DEUTERIUM BUILDUP IN THE ROLLED JOINT REGIONS OF OPERATING CANDU PRESSURE TUBES

by

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ABSTRACT

Deuterium uptake by operating pressure tube material is usually enhanced at the rolled joint regions relative to the ingress into the main body of the tube. Predicting the concentration of hydrogen isotopes in the rolled joint regions as a function of reactor operating time is important in structural integrity evaluations such as flaw assessments and end of life predictions.

A model, accounting for the ingress and changing distribution due to diffusion and precipitation, has been developed, and updated over the past few years, to make predictions of the axial profile of the hydrogen isotope concentration in the rolled joint regions of operating pressure tubes as a function of reactor operating time. The updates included several improvements such as the introduction of the effect of an initial thermal spike (at the inlet of inner zone channels at Bruce A) on the solubility of hydrogen that improved the accuracy of the predictions. As non-linear (with time) models were developed for deuterium ingress due to corrosion in the body of the tube, the rolled joint model was modified to incorporate the observed non-linear behaviour.

1.0 INTRODUCTION

CANDU fuel channels consist of cold-worked Zr-2.5%Nb pressure tubes connected by a rolled joint at each end to a stainless steel end fitting. The pressure tube contains the fuel bundles and the heavy water coolant of the primary heat transport system. The fuel channel is shown schematically in Figure 1. Measured axial deuterium concentration profiles from removed pressure tubes show that deuterium ingress occurs at a much greater rate in the rolled joint region than the main body of the tube. The diffusion of deuterium inboard from the rolled joint region can potentially lead to hydride precipitation inboard of the burnish mark (BM)[1], once the terminal solid solubility (TSS) of hydrogen isotopes is exceeded. The residual tensile stresses, just inboard of the BM, make this region of the pressure tube susceptible to delayed hydride cracking (DHC) in the presence of flaws. Therefore, predicting the concentration and distribution of hydrogen isotopes in the rolled joint regions for operating pressure tubes as a function of time is considered essential for fitness-for-service assessments.

The predictions are made by modelling the buildup and redistribution of deuterium in the rolled joint regions of operating pressure tubes [1-2]. The objective of the modelling is mainly to determine when TSS will be exceeded at the BM at reactor operating temperatures. The model incorporates localized deuterium ingress at the rolled joint as well as deuterium pickup along the pressure tube due to uniform corrosion and describes the diffusional redistribution of deuterium taking into account hydride precipitation when TSS is exceeded. At present, it is not possible to determine the localized rolled joint ingress rates *a priori*. The adopted approach is to derive these rates from analyses of measured deuterium concentration profiles in the rolled joint region of removed pressure tubes from CANDU reactors. The results show that the localized ingress rate at the rolled joint is declining with time. For conservatism, the upper bound to the measured ingress rates is extrapolated and used in the model to predict deuterium buildup over the pressure tube operating lifetime.

The rolled joint deuterium ingress model and its database are continually updated as information from the corrosion and hydrogen ingress R&D programs and data from removed pressure tubes become available. The updates include several improvements such as the introduction of the two TSS concept; the dissolution TSS (TSSd) and the precipitation TSS (TSSp), as well as the introduction of the effect of an initial thermal spike during reactor startup for inner zone inlets of Bruce A pressure tubes. Both measures improved the accuracy of the predictions especially in the BM region [2]. Also, as non-linear (with time) models were developed for deuterium ingress due to corrosion in the body of the tube, the rolled joint model was modified to incorporate the non-linear behaviour of deuterium ingress due to uniform corrosion of the waterside of the tube.

This paper describes the observed ingress at the rolled joint regions, the elements of and the advances in the on-going work of modelling of axial hydrogen isotope concentration profiles in operating pressure tubes at the rolled joints.

[1] The inboard limit of the rolled region is referred to as the "burnish mark"

2.0 ROLLED JOINT INGRESS RATES FROM REMOVED TUBES

Measured axial profiles of deuterium concentration from removed tubes have shown enhanced deuterium pickup at the rolled joint region. This is illustrated in Figure 2. It shows a typical deuterium distribution along the length of a Bruce A pressure tube removed from the reactor after ~17 years of operation. The deuterium concentration profiles at the ends of the pressure tubes result from deuterium diffusion inboard from points of ingress at the rolled joint. At each pressure tube end, the part of the profile attributed to rolled-joint ingress is confined to a small section of the tube, (<100 cm from each end of the pressure tube).

It is generally accepted that ingress occurs at the rolled joint at areas of metal-to-metal contact between the pressure tube and the end fitting. The metal-to-metal contacts are formed during rolling by the breakage of some of the oxide on the outside surface of the pressure tube and serve as windows for deuterium entry into the pressure tube. During reactor operation, pressurized coolant can diffuse along the out-board crevice between the pressure tube and end fitting and re-oxidize exposed areas of the tube. The enhanced corrosion in the crevices formed between the galvanically-coupled pressure tube and end fitting (Figure 1) favours the occurrence of the cathodic reaction on the steel half of each crevice. The deuterium generation on this surface will likely assume a maximum value near points of intimate contact between the pressure tube and end fitting. In addition, a unique electrochemical environment may exist in the crevices. Tests on small specimens coupled by swaging showed that contributions from galvanic and crevice effects lead to enhanced ingress at the rolled joint [1].

