

Letter to the Editors

# Volume conservation during irradiation growth of Zr–2.5Nb

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## Abstract

Dimensional changes are reported in three dimensions for cold-worked Zr–2.5 Nb pressure tube material irradiated to a fast fluence of  $174 \times 10^{24} \text{ nm}^{-2}$ ,  $E > 1 \text{ MeV}$  at a nominal temperature of 250 °C. The observed dimensional changes in the longitudinal and transverse directions (up to  $\sim 1.2\%$  and  $\sim -0.5\%$ , respectively) are consistent with earlier data at 280 °C and 310 °C, and the previously reported negative temperature dependence. The observed growth in the radial direction is negative (up to  $\sim 0.7\%$ ). Initially, there is a small volume increase (0.05–0.1%) but this gradually decays to  $< 0.05\%$  and the long term rate of volume change is negligible, within the accuracy of the measurement, demonstrating that the phenomenon observed is, indeed, irradiation growth.

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## 1. Introduction

Irradiation growth is defined as shape change at constant volume in the absence of applied stress during irradiation [1]. For zirconium alloys, the approximate conservation of volume has been demonstrated in a few studies, see reference [2] for example, but generally, irradiation growth measurements have been made on thin walled tubes and relatively thin sheet, where measurements are made in only one or two dimensions. With few exceptions (see reference [3] for example) the phenomenon being examined is assumed to be growth, without any evidence being offered to support this contention. This has certainly been the case for studies on Zr–2.5Nb pressure tube material [4–7].

## 2. Materials

The material discussed here is taken from production CANDU cold-worked Zr–2.5Nb pressure tube H737 (104 mm diameter  $\times$  4.2 mm wall thickness). The manufacturing details, metallurgical characteristics and chemical composition of this tube are given in reference [8]. The specimens from which the irradiation growth measurements are taken are 17 mm wide (axial direction)  $\times$  18.4 mm high (transverse direction)  $\times$  4.2 mm (thickness direction) curved compact tension specimens from which the fracture properties of the tube were subsequently taken. This particular tube contained, by weight, 550 ppm Fe, 1096 ppm O and 2.6% Nb.

## 3. Irradiation conditions

The specimens with a fast neutron fluence of  $2.9 \times 10^{24} \text{ nm}^{-2}$ ,  $E > 1 \text{ MeV}$ , were irradiated in the light water U5 loop of NRU and Chalk River Laboratories

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in a fast neutron flux of about  $1.5 \times 10^{17} \text{ nm}^{-2}\text{s}^{-1}$ ,  $E > 1 \text{ MeV}$ . The insert was operated at  $265^\circ\text{C}$ . Since nuclear heating was low, the specimens had little temperature gradient ( $\ll 5^\circ\text{C}$ ) and were close to the water temperature.

The specimens with higher fast neutron fluences were irradiated in a central site of the OSIRIS Reactor (CEA, CEN de Saclay) where the fast neutron flux is about  $1.8 \times 10^{18} \text{ nm}^{-2}\text{s}^{-1}$ ,  $E > 1 \text{ MeV}$ . The OSIRIS insert contained 16 tensile specimens and 16 cantilever beam specimens as well as the 16 compact tension specimens of interest here. The insert was designed so that the specimens in the bottom half could be replaced periodically, allowing a number of different fluences to be obtained. Specimens were replaced after 2, 7 and 17 reactor cycles resulting in cumulative exposures of 2, 5, 10, 34 and 51 cycles giving peak fast neutron fluences of 6.8, 16.5, 27.5, 119 and  $174 \times 10^{24} \text{ nm}^{-2}$ ,  $E > 1 \text{ MeV}$ . The fast neutron fluences were estimated from the activation of copper wires. Each reactor cycle was about 20 days and the total elapsed time of the experiment was about seven years. The flux distribution in OSIRIS is a cosine in which the specimen string is centered. The minimum flux experienced by any specimen in each string is about 74% of that experienced by the specimen in the peak flux position.

The specimens were irradiated at a nominal temperature of  $250^\circ\text{C}$ , controlled to within  $\pm 10^\circ\text{C}$  by resistance heating. They were immersed in NaK to remove heat deposited by neutron and  $\gamma$ -radiation. The nuclear heating gave rise to a calculated temperature gradient of  $11^\circ\text{C}$  from center to surface of the compact tension specimens discussed here.

#### 4. Measurements

The measurements were made using a mechanical comparator equipped with two LVDT (linearly variable differential transformer) probes, Fig. 1. The specimens were clamped in one of three orientations, longitudinal, transverse and radial, Fig. 2. The axial positions of the probes were adjustable such that the LVDT probes were at the centre of their travel for the nominal dimension of the specimen in each of the three orientations. For each orientation, the specimens were positioned reproducibly using shims and blocks, and clamped by horizontal and vertical pneumatic rams. The sensitivities of the LVDT probes were calibrated using stepped gauge blocks, and each dimension was referenced to a standard, unirradiated specimen of the same nominal dimensions as the irradiated specimens.

