Irradiation Strengthening and Effect of Test Temperature on Zr-2.5Nb Pressure Tube Material

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ABSTRACT

It has been known that the strength of Zr-2.5Nb pressure tube material, as measured by the yield stress or the ultimate tensile strength, increases when exposed to fast neutron irradiation. The function describing the dependence of strength on fast neutron fluence is required in modeling the creep and fracture properties of pressure tube materials used in CANDU nuclear reactors. A semi-empirical expression describing the strengthening effect as a function of fast neutron fluence is derived. In addition, the effect of both the irradiation temperature and test temperature is also assessed.

IRRADIATION HARDENING DUE TO FAST NEUTRON FLUX

Strengthening of a material that is exposed to fast neutron irradiation (all neutron flux and fluence values used in this work are defined for energies E > 1 MeV) can be quantified by means of the increase in the 0.2% yield stress, σ_y, or the ultimate tensile strength, UTS. Sagat et al. [1] have shown that at 523 K, the σ_y of pressure tube (PT) material initially increases rapidly with fast fluence. When fluences in the order of 1×10^{24} n/m^2 or higher are reached, the slope of the curve decreases dramatically and any subsequent increase in σ_y becomes much lower than that at low fluences. A semi-empirical expression describing the relative increase of either σ_y or UTS as a function of fast fluence Φ is described next.

The increase in strength observed when a material is under a fast neutron flux is attributed to the accumulation of irradiation damage in the form of dislocation loops. It is assumed that the effect of accumulated damage on dislocation glide is similar to that of small particles, namely, similar to that observed due to Orowan hardening that was shown to be consistent with data from Zircaloy-2 tests [2]. In this case the expression relating the increase in strength, as measured by σ_y, with damage is:

$$\Delta\sigma_y = A\cdot(Nd)^{0.5} \qquad (1)$$

$$A = F\alpha Gb \qquad (1a)$$

Here N, the average loop density due to irradiation damage only
d, the average visible loop size,
F, the Taylor factor
α, a constant that describes the obstacle strength,
G the shear modulus and
b the Burgers vector.

The values of these constants used in this work are $d = 1 \times 10^{-8}$ m, F = 3.6, α = 0.465, G = 34800 MPa and $b = 3.3 \times 10^{-10}$ m. As was discussed in more detail in [2], the parameter F depends on crystallographic texture. However, it was shown in [2] that the value of F that fits best Zr based alloys lies between 3.5 and 4.0 and therefore the value found above appears reasonable. The value of α that is consistent with Zr-2 data is about 0.35, i.e., lower than that found above for Zr-2.5Nb. However, the

Zr-2 samples in [2] were irradiated at 573 K and the mechanical tests were carried out at room temperature. It is therefore possible that the obstacle strength presented by the loops in Zr-2 is different from that presented by the loops generated in Zr-2.5Nb at a lower irradiation temperature. Alternatively, the difference in α could reflect some differences in solution strengthening, i.e., Sn and Nb having different binding energies with dislocations. Note that the undersized Nb atoms tend to shrink the lattice whereas the oversized Sn atoms tend to spread the lattice in the <a> direction and this might be consistent with the higher value of α found here.

The increase in strength is attributed to the increase in N with fluence. This is consistent with the observed increase of the <a> type dislocations (see Figure 18 of [1]) and partially to the <a> component of the <c+a> type loops forming due to irradiation. Assuming that there is no difference between the interactions of the strain fields of straight dislocations and loops, the increase of the <a> type dislocation density as a function of Φ can be used to derive the dependence of N on Φ. An expression describing the evolution of N with Φ was suggested by Wolfer [3] and is given by:

$$dN/dt = B \cdot N - C \cdot N^2 \tag{2}$$

The first term in the right hand side (rhs) of Eq. (2) describes the generation rate of loops and the second term the recovery rate in a manner similar to that describing the thermal recovery of dislocations. Integration of Eq. (2) gives:

$$N = \frac{N_s}{1 - \left(1 - N_s / N_o\right) \cdot \exp\left(-C \cdot N_s \cdot t\right)} \tag{3}$$

Here N_s and N_o are the saturation and initial values of N, respectively.