However, from a quantitative analytical point of view, the mechanisms of deuterium ingress at the rolled joint are, at present, not fully understood. As a result, it is not possible to determine the rolled joint ingress rates *a priori* and the approach currently adopted is to derive these rates from measured deuterium profiles in the rolled joint region of removed surveillance pressure tubes. For example, Figure 3 shows the deuterium concentration profile at the outlet end of a Bruce B tube which is typical of profiles at the ends of pressure tubes and result from deuterium diffusion inboard from points of ingress at the rolled joint. From the measured deuterium concentration profile and the extent of deuterium diffusion inboard, the total amount of deuterium picked up and the contributions from rolled joint ingress and pickup along the pressure tube due to corrosion (assumed to be uniform) can be determined as depicted in Figure 3. Using this approach, a database on rolled joints removed from operating reactors (mainly from Bruce A) has been compiled. The results for the inlet rolled-joint region is plotted in Figure 4, which shows the amount (mass) of deuterium picked up via rolled joint ingress as a function of operating time in hot years[2] (each measured profile is represented by one point on the chart).

[2] Hot operating time (hot years or hot hours) refers to time of operation plus time at hot shutdown.

The functional form of the time dependence of the rolled joint ingress rate has been previously discussed in reference [2]. Briefly, the ingress rate, R, is assumed to be inversely proportional to the oxide thickness, which in turn is assumed to grow with time at the parabolic rate law commonly observed:

$$R(x,t) = \frac{F(x)}{at^{1/2} + b} \tag{1}$$

Where F(x) is the assumed shape of the ingress spatial profile, x is the position, t is the time and a and b are constants determined from a fit to the measured deuterium mass, M, picked up via rolled joint ingress. Integrating Equation (1) gives:

$$M(t) = 2\pi rw\rho\lambda\beta\left[\alpha t^{1/2} - \ln\left(1 + \alpha t^{1/2}\right)\right] \tag{2}$$

Where α=a/b and $\beta=2b/a^2$, r and w are the pressure tube radius and wall thickness, respectively, ρ is the density of the pressure tube material and λ is given by:

$$\lambda = \int F(x)dx \tag{3}$$

A non-linear least-squares fit procedure is then used to obtain the coefficients α and β. The results are also shown in Figure 4 for the inlet rolled joints. Most of the data to date are from Bruce A rolled joints (shown in red) and the initial fit in Figure 4 (solid red curve) was to the data from Bruce A only. The dashed curves, giving the upper and lower bounds, represent the observed scatter in the data.

3.0 CIRCUMFERENTIAL SCRAPING IN THE ROLLED JOINT REGION

Microsampling (scraping) is a process in which a pressure tube is sampled by removing a thin layer of metal from the inside surface. To avoid the inclusion of high deuterium concentration material from the oxide and near-surface region in the analysis, double scrape samples are taken with the first one designed to remove the oxide and near-surface metal layer, and the second one, from within the crater of the first, to remove a metal sample that is subsequently analyzed to determine the hydrogen isotope concentration. Scrape thickness varied from 75 - 125 µm. Scrape lengths have ranged from 40 mm to 75 mm with a nominal width of ~6 mm. Scraping is a non-destructive and less expensive method of acquiring the hydrogen isotope concentration in operating pressure tubes than tube removal (where through-wall punches can be taken as samples). However, the length of an axial scrape (the common form of scraping the body of tube) does not allow for detailed axial deuterium concentration profiling at the rolled joint region where the axial region 0-100 mm from the pressure tube end is critical. Therefore, circumferential scraping was developed especially for sampling in the rolled joint regions.

The first circumferential scrape samples (from an operating pressure tube) were taken from the inlet rolled joint area of a Bruce 7 pressure tube and analyzed for hydrogen and deuterium concentration. The scrapes were taken at three typical locations approximately 12, 27 and 43 mm from the pressure tube end, respectively. The deuterium concentrations at these locations were determined by hot vacuum extraction mass spectrometry (HVEMS) to be 41, 41 and 38 ppm, respectively. Although the results were not sufficient to determine the ingress rate at the rolled joint, an estimate of the ingress rate was obtained using the rolled joint ingress model and its database (based on removed tubes) by adjusting the rolled joint ingress rate to fit the measured concentrations at the scrape locations. Since there were no data away from the rolled joint, it was not possible to determine the deuterium pickup rate along the tube due to corrosion. It was therefore assumed, in this particular case, that the corrosion pickup rate at the inlet end of the Bruce 7 tube was the same as that for the inlet end of a removed tube from Bruce 6. For the latter, the corrosion pickup rate had been determined to be 0.75 ppm deuterium per year. The results of the fit are shown in Figure 5. The inlet operating temperature for the Bruce 7 tube is 266°C and the scrapes were taken after 78,000 hot hours. The initial hydrogen concentration in the pressure tube is 10.3 ppm. Subsequently, circumferential scrapes were taken from the inlet rolled joints of 4 other pressure tubes from Bruce B (with operating time ranging from 87,000 to 102,000 Hot Hours) and the results are also shown in Figure 5.

Deuterium pickup due to rolled joint ingress, determined from the fits in Figure 5, is compared to other Bruce inlet rolled joints in Figure 4. The results show that deuterium pickup in the Bruce B rolled joints is low compared with that in Bruce A inlet rolled joints. The reasons for this difference are not fully understood at the present time.