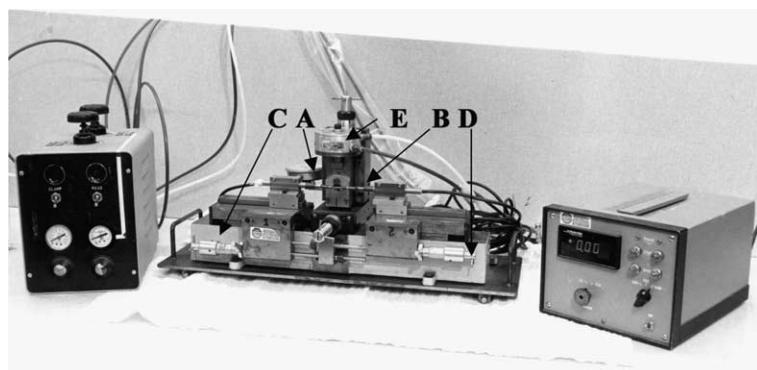


Fig. 1. Measurement rig (centre) with pneumatic controls (left) and LVDT readout (right). LVDT probes are indicated at A and B, adjustment slides at C and D, and vertical pneumatic clamp at E.

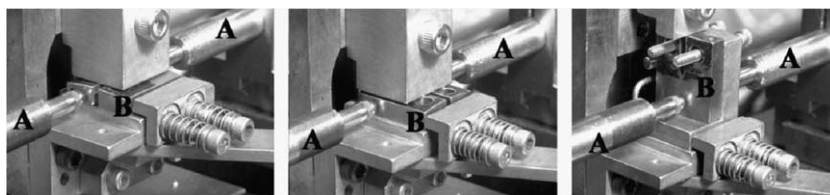


Fig. 2. Specimen clamped in mechanical comparator for measurements of longitudinal (left), transverse (centre) and radial dimension (right). Barrels of LVDT probes are indicated at A, specimen at B.

### 5. Results

The results are shown in Figs. 3 and 4. In addition to the strains in the longitudinal, transverse and radial directions of the specimen, the volume change for each specimen is shown. This has been calculated in two ways: Column BU refers to the volume change calculated by comparing the product of the three dimensions of the specimen after irradiation to the initial product of the three specimen dimensions and Column BF refers to the sum of the strains in the three directions. Fig. 1 shows that there is a small increase in volume (density

decrease) at low fluences (0.05–0.1%) which decreases slightly with fast fluence.

Fig. 2 shows that four specimens irradiated to fast fluences between  $90$  and  $130 \times 10^{24} \text{ nm}^{-2}$ ,  $E > 1 \text{ MeV}$  exhibited a significant volume decrease resulting from substantial negative strains in the normal direction. These are believed to be artifacts, resulting from some fretting of the specimens against their holders during irradiation. Ignoring these results is justified since the other results from that set show a continuous trend with the results from the final set, where such large negative strains are not observed. The volume change for fluences

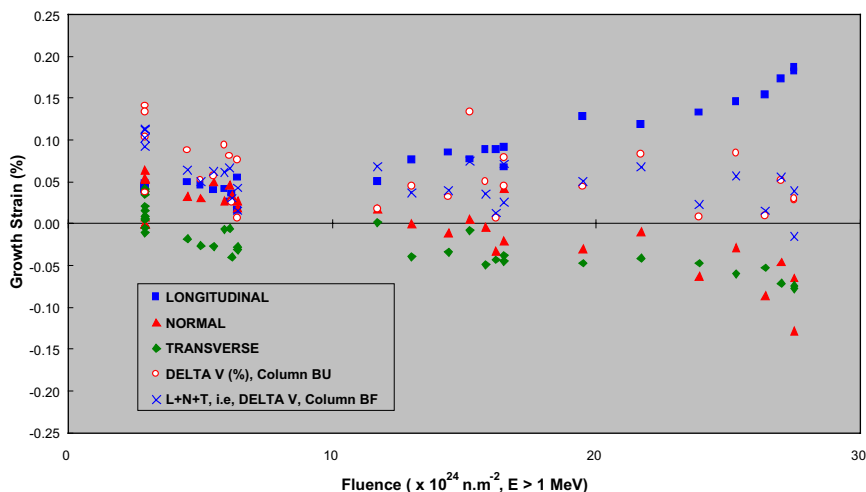


Fig. 3. Irradiation growth and volume changes in Zr–2.5Nb pressure tube H737 at low to medium fast neutron fluence. NRU results are at far left ( $2.9 \times 10^{24} \text{ nm}^{-2}$ ,  $E > 1 \text{ MeV}$ ). All other results are from Osiris.

