

University of Western Ontario – Robert Klassen CRD

NSERC/UNENE Collaborative Research and Development Grant on Microindentation studies of the local ductility of Zr-2.5Nb CANDU pressure tubes

Principal Investigator: Robert J. Klassen
University of Western Ontario

Co-Investigator: Brad J. Diak
Queen's University



Overview

The objective of this research project is to use microindentation testing techniques; namely spherical- and pyramidal-microindentation, to assess the local mechanical properties of the Zr-2.5Nb alloy used in CANDU pressure tubes. Spherical micro-indentation is used to deduce the flow stress versus plastic strain relationships in the radial-, axial-, and transverse-directions of a pressure tube at temperatures from 25°C to 400°C and, thus, provide data on the directional anisotropy of the plastic flow properties. Pyramidal micro-indentation creep tests and indentation strain rate-change tests are used to assess the effect of indentation direction, temperature and irradiation damage (as simulated with Zr⁺ ion irradiation) on the parameters that define the kinetics of the plastic deformation of the Zr-2.5Nb alloy.

The data from this investigation will allow more accurate predictions to be made of the extent of local stress relaxation occurring as a result of creep deformation around surface flaws, such as scratches and fretting flaws, in CANDU pressure tubes during service and will provide a testing methodology that can potentially be used for in-cell assessment of local variations in the mechanical properties of irradiated fuel channel materials.

Outcomes from this research

1) Spherical micro-indentation testing of Zr-2.5Nb material
Spherical micro-indentation tests were performed at 25°C in the radial-, axial-, and transverse-directions of a non-irradiated Zr-2.5Nb pressure tube off-cut. The average indentation stress σ and plastic strain ε are determined from the measured indentation force F , indentation depth h , and the indenter contact radius a using the following expressions:

$$\sigma = \frac{F}{\psi h^2} \text{ and } \varepsilon = \alpha \left(\frac{a}{R} \right) \quad (1)$$

The parameters ψ and α were determined by fitting the calculated σ , ε values to σ - ε curves obtained from uniaxial compression tests performed on the same Zr-2.5Nb material. It was found that the values of ψ and α are different by about 15% for the axial-, radial-, and hoop-directions. This reflects the mechanical anisotropy of the highly textured Zr-2.5Nb pressure tube material when deformed at room temperature. Spherical indentation and uniaxial compression tests are now being done at elevated temperatures (100, 200, 300, and 400°C) and the methodology developed from the room temperature tests will be applied to analyse the data. This research will result in empirical expressions for ψ and α as a function of indentation direction and temperature which will allow the use of spherical micro-indentation testing to obtain σ versus ε curves for localized regions of pressure tube material without the need to make large uniaxial tension or compression specimens. This has an obvious application for obtaining σ - ε flow curves from radioactive material removed from nuclear reactors.

Pyramidal micro-indentation hardness tests were performed on sectioned spherical indentations to map the variation in hardness, and hence the variation in the equivalent plastic strain, around the indentation. Similar tests will be performed on spherical indentations made at 100, 200, 300, and 400°C. Knowing the distribution of plastic strain around spherical indentations made in pressure tube material in the three orthogonal directions at various temperatures will provide benchmark data against which researchers can assess the accuracy of their finite element models.

2) Microindentation creep testing of non-irradiated Zr-2.5Nb
Constant-force indentation creep tests, of one hour duration, were performed at indentation depths of $h_0 = 0.1, 0.5, 1.0, 5.0, 8.0,$ and $10.0 \mu\text{m}$ in the radial-, axial-, and transverse-directions of non-irradiated Zr-2.5Nb pressure tube material at temperatures of 25, 100, 200, 300, and 400°C.

The indentation strain rate $\dot{\varepsilon}_{ind}$ was calculated from the constant-force indentation creep test data and was expressed as a function of temperature T and indentation stress σ_{ind} by the following equation which expresses the strain rate resulting from a creep deformation process occurring by a mechanism of dislocation glide limited by interaction with discrete obstacles distributed in the microstructure.

$$\dot{\varepsilon}_{ind} = \dot{\varepsilon}_0 e^{-\frac{\Delta G_0}{kT} \left(1 - \left(\frac{\sigma_{ind}}{\sigma} \right)^p \right)^q} \quad (2)$$

In this equation ΔG_0 is the activation energy of the obstacles that are limiting the dislocation glide through the indented material. By fitting Eq. (2) to the measured $\dot{\varepsilon}_{ind}$ and σ_{ind} data ΔG_0 was found to increase with increasing temperature for the Zr-2.5Nb samples tested.

The magnitude of the measured $\dot{\varepsilon}_0$ and ΔG_0 suggests that the obstacles controlling the creep rate of Zr-2.5Nb at 25°C are dislocation / dislocation or dislocation/grain

boundary interactions. This was confirmed by TEM micrographs of foils that were extracted from directly below crept microindentations using focused ion beam milling.

3) Microindentation creep testing of ion-irradiated Zr-2.5Nb Constant-force indentation creep tests (of one hour duration) were also performed on Zr-2.5Nb pressure tube material that was irradiated with 8.5 MeV Zr^+ ions to levels corresponding to 5 and 30 displacements per atom (dpa) at a depth of about 3 μm . These levels of ion irradiation were chosen to simulate the microstructural damage imparted to Zr-2.5Nb from neutron irradiation over the lifetime of a CANDU pressure tube.