The use of scrape data with the model to analyze rolled joint ingress can be helpful in providing lifetime predictions and is a major improvement over generic predictions using (for conservatism) upper bound ingress rates based on removed pressure tubes. To determine the deuterium pickup due to corrosion, and hence determine more precisely the contribution from rolled joint ingress, it is necessary that future monitoring campaigns consider additional measurement be taken further inboard (500 mm from the pressure tube end), if possible, to help infer the ingress due to corrosion.

4.0 BRUCE B ROLLED JOINT DEUTERIUM INGRESS RATES

Figure 4 shows that deuterium pickup at the inlet rolled joints of Bruce B pressure tubes is lower than pickup in Bruce A rolled joints. This observation suggested that a Bruce B specific rolled joint ingress rate should be derived. The data obtained from the removed and analyzed tube from Bruce 6, the results from the above mentioned circumferential scraping of the inlet rolled joint region of five tubes, were used to derive a rate curve specific for rolled joint ingress for Bruce B reactors. The Bruce B inlet data are shown along with the data from Bruce A on the rolled joint ingress rate graph in Figure 4. To construct a new curve for Bruce B, the functional shape for the rate curve was maintained (from equations 1-3) but scaled down to pass through the average of all the Bruce B points at ~10 hot years. This was considered a mean curve for Bruce B inlet rolled joints with ±2σ for upper and lower bounds as depicted in Figure 4. The upper bound, which is slightly lower than the mean curve for the reference (Bruce A) curve, is used for current Bruce B inlet predictions.

At the outlet rolled joint region, it was assumed that the relationship between the inlet rate and the outlet rate from the reference curves of Bruce A is maintained for Bruce B inlet and outlet. Thus, an outlet rate curve for Bruce B was obtained using the scaling factor used at the inlet. The mapped curves are shown in Figure 6 compared to the reference Bruce A outlet curves. The upper bound of the mapped Bruce B outlet curve is used for Bruce B outlet predictions.

Subsequently, data from two removed Bruce B tubes were obtained. The results from the two tubes are at or below the mean line of each of the rate curves as illustrated in Figures 4 and 6.

5.0 DISCUSSION

Predicting the concentration of hydrogen isotopes in the rolled joint region of operating pressure tubes as a function of reactor hot hours is a key objective of the modelling. The total concentration of all hydrogen isotopes, is defined as equivalent hydrogen concentration, $[H]_{eq}$:

$$[H]_{eq} = [H] + 0.5\,[D] \tag{4}$$

Where [H] is the initial hydrogen concentration and [D] is the deuterium concentration. The equivalent hydrogen concentration in the rolled joint region of operating pressure tubes is a key factor in integrity assessments such as those for fuel bundle bearing pad fretting flaws in the high stress region of the rolled joint (e.g. BM). The objective of the assessments usually includes predicting when such locations will reach TSS to avoid the possibility of DHC. The Bruce B predictions based on the new rate curves are expected to show a reduction in [D] in the rolled joint region as a result of using lower rolled joint ingress rate curves; this will result in an increase in the time to reach TSS at, for example, the BM. As an example, predicted $[H]_{eq}$ profiles versus operating time for an inlet axial band (60-90 mm from inlet end of the tube) containing the BM for the reference analysis and the updated Bruce B analysis are compared in Figure 7. Note that the time to reach TSS is significantly extended with the new predictions (shown as BB). The results presented here were based on the 2σ upper bound to the observed rolled joint ingress data and hence were intended to be conservative. It is emphasized that these results were based on a somewhat limited database and as more reactor data are obtained, the assumptions and the predictions should be reviewed and updated accordingly. In addition, the calculations are usually based on a set of assumptions so that the confidence in the predictions is dependent on the validity of the assumptions [3].

6.0 SUMMARY

The rate of localized deuterium ingress at the rolled joint is modelled using an empirical rate curve based on measured axial profiles mainly from Bruce A removed tubes. The rolled joint ingress rate curve is a relationship between the amount of deuterium picked up at the rolled joint and hot years. The amount of deuterium picked up through the rolled joint is equal to the integrated area under a measured axial [D] profile in the rolled joint region less the uniform ingress due to inside surface corrosion.

The results from circumferential scraping of the inlet rolled joint region of five Bruce B tubes, in addition to the data obtained from a removed tube from Bruce 6, were used to derive a rate curve specific for Bruce B reactors. The functional shape of the reference Bruce A rate curve was maintained but scaled down to pass through the average of all Bruce B points roughly at 10 hot years. This was considered a mean curve for Bruce B inlet with $\pm 2\sigma$ for upper and lower bounds. The upper bound is used for Bruce B inlet predictions. At the outlet region, assuming that the relationship between the inlet rate and the outlet rate from the reference curves is maintained for Bruce B inlet and outlet, a rate curve for Bruce B outlet was obtained using the same scaling factor. The upper bound of the mapped Bruce B outlet curve is used for Bruce B outlet predictions.

The new predictions show a significant reduction in the predicted $[H]_{eq}$ profiles both at the inlet and the outlet rolled joints as a result of using the new ingress rate curves compared to the previously used reference curves (from Bruce A).