Eq. (3) saturates with time, whereas the evolution of N with Φ does not appear to reach a constant value N_s in Figure 18 of [1]. It was therefore decided to derive a different expression that describes the observed behaviour more accurately. Instead of Eq. (2) the generation rate of N with Φ can be expressed as follows:

$$dN/d\Phi = B - A_o \cdot (1 - e^{-\Phi/\Phi_o}) \tag{4}$$

Here B is the generation rate and the second term describes the recovery rate of loops under irradiation (analogous to the thermal recovery).

At $\Phi = 0$ the initial generation rate is B, while at $\Phi >> \Phi_o$ $(dN/d\Phi)_{\Phi \to \infty} = B - A_o$. If $A_o = B$ then $dN/d\Phi$ tends to 0 and therefore N tends to saturate with Φ and tends to the value $B\Phi_o$. However, if $A_o < B$ then N does not saturate with Φ. Integrating Eq. (4):

$$N = (B - A_o) \cdot \Phi - A_o \Phi_o \cdot (e^{-\Phi/\Phi_o} - 1) \tag{5}$$

It is evident that if $A_o = B$, at $\Phi >> \Phi_o$, N tends to saturate to the value $B\Phi_o$. In [2] it was shown that the relationship between N and the increase in the <a> dislocation density $\rho_{<a>}$ is given by

$$N = \Delta\rho_{<a>} / (\pi d) \tag{6}$$

Here $\Delta\rho_{<a>} = \rho_{<a>} - \rho_o$, where ρ_o is the initial <a> type dislocation density. Using Eqs. (5) and (6) and the data from Fig. 18 of [1], the constants B, A_o, ρ_o and Φ_o can be determined. These are, $\rho_o = 3.5 \times 10^{14}$ m^{-2}, $B = 5.2 \times 10^{-3}$, $A_o = (0.987 \times B)$ and $\Phi_o = 2.3 \times 10^{24}$ n/m^2. The dislocation density change with irradiation can be assumed to also describe the loop density change with irradiation. Using these values, the fit to the data in Figure 18 of [1] is shown in Figure 1 below.

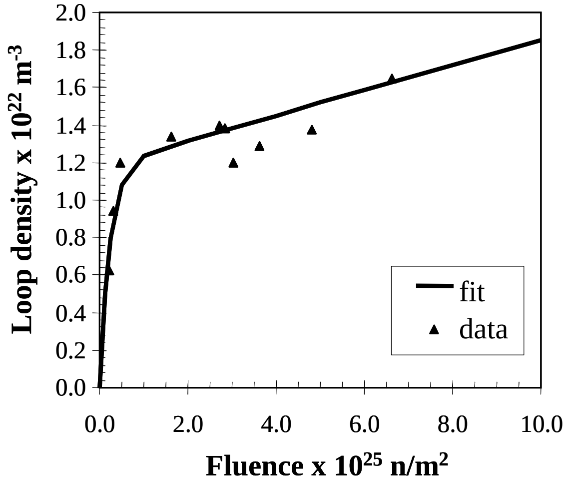

Figure 1: Evolution of loop density with fast neutron fluence (E > 1 MeV) (data from [1]).

Substituting these values in Eq. (5), the derived expression giving the relative increase in strength σ_y / σ_y^o, where σ_y^o is the yield strength before irradiation, can be derived from Eq. (1) and is given by:

$$\sigma_y / \sigma_y^o = 1 + C_1 \sqrt{C_2 \cdot \Phi - C_3 \cdot \{e^{-\Phi/\Phi_o} - 1\}} \tag{7}$$

Here $C_1 = 3.204 \times 10^{-12}$, $C_2 = 6.76 \times 10^{-5}$ m^2/n, $C_3 = 1.18045 \times 10^{22}$. Eq. (7) is compared with data from [1, 4] in Figure 2. It is evident from Figure 2 that the model of irradiation strengthening described above describes the observed increase in strength in pressure tube material well to end-of-life accumulated fluences. Note that the solid points in Figure 2 all came from the same pressure tube, whereas the open points are from various surveillance pressure tubes.

