are generally used – See report by C. Coleman et al: *Evaluation of a Leaking Crack in an Irradiated CANDU Pressure Tube.*”, AECL Report No: AECL-9733, issued June 1988.

The CRACLE data show D<sub>2</sub>O leak rates are poorly correlated with crack lengths; nevertheless, researchers have used what they considered to be “best fits” to various mathematical functions to conduct quantitative leak-before-break analyses. For example:

- In the Bruce B AGS Design Manual, (See Bruce Power document DM-NK29-34980/63498, issued 1991), we have a *linear* functional relationship proposed for the time evolution of the leak rate Q, measured in kg/hr, vs. the through-wall pressure tube crack length L, measured in mm:

$$Q = 0.614L - 16.58$$

where it is assumed that L = 27 mm when Q = 0

- In a report by K.M Nho et al. entitled: “*Assessment of Leak Detection Capability of CANDU 6*”, Journal of the Korean Nuclear Society, Vol 30 (5), 405, (1998), we have another linear relationship, (using the same notation as above):

$$Q = 0.7618L - 14.86$$

where it is assumed that L = 19.5 mm when Q = 0

- However, CSA Nuclear Standard N285.8 recommends a *cubic* leak rate equation:

$$Q = 0.0014L^3 - 11.2$$

Where, L = 20 mm when Q = 0

The lower bound (95% confidence) of this cubic relationship is shown in an overhead from a September 2002 presentation by Doug Rogers, AECL’s Director of Fuel Channel Research.



CANDU reactors are claimed to have “layers” of safety systems in place, giving them “defence-in-depth” against catastrophic accidents.

The safety of a reactor largely depends on the integrity of its pressure-boundaries such as feeder pipes and other heat transport system components. It is also crucial to be able to accurately predict how these boundaries behave in the event of failure.

For pressure tubes, the key element of its “defence-in-depth” is its predicted leak-before-break, (LBB), behavior in the event of a through-wall crack failure.

The “failure” of a pressure tube can range from a minor leak of ~ 1kg D<sub>2</sub>O per hour or less, to a catastrophic rupture involving the discharge of > 100 kg of D<sub>2</sub>O per second.

The former type of leak occurs when a small, *slowly growing crack* first penetrates the 4-mm wall-thickness of the tube; the latter is due to a *fast-uncontrolled rupture* that is known to occur once a critical crack length, (CCL) is reached.

The outside of every pressure tube in a CANDU reactor is surrounded by a CO<sub>2</sub>-filled annulus gas system (AGS) that is used for the detection of pressure tube leaks through continuous dew point measurements.

The Leak-Before-Break methodology is based on the assumption that the time required for a leak to grow to its critical crack length is always *greater* than the time required to *detect* the leak

The detection of a pressure tube leak

#### APPLICABILITY OF THE LEAK BEFORE BREAK CONCEPT

IAEA, VIENNA, 1993

IAEA-TECDOC-710

June 1993

It has still not been shown that the leak rate can be measured with sufficient accuracy. The possibility exists that large cracks may initially produce only low leak rates. This situation could arise because of corrosion plugging or fouling of relatively slow growing cracks or the relatively uniform growth of a long crack before penetration. In such cases the time required for a small leak to become a significant leak or rupture could be short, depending on crack geometry, pipe loading, and transient loading.

REJECTION INDEX  
FOR PRESSURE TUBES

by  
A.B. Mitchell and D. Meneley

October 1989

The inherent leak detection sensitivity of each reactor unit varies depending on the capabilities of their particular detection systems and other mechanical considerations. For complete reactor monitoring, any system must guarantee sensitivity for each fuel channel. Instances of deficiency in this regard have been detected at Pickering-A (damaged bellows, suspected stagnation, etc.), Bruce-A (prolonged identification of leaking tubes), and Bruce-B (low flow, tight pressure tube bearings). For an LBB scenario to be acceptable, the situation for each fuel channel would need consideration and alternate inspection methods provided for deficient channels.

In addition, we had difficulty in establishing the operating duration for which LBB remained valid because of perceived uncertainties in critical metallurgical data. Further research effort is recommended to determine:

- an acceptable lower bound relationship between CCL, fluence and hydrogen/deuterium effects,
- the effect of flux and fluence on delayed hydride cracking velocity, and
- the influence of stress intensity on delayed hydride cracking velocity.