Subsequent data from two Bruce B removed tubes fell at or below the mean line of the new Bruce B curves. However, these curves are based on a somewhat limited database and as more reactor data are obtained, the assumptions and the predictions should be reviewed and updated accordingly. It is clear that circumferential scraping could be used to enhance the crucial rolled joint ingress database for all reactors. This should result in a significant improvement in fitness-for-service assessments.

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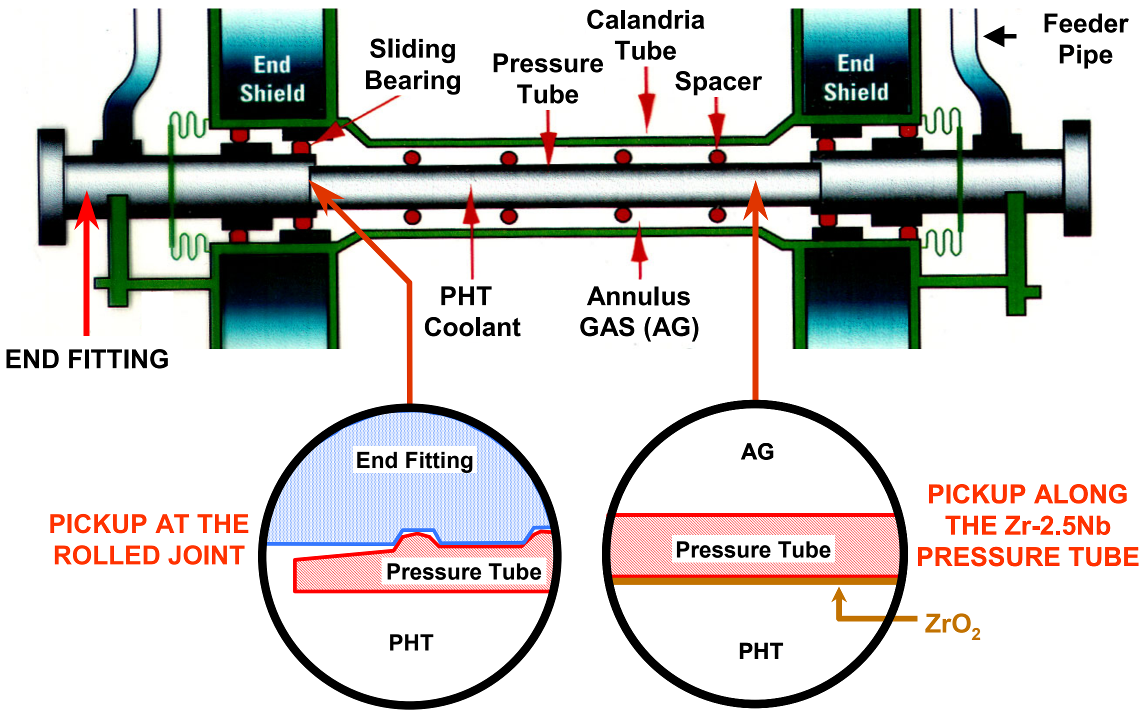


Figure 1: A schematic of the CANDU fuel channel. Under normal operating conditions, CANDU pressure tubes pick up deuterium along the inside surface due to oxidation by the coolant. Additional ingress at the rolled joint results from galvanic and crevice effects between the Zr-2.5%Nb pressure tube and the stainless steel end fitting.