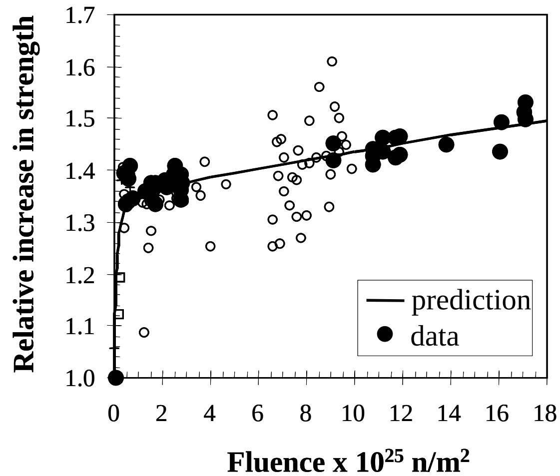

Figure 2: Comparison of the calculated relative increase in strength from Eq. (7) with fluence [1, 4].

EFFECT OF TEMPERATURE ON IRRADIATION HARDENING

The strengthening observed during neutron irradiation depends on both the irradiation temperature and test temperature. However, in the past it was mainly the effect of test temperature that has been assessed in detail. For instance, Figure 3 shows the UTS as a function of fluence for only two test temperatures. The data in Figure 3 were obtained from a number of surveillance tubes. In the case of the low temperature test, the irradiation temperature of the samples was in the range of 520 – 580 K. In the case of the high temperature tests, the irradiation and test temperatures were the same. However, it is known that if the test temperature is substantially higher than the irradiation temperature, the material strength will approach that of the un-irradiated state, since high temperature promotes rapid recovery of irradiation damage.

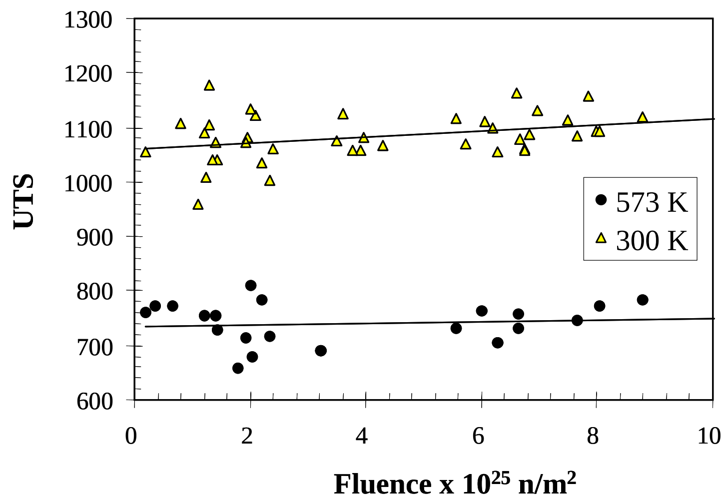

Figure 3: Effect of test temperature on the UTS of PT material.

The temperature dependence of the UTS of un-irradiated and irradiated PT material is shown in Figure 4. In this figure the data were normalised with the UTS of the unirradiated material measured at room temperature, i.e., 833 MPa. An age hardening plateau due to dynamic strain aging is observed in unirradiated Zr-2.5Nb pressure tube materials [6]. This plateau is not normally evident in irradiated pressure tube material. Dynamic strain aging may be suppressed in irradiated material due to trapping of oxygen in irradiation-induced defect clusters [7], or the enhanced precipitation of Nb in the α-phase, which reduce the ability of the Nb atoms in locking up mobile dislocations [8]. Kelly and Smith [9] as well as Veevers et al. [10] showed that there was no static strain aging in zirconium alloys irradiated to 3.7×10^{24} n/m^2. Himbeault et al. [7] demonstrated in one irradiated pressure tube material that the strain aging parameter ($\Delta\sigma/\sigma$) is greatly reduced, but not completely absent. It was speculated that the small strain aging parameter observed is due to the small grain size of the pressure tube material. In the analysis presented here it is assumed that the hardening, as measured by the relative increase in the UTS, is a linear function of test temperature.