The initial indentation stress $\sigma_{ind \ t=0}$ was found to increase with increasing ion irradiation. $\sigma_{ind \ t=0}$ show a similar dependence upon indentation depth and indentation direction as the non-irradiated Zr-2.5Nb however ΔG_0 (Eq. 2) increases with increasing ion irradiation. This indicates that the ion damage induces a population of crystal defects into the Zr-2.5Nb that directly affect the activation energy associated with creep deformation.

4) Microindentation strain rate change tests of non-irradiated Zr-2.5Nb Pyramidal microindentation tests were performed, at room temperature, to indentation depths of 2 μm and 10 μm on the non-irradiated and ion-irradiated Zr-2.5Nb samples. Periodically during the tests the indentation strain rate was changed suddenly by a factor of ten to assess the effect of irradiation hardening on the strain rate sensitivity of the hardness of Zr-2.5Nb. The first stage of these tests were performed at Queen's university and the data are presently being analysed.

Research Facilities

This research project has made use of facilities located in three Canadian universities (the University of Western Ontario, Queen's University, and McMaster University). The majority of the microindentation tests were performed with the high-temperature microindentation and nanoindentation testing facilities located in the Faculty of Engineering at the UWO. The Zr^+ ion irradiations were performed with the Tandetron ion accelerator and the fabrication of TEM samples using focused ion milling was performed at the nanofabrication laboratory both of which are located in the Faculty of Science at the UWO. The TEM investigations were performed at the Brockhouse Institute for Materials Research at McMaster University. The microindentation strain rate change tests were performed in the Faculty of Engineering and Applied Science at Queen's University.

Research Team

Principal Investigator (R.J. Klassen)

- Primary organizer of the project. Recruited the four graduate students (2 PhDs and 3 MEngs) and one summer student.
- Set up the test matrix.

- Scheduled all testing.
- Liased with the supporting company (AECL (UNENE member)).

Co-investigator (B. Diak)

- Prof. Brad Diak is supervising the microindentation strain rate change tests at Queen's University. Prof. Diak's expertise in the kinetics of plastic deformation is essential in correctly interpreting the results of the pyramidal indentation creep and strain-rate change tests performed on the non-irradiated and the ion-irradiated Zr-2.5Nb samples.

PhD Grad. Student (B. Bose)

- Completed the bulk of the micro-/nano-scale indentation creep tests at temperatures of 25, 100, 200, 300, and 400°C.
- Completed all the indentation creep testing of the Zr⁺ ion irradiated (5 and 30 dpa) samples.
- Performed TEM analyses of the indentation plastic zone of non-irradiated and ion-irradiated Zr-2.5Nb samples.
- Performed indentation strain rate change tests at Queens University.

PhD Grad. Student (R. Oviasuyi)

- Finished the room temperature spherical micro-indentation testing and the uniaxial compression tests on the Zr-2.5Nb samples.
- Assessed the anisotropic plastic strain around spherical indentations.
- Performed uniaxial compression tests at 100, 200, 300, and 400°C.

MEng Grad. Students (N. Dorairaj, N. Islam, M.A. Bashar)

- Worked on characterising the plastic zone beneath spherical indentations.
- Worked on assessing the effect of internal oxidation on the indentation creep of zirconium alloys.

Publications

The findings from this research have resulted in two conference presentations [1,2] and one submitted journal paper [3].

1. V. Bhakhri and R.J. Klassen; "Haasen plot activation analyses of constant-force indentation creep" Oral presentation at the Materials Research Council Fall Meeting (Symposium AA), Boston MA, October 25 – 29, 2007.
2. B. Bose, R.J. Klassen; "Assessment of Local Plastic Deformation of Zr-2.5%Nb CANDU Pressure Tubes at 298K" presented at the 21st Canadian Materials Science Conference, Kingston ON, June 2009.
3. B. Bose, R.J. Klassen; "Effect of ion irradiation and indentation depth on the kinetics of deformation during microindentation of Zr 2.5Nb pressure tube material at 25°C", submitted to the Journal of Nuclear Materials, October 2009.

Interactions with Industrial Sponsors

The results that were achieved to date were presented at the Chalk River Laboratories (CRL) on May 12, 2009 to an audience of about 30 Researchers/engineers who were involved in pressure tube flaw assessments.

Considerable interest arose from this presentation particularly in the use of spherical micro-indentation testing for performing in-cell measurements of the mechanical properties of highly radioactive material.

Based upon the findings presented there was agreement in principle to:

- 1) Use spherical microindentation to test the flow stress of Zircaloy circular disks that were previously irradiated to high neutron fluence at the ATR reactor in Idaho Falls and which are now in storage at CRL.
- 2) Use the pyramidal nano-indentation techniques to assess the effects of Oxygen ingress and internal oxidation on the hardness of Zr-4 fuel sheathing exposed to high temperature similar to that during a LOCA event.

This demonstrates the usefulness of the findings of this study to players in the Canadian nuclear industry.