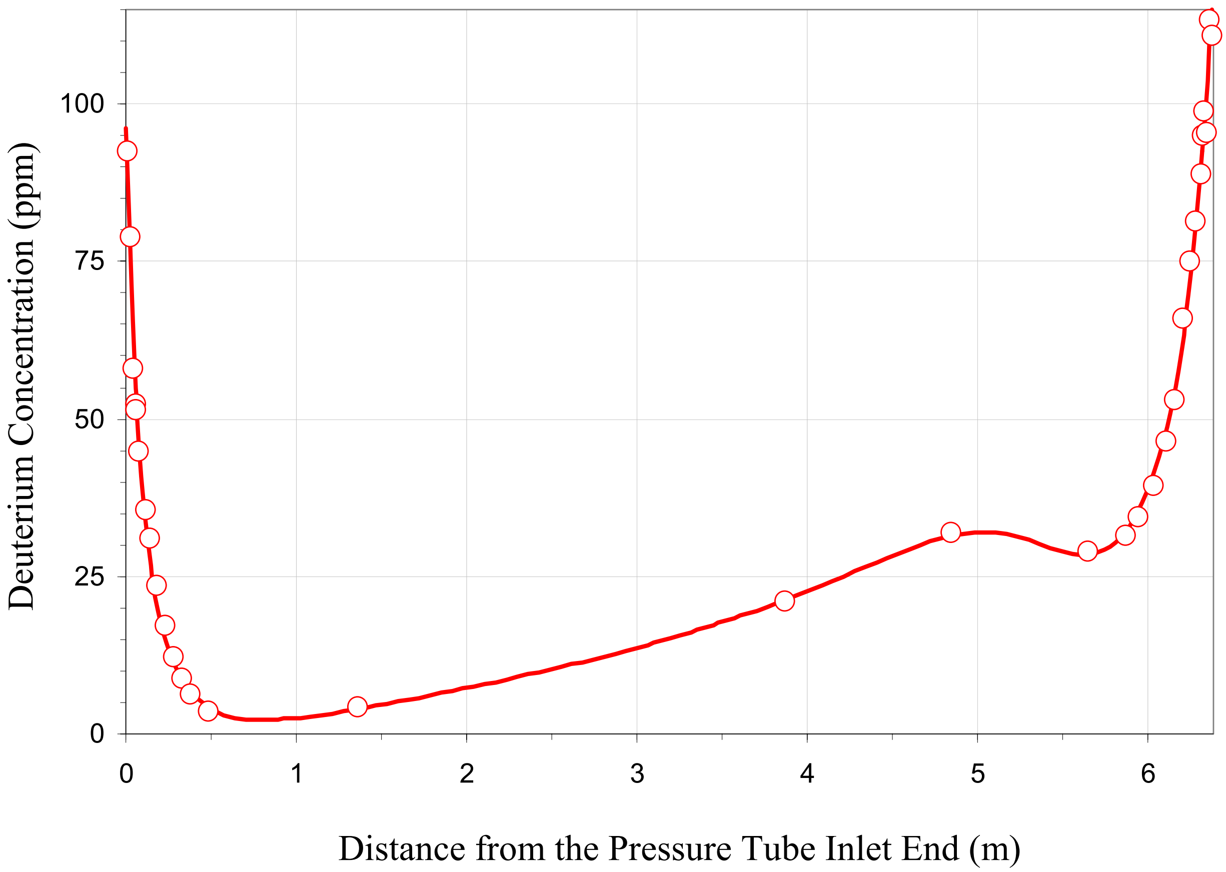
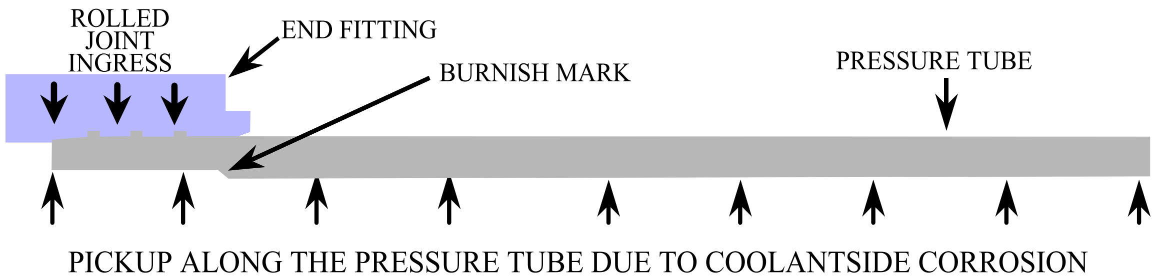


Figure 2: Deuterium concentration profile along a pressure tube from Bruce A removed after ~17 hot years of operation.



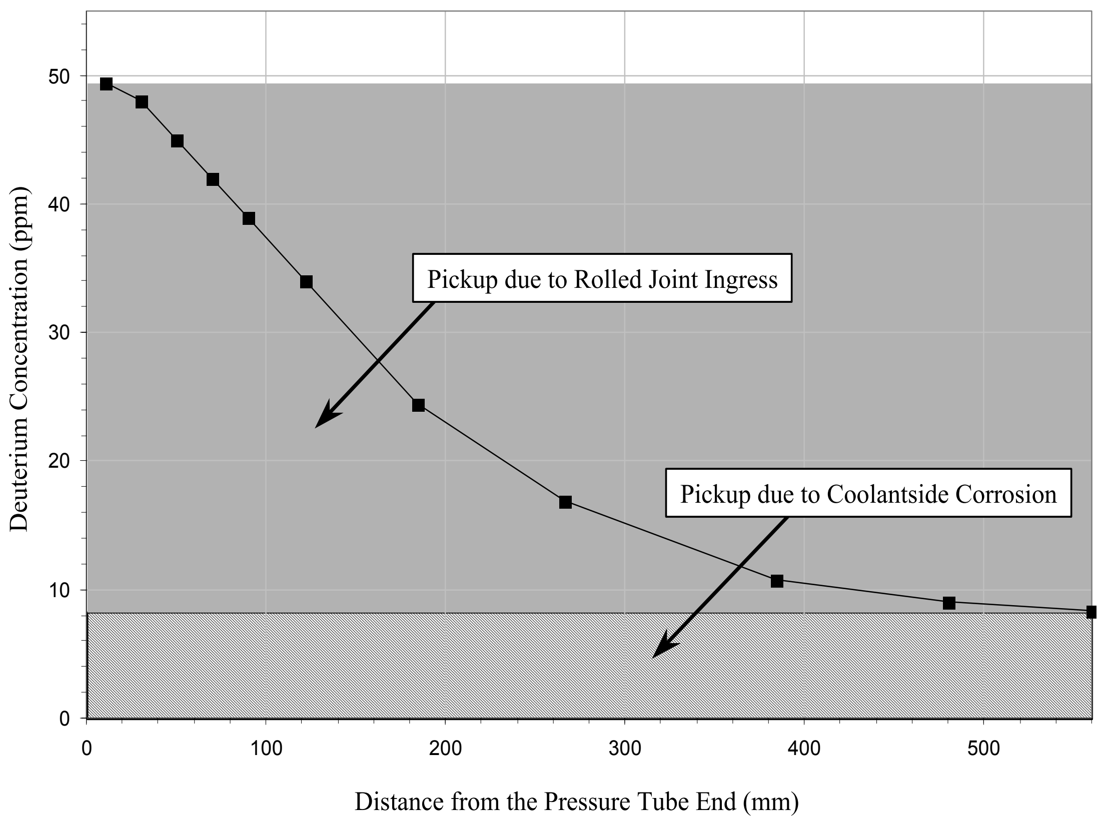


Figure 3: Measured deuterium concentrations at the outlet rolled joint of the Bruce 8 pressure tube. The plot indicates the amounts of deuterium picked up due to rolled joint ingress and as a result of corrosion on the coolant side.