The manufacturing procedure has evolved with time and as a result, slight microstructural variations exist, which in turn affect the tube's behaviour. The scatter observed in Figure 5 is partly due to the material variability resulting from changes in the manufacturing procedures. The scatter is also partly a reflection of differences in the testing procedure since the data are from pressure tubes removed from CANDU reactors that were tested over approximately 20 years as part of ongoing surveillance examinations. To minimize the scatter, data from specimens from either the 3:00 or 9:00 in-reactor o'clock positions were preferentially selected. This minimizes the influence of a local variation in irradiation temperature between the top (12 o'clock) and bottom (6:00 o'clock) positions [11]. The majority of specimens were also tested at temperature below the nominal irradiation temperature to eliminate the possibility of recovery of radiation damage. However, given the short duration of the tests it is unlikely that recovery is occurring which is supported by measurements of the dislocation density by X-ray diffraction analysis (data not included). To eliminate strain rate effects only data from specimens tested at strain rates of ~10^{-3}/s were included. The effect of fluence is also not considered in this analysis since, as shown in Figure 3, the increase in strength following the initial transient is small. Figure 5 shows the effect of test and irradiation temperature on the normalised UTS of the material. The normalizing UTS value is that used in Figure 4.

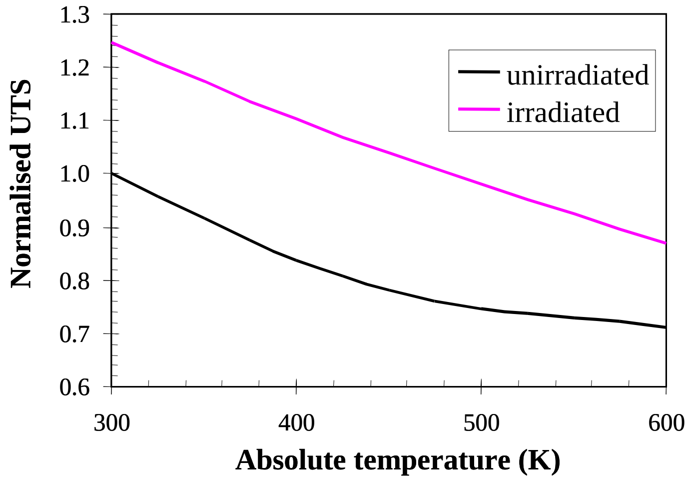

Figure 4: Temperature dependence of UTS of un-irradiated and irradiated PT material.

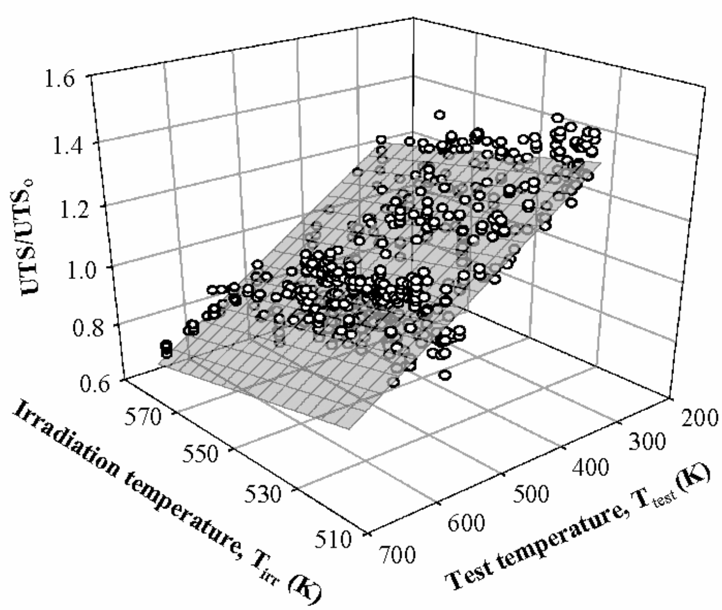

Figure 5: Effect of irradiation temperature T_{irr} and test temperature T_{test} both in K, on the normalized transverse UTS of surveillance tubes removed from CANDU Nuclear Generating Stations.