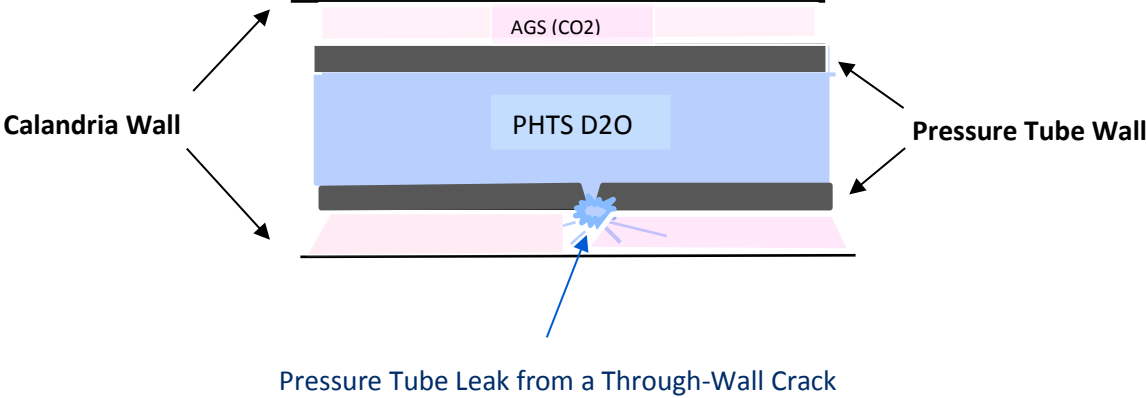
The authors feel that insufficient attention has been given to pressure tubes in several reactors, which may be excluded from LBB monitoring because of annulus gas system monitoring deficiencies. A substitute inspection scenario for these unmonitored tubes, and for the reactor as a whole if LBB becomes invalid in later life, needs to be developed. The overall effectiveness of LBB can only be determined if a detailed LBB analysis is carried out for each operating reactor unit and it is recommended that future studies be less generic and more specific to actual reactor units.

The US NRC Safety Assessment Report on a 2004 AECL submission for the licensing of an advanced CANDU reactor raised the following concerns about the leak detection capabilities of the Annulus Gas System:

US NRC Staff asked AECL if an AGS blockage could go undetected during reactor operation, and what would be the implications of an undetected blockage. AECL responded that a blockage could potentially go undetected for an unspecified period because there is not a uniform practice for monitoring the flow rotameters at utilities operating CANDU reactors. Also, according to AECL, blockage would result in a longer response time to system leakage.

The US NRC Staff considers this issue unresolved because AECL did not address the following: (1) The effects of operating without a fully functional AGS; (2) The corrective actions to be taken following the identification of a blockage; (3) The absence of a flow monitoring program.

# A Pressure Tube Through-Wall Leak into an Annulus Gas System



## CONCLUSIONS

- A pressure tube through-wall crack is expected to start when it's about 27 mm long, and grow at an initial rate of 1.44 mm/hr
- This means, for a *critical crack length* of 40 mm, a catastrophic fast rupture of the leaking tube is likely to occur after only 4½ hrs
- Even for small leaks ~ 100 g/hour, dew point meters will cease to function within 2 hrs due to D<sub>2</sub>O condensation in the AGS
- An AGS can retain up to 12 kg of D<sub>2</sub>O before sufficient liquid reaches a beetle detector and triggers an alarm
- Even for a fairly substantial pressure tube leak of 1 kg/hr, it may take up to 12 hrs for the leak to be detected.
- The Leak-Before-Break acceptance criteria set out in CSA N285.8 are not satisfied by currently operating CANDU reactors

## **Problems with Current Pressure Tube Leak Detection**

1. CSA N285.8 fails to address leak detector performance, calibration checks and/or maintenance requirements
2. Dew Point Meters and Beetles have many short-comings:

### **Dew Point Meters**

Narrow dynamic range  
Need frequent re-calibrations  
Subject to detector drift  
No leak location capability  
Slow recovery after overloading

### **Beetles**

Poor sensitivity  
Not quantitative  
Delayed response  
No leak location capability  
No backup system

A leak-before-break (LBB) assessment requires that the leak rate for a given pressure tube crack length should be determined.

- In 1990 AECL developed the *MARATHON* code to model the leak rate  $Q$  (kg/hr) from a pressure tube crack of length  $L$  (mm) using a *linear* function:

$$Q = 0.5L - 9.0$$

where it was assumed that  $L = 18$  mm when  $Q = 0$

- In the Bruce B AGS Design Manual, DM-NK29-34980/63498, issued 1991, we have a *linear* functional relationship proposed for the time evolution of a leak rate  $Q$  vs. the through-wall pressure tube crack length  $L$ :

$$Q = 0.614L - 16.58$$

where it was assumed that  $L = 27$  mm when  $Q = 0$

- In a 1998 report by K.M Nho, yet another *linear* relationship, (using the same notation as above) is proposed:

$$Q = 0.7618L - 14.86$$

where it was assumed that  $L = 19.5$  mm when  $Q = 0$