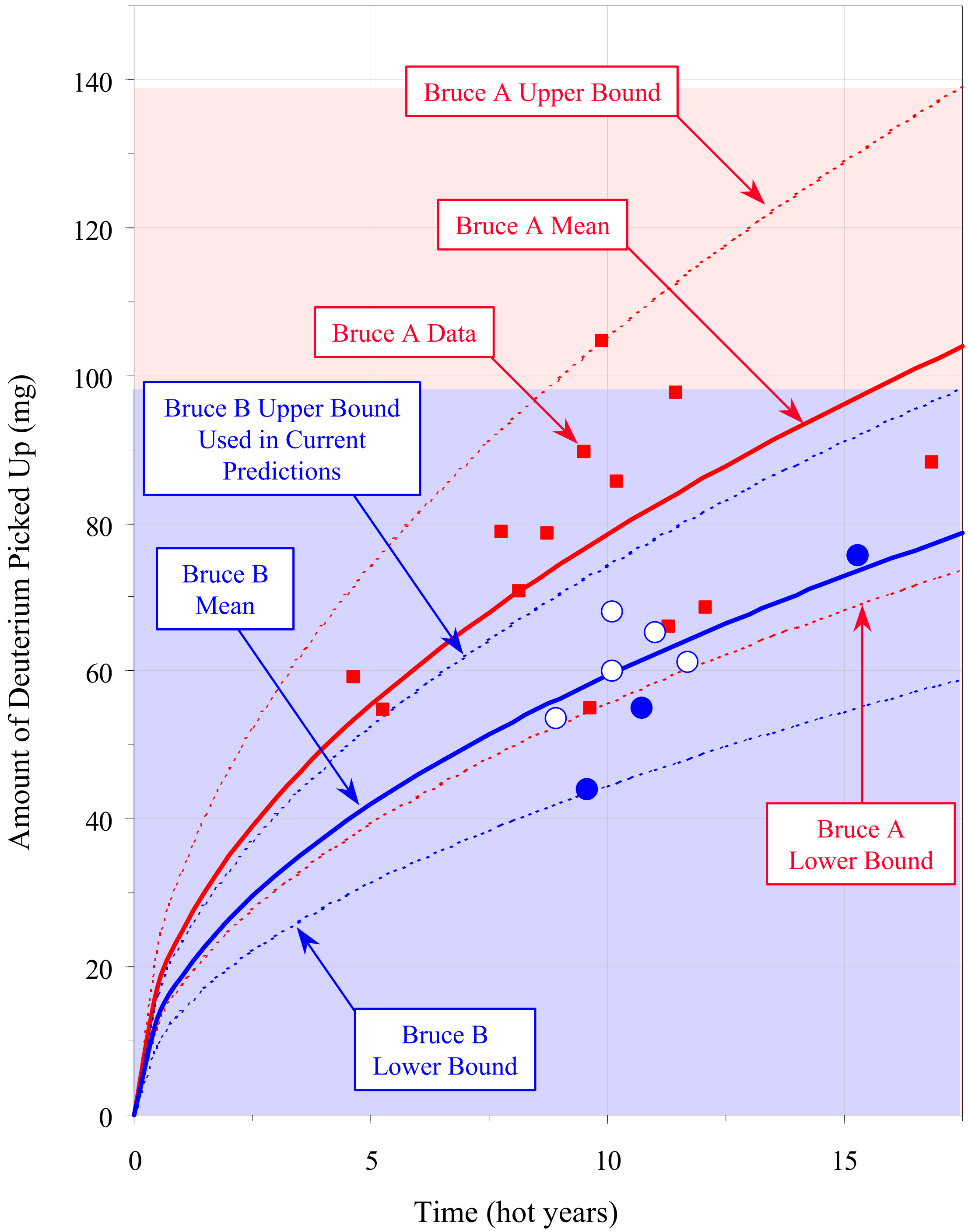


Figure 4: Amount of deuterium picked up via rolled joint ingress as a function of time for Bruce A inlets (solid squares). The solid curve is the best fit assuming a declining ingress rate and the dashed curves represent upper and lower bounds. Also shown are Bruce B inlet rolled joint ingress rate curves (mean, upper and lower bounds shown in blue); open and solid circles are data from scraped and removed tubes, respectively.