A best fit linear equation describing the temperature dependence of the UTS is given below:

$$UTS/UTS_o = D_1 - D_2 \cdot T_{irr} - D_3 \cdot T_{test} \quad (8)$$

Here $D_1 = 3.14$, $D_2 = 2.53 \times 10^{-3}$ 1/K, $D_3 = 1.47 \times 10^{-3}$ 1/K and T_{irr}, T_{test} are the irradiation and test temperatures in K respectively. This equation shows that the irradiation temperature has a much stronger effect on the hardening of the material than the test temperature. It is evident from Figure 5 that Eq. (8) predicts the dependence of material strengthening with test and irradiation temperature well.

The strength of a pressure tube prior to installation in reactor increases from the Front-end to the Back-end. The Front-end emerges first during the extrusion process and has a lower strength than the Back-end. For a pressure tube oriented with its Back-end at the Inlet (B/I), the inlet end will have the highest possible strength at the inlet temperature after irradiation due to the high initial strength and low irradiation temperature. The outlet end will have the lowest strength (at the outlet temperature) due to the high irradiation temperature and low initial strength. Alternatively if the tube is installed with its Front-end at the Inlet (F/I), its strength at the inlet end will be less than the tubes with B/I orientation at the corresponding location because of its lower initial strength. Figure 6 shows planar fits to the normalized UTS as a function of irradiation and test temperature with the data separated according to orientation. For clarity, individual data points are not shown. Clearly, the UTS of the tube is affected by the orientation and the lowest strength is with the Front-end at the outlet.

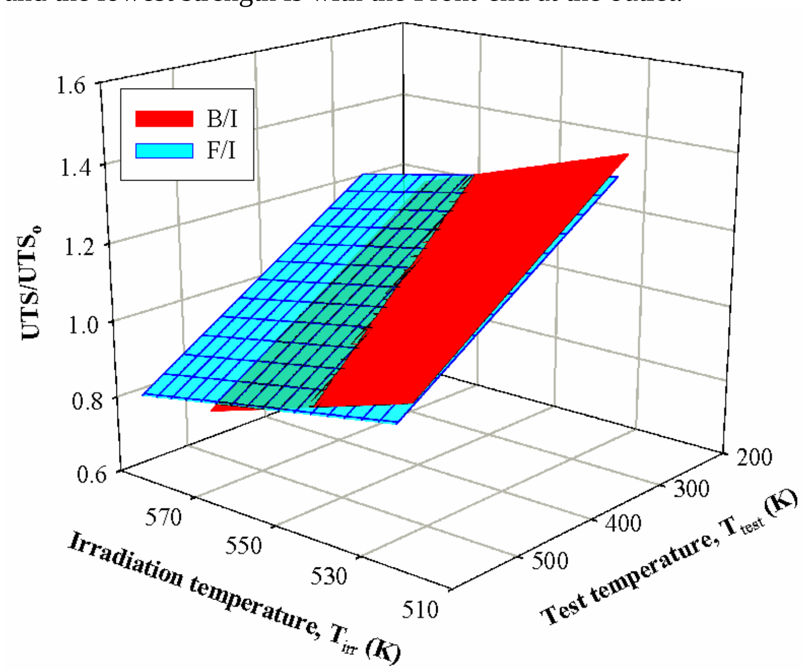

Figure 6: Effect of orientation on the normalized UTS of irradiated pressure tubes.