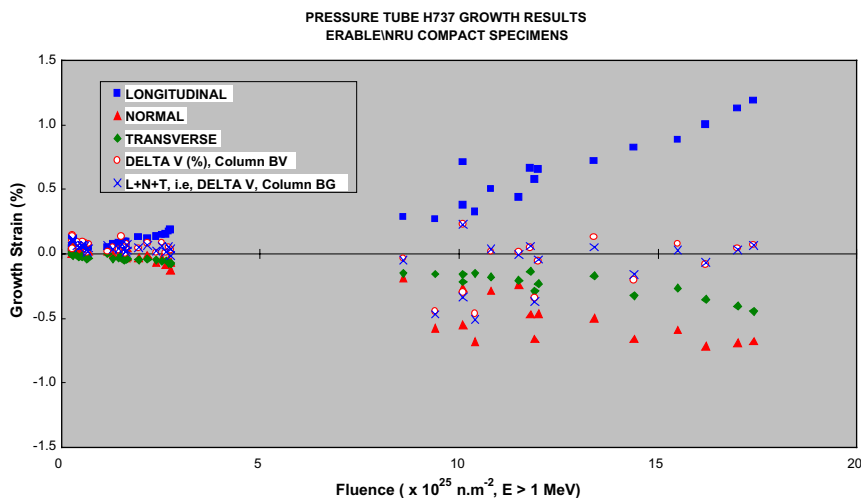


Fig. 4. Irradiation growth of Zr–2.5Nb and volume changes in pressure tube H737 to high fast neutron fluence. NRU results are at far left ( $0.29 \times 10^{25} \text{ nm}^{-2}$ ,  $E > 1 \text{ MeV}$ ). All other results are from Osiris.

of  $>75 \times 10^{24} \text{ nm}^{-2}$ ,  $E > 1 \text{ MeV}$  is thus zero, within the accuracy of the measurement. There may still be a small residual density decrease from lower fluences ( $<0.05\%$ ).

## 6. Discussion

The current results verify the interpretation of long term data for unstressed Zr–2.5Nb irradiated at temperatures near  $250^\circ\text{C}$  as irradiation growth, accounting for linear strains of  $>1\%$ . There is a small volume increase in the early stages of irradiation, between  $0.05\%$  and  $0.1\%$ , but this appears to gradually decay to fast fluences of  $30 \times 10^{24} \text{ nm}^{-2}$ ,  $E > 1 \text{ MeV}$ , and is less than  $0.05\%$  at high fast fluences where the rate of volume change is negligible within the accuracy of the measurements.

The irradiation growth strains reported here for the longitudinal and transverse directions are consistent with those previously reported at  $280^\circ\text{C}$  and  $310^\circ\text{C}$  for similar materials, Table 1 [7], considering the negative temperature dependence of the magnitude of the strain, Table 2. The ratio of transverse to axial strain, about  $-0.3$ , is numerically smaller than seen at the higher temperatures (typically  $-0.05$ ), suggesting that the growth anisotropy changes with temperature. This is consistent with earlier observations that, at even higher temperatures ( $414\text{--}433^\circ\text{C}$ ), the ratio of the transverse (negative) to axial (positive) growth strains is numerically even larger, about  $-1.4$  after,  $1.5 \times 10^{26} \text{ nm}^{-2}$   $E > 1 \text{ MeV}$ , Table 2.

These results verify also the anisotropy of the growth strain rate tensor that has been used for modeling the

deformation of Zr–2.5Nb pressure tubes [10] i.e., a substantially larger negative strain-rate in the normal direction than would be expected if the individual  $\alpha$ -Zr crystals exhibited isotropic expansion in the basal plane, accompanied by shrinkage along the  $c$ -axis. This could be attributed either to the role of the grain boundaries (the grain shape is highly anisotropic) or to variations in the single crystal strain rate tensors with crystal orientation. This has been hypothesized on the basis of observed variations in dislocation substructure with orientation in both Zircaloy-2 [9,10] and Zr–2.5Nb [7,11], and the growth of Zr–2.5Nb has been successfully modeled based on this assumption [12].

## Acknowledgments

The specimens that we report upon above were part of an experiment managed successively by C.E. Coleman, R.R. Hosbons and P.H. Davies. Our good friend and colleague, the late V. Fidleris initiated the measurement program on these specimens. M.A. Miller developed the measurement technique and performed the earlier measurements and A.K. MacCormack performed the later measurements.

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Table 1

Texture parameters (resolved fractions of basal plane normals),  $F_d$ , in the 'd' direction for the materials used in this and previous studies [7]

Material	$F_r$	$F_t$	$F_a$
A	0.34	0.63	0.04
B	0.32	0.59	0.09
C	0.31	0.61	0.08
This work	0.34	0.62	0.05

Table 2

Approximate growth strains interpolated to a fast fluence of  $125 \times 10^{24} \text{ nm}^{-2}$  for this and earlier work [7]

Material	Nominal temperature ( $^\circ\text{C}$ )	Longitudinal strain (%)	Transverse strain (%)
This work	250	0.7	-0.2
A – Ref. [7]	280	0.5 to 0.6	-0.2 to -0.3
B – Ref. [7]	280	0.6 to 0.7	-0.3 to -0.4
C – Ref. [7]	280	0.35 to 0.45	-0.2 to -0.25
Others – Ref. [7]	280	0.3 to 0.8	-
A – Ref. [7]	310	0.2 to 0.3	-0.1 to -0.2
B – Ref. [7]	310	0.3 to 0.4	-0.25
C – Ref. [7]	310	0.1	-
Others – Ref. [7]	310	0.0 to 0.4	-

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