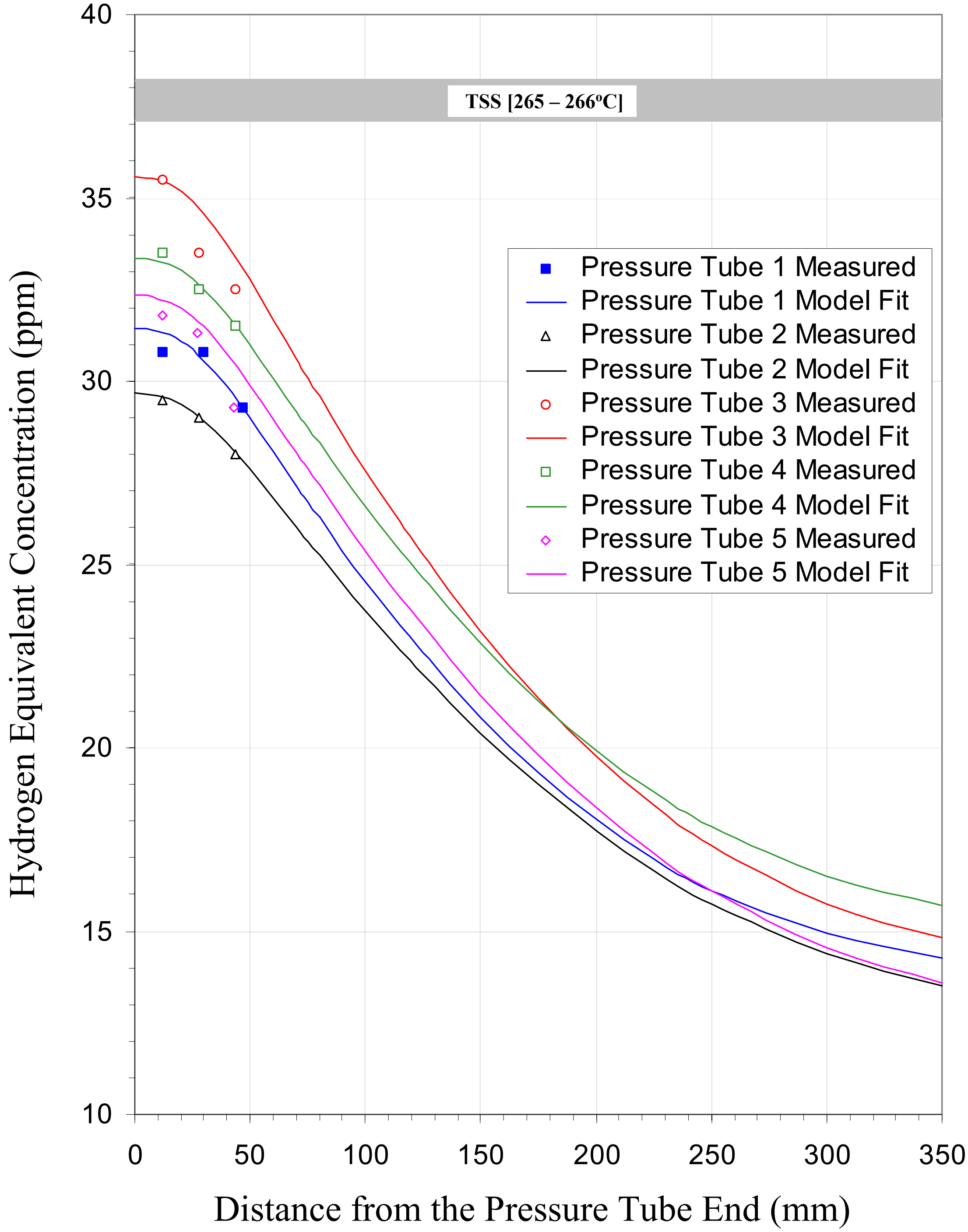


Figure 5: Hydrogen equivalent concentration at the inlet ends of five Bruce B tubes. The model profiles were obtained by adjusting the rolled joint ingress rate to fit the measured (scrape) data.