For some surveillance pressure tubes offcut material (material removed prior to installation in reactor) from the Front-end and Back-end was available for testing. For these tubes the tensile properties at both ends prior to installation in reactor can be measured. Assuming the strength varies linearly along the length of the tube then the strength at any location can be determined. If the initial strength is known then the increase in strength due to irradiation can be derived. In this case to eliminate the effect of test temperature, only data from tests at 250°C are included in this analysis. Figure 7 shows the UTS after irradiation as a function of irradiation temperature and the initial local UTS estimated from the unirradiated Front and Back-end off cuts, assuming linear variability. There is a positive relationship between the initial strength and the irradiated UTS. However, the slope of less than 1 (0.84) indicates that the stronger the initial material, then less additional strengthening occurs as a function of fluence.

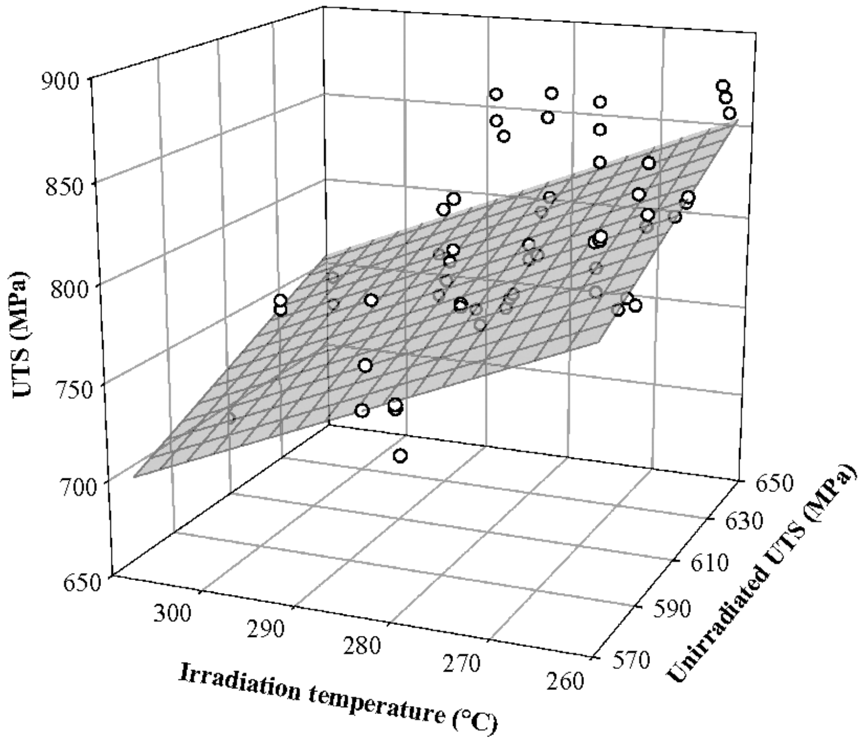

Figure 7: Effect of irradiation temperature T_{irr} and initial local UTS on irradiated UTS

DISCUSSION – CONCLUDING REMARKS

The relative increase in the strength of PT material is consistent with irradiation damage that is introduced when highly energetic neutrons create damage cascades that eventually form dislocation loops. These loops can be treated as obstacles to dislocation motion when the material is subjected to external stresses. As a result of the increased resistance to these obstacles, the material exhibits higher strength, measured either by the 0.2% yield stress or the UTS. It appears that the increase in strength with accumulated fast fluence can be treated as Orowan hardening and the resulting formulation describes well the transient and steady state regimes observed during testing.

In addition to the dependence of strength on fast fluence, the material exhibits a strong dependence on test temperature T_{test} and irradiation temperature T_{irr}. When the relative increase in strength is plotted against these two variables, a linear dependence appears to describe the results well. From the linear fit, T_{irr} has a greater effect on the strength of the material than T_{test}. The UTS of irradiated material is also dependent on the strength prior to installation in reactor. At the lower operating temperature found at the inlet end of the fuel channel pressure tubes oriented with the Back-end at the inlet have the highest strength. With the reverse orientation (Front-end at the inlet) the strength at the same location will be less because of its lower initial strength. Differences in the initial strength, affects the strengthening that occurs with irradiation.

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