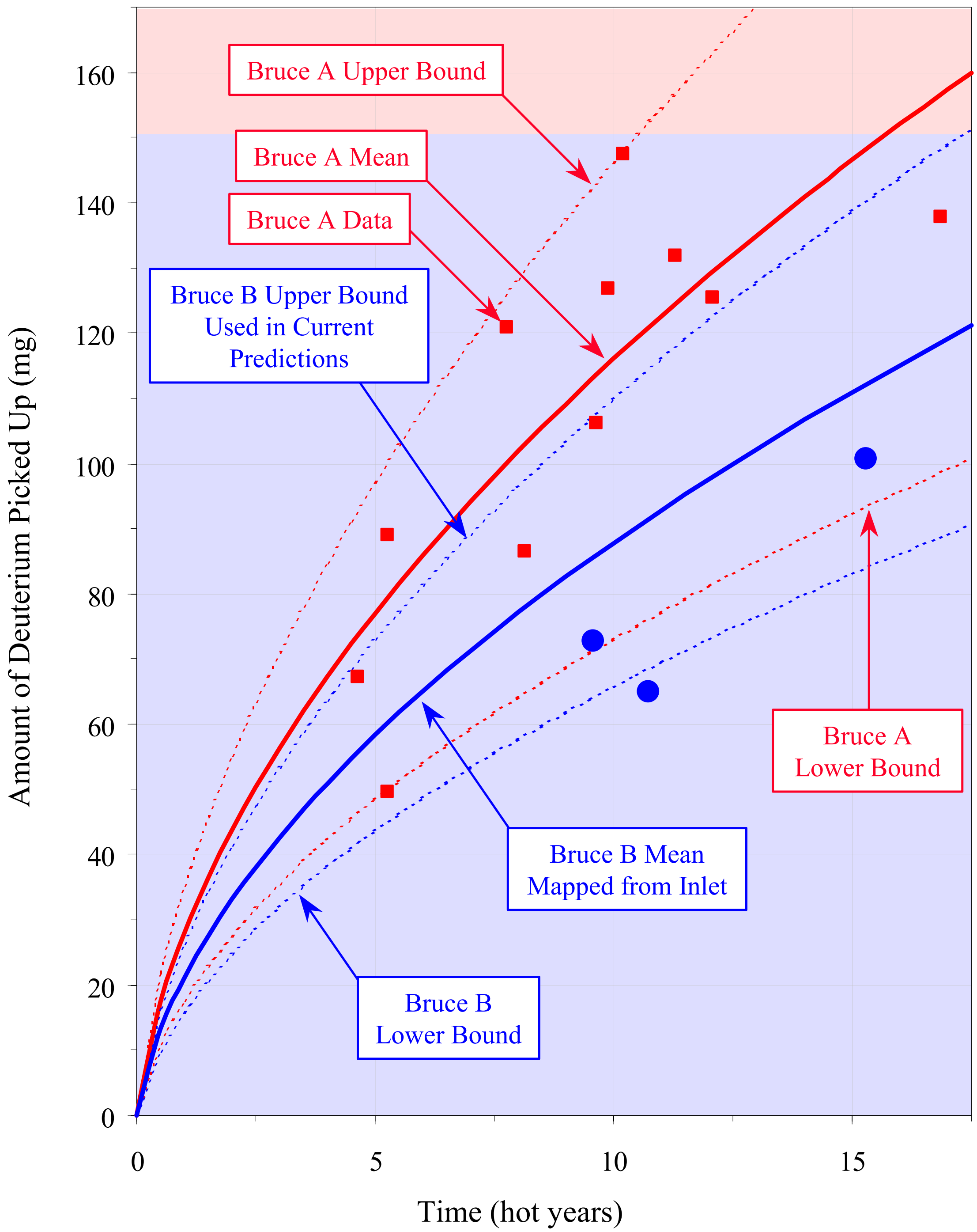


Figure 6: Amount of deuterium picked up via rolled joint ingress as a function of time for Bruce A outlets (solid squares). The solid curve is the best fit assuming a declining ingress rate and the dashed curves represent upper and lower bounds. Also shown is Bruce B outlet rolled joint ingress rate curves (mean, upper and lower bounds shown in blue).

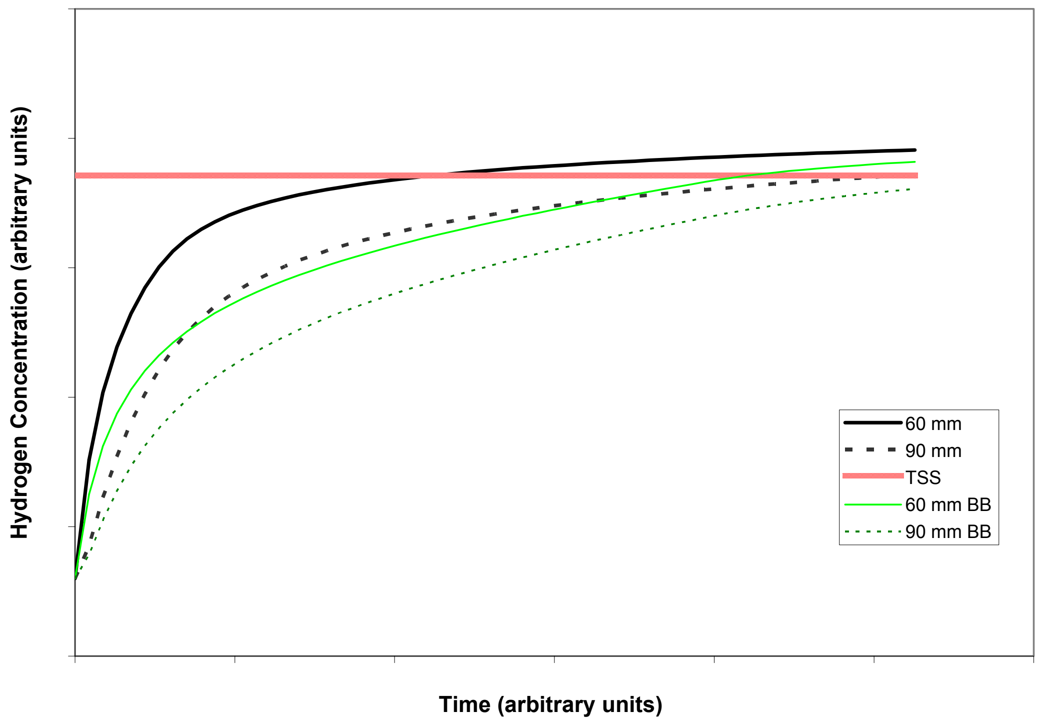


Figure 7: A comparison between previous and present predictions of the evolution of $[H]_{eq}$ with operating time at Bruce B outer zone inlet rolled joint at axial locations of 60 and 90 mm from the pressure tube (BB indicates the new predictions).