Fatigue Behavior of Pressure Tube Material Zr-2.5Nb in Air and in Simulated CANDU-Reactor Water Environments

Final Report

Nuclear Engineering Division

About Argonne National Laboratory

Argonne is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC under contract DE-AC02-06CH11357. The Laboratory's main facility is outside Chicago, at 9700 South Cass Avenue, Argonne, Illinois 60439. For information about Argonne and its pioneering science and technology programs, see www.anl.gov.

DOCUMENT AVAILABILITY

Online Access: U.S. Department of Energy (DOE) reports produced after 1991 and a growing number of pre-1991 documents are available free via DOE's SciTech Connect (<http://www.osti.gov/scitech/>)

Reports not in digital format may be purchased by the public from the National Technical Information Service (NTIS):

U.S. Department of Commerce National
Technical Information Service 5301
Shawnee Rd
Alexandra, VA 22312
www.ntis.gov
Phone: (800) 553-NTIS (6847) or (703) 605-6000
Fax: (703) 605-6900
Email: **orders@ntis.gov**

Reports not in digital format are available to DOE and DOE contractors from the Office of Scientific and Technical Information (OSTI):

U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
www.osti.gov
Phone: (865) 576-8401
Fax: (865) 576-5728
Email: **reports@osti.gov**

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor UChicago Argonne, LLC, nor any of their employees or officers, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of document authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof, Argonne National Laboratory, or UChicago Argonne, LLC.

Fatigue Behavior of Pressure Tube Material Zr-2.5Nb in Air and in Simulated CANDU-Reactor Water Environments

Final Report

prepared by
Meimei Li, W. K. Soppet, and K. Natesan
Nuclear Engineering Division, Argonne National Laboratory

Work supported by
Atomic Energy of Canada Limited/Chalk River Laboratories

September 2009

This page is intentionally left blank

Executive Summary

An experimental program was initiated to develop fatigue data in strain-controlled conditions for an assessment of the fatigue behavior of Zr-2.5Nb pressure tube material in support of CANDU reactors for Atomic Energy of Canada Limited (AECL)/Chalk River Laboratories (CRL). Fatigue tests were conducted on specimens fabricated from a Zr-2.5Nb pressure tube to determine the fatigue performance in air and in simulated CANDU-reactor water environments. This report summarizes the fatigue data of Zr-2.5Nb material obtained in both environments.

Fatigue tests in air were performed under constant total strain amplitudes in the fully reversed ($R=-1$) strain control. Fatigue behavior of Zr-2.5Nb pressure tube material was examined over a range of total strain amplitudes from 0.4 to 0.75% at 300°C with a balanced triangular wave form (equal strain rates in tension and in compression) at a strain rate of 0.4%/s. The temperature dependence of the fatigue response was examined at 20 and 100°C at total strain amplitude of 0.6%. The strain rate dependence of the fatigue performance was investigated by applying an unbalanced triangular waveform with a slower strain rate of 0.004%/s in tension and a faster strain rate of 0.4%/s in compression.

Fatigue tests in water environments were performed under the stroke control mode. The axial stroke response was correlated with the axial strain control from the air tests, and was used to provide control for the fatigue tests in the autoclave system. Fatigue behavior of Zr-2.5Nb pressure tube material was examined over a range of total strain amplitudes from 0.45 to 0.75% at 300°C with an unbalanced triangular waveform with a slower strain rate of 0.004%/s in tension and a faster strain rate of 0.4%/s in compression in low-dissolved oxygen (DO) LiOH-containing water.

Fatigue data were analyzed to obtain the strain-life relation, the cyclic stress response during fatigue cycling, half-life hysteresis loops, and half-life cyclic stress-strain curves. The cross-sectional fracture surfaces and longitudinal surfaces of the gage section of tested specimens were examined by scanning electron microscopy (SEM) to understand the fatigue crack initiation and propagation behavior in this material.

The fatigue life of Zr-2.5Nb was significantly reduced in simulated CANDU-reactor water environments at 300°C, compared to the fatigue data obtained in air. The synergistic effect of the aqueous environment and the strain rate is most detrimental to the fatigue resistance of Zr-2.5Nb. The fatigue life of Zr-2.5Nb in water under the slow-fast cyclic loading was reduced in both high-strain and low-strain amplitudes when compared with the fatigue data in air under the fast-fast cycling. The influence of the aqueous environment on the fatigue resistance is more pronounced in the high strain amplitudes than in the low strain amplitudes when compared with the fatigue data in air under slow-fast cyclic loading.

Significant reduction in fatigue life was observed at 300°C in air when a lower strain rate (0.004%/s) was applied in tension during cycling. The strain rate effect was more pronounced in the low strain amplitude and long fatigue life regime.

Fatigue behavior of Zr-2.5Nb was strongly affected by the test temperature. Fatigue life increased significantly as temperature decreased along with considerably higher elastic

strain component and lower plastic strain component at lower temperatures. It is apparent that the fatigue performance of Zr-2.5Nb at low temperatures is dominated by the material strength.

The cyclic stress response during cycling at 300°C can be characterized by subsequent transitional cyclic softening, cyclic saturation, and secondary cyclic hardening. Secondary cyclic hardening was more pronounced at lower strain amplitudes and long fatigue lives. The cyclic stress response was strongly affected by the strain rate, and less sensitive to the applied strain amplitude and the test temperature. The extent of cyclic softening in the cyclic stress response was less when a slow strain rate was applied in tension at 300°C. The cyclic stress-strain response showed an inverse strain-rate dependence of cyclic strain hardening. While the aqueous environment has little influence on the cyclic stress-cycle profile, the cyclic strain hardening at the half-fatigue life was much more pronounced under the combined effect of the low strain rate and the aqueous environment.

The fracture surfaces of all fatigue-tested specimens showed typical surface crack initiation, crack propagation fatigue zone, and fast fracture zone. A single crack initiation site was often observed in specimens tested at lower strain amplitudes and lower temperatures, while multiple crack initiation sites were observed in specimens tested at higher strain amplitudes and higher temperatures. Fatigue zones were covered with striation-like features and heavy laminated secondary cracking in all the specimens. Early onset of stage II crack propagation was evident in specimens with shorter fatigue lives. Fatigue striations were well developed in the fatigue-tested specimens in water. Secondary surface cracking along the gage was more pronounced at higher strain amplitudes, higher temperature, and lower strain rates. The aqueous environment has little influence on the fatigue fracture modes of Zr-2.5Nb, but secondary surface cracking along the gage was more severe in the specimens tested in water than in air.

Several outstanding issues have been identified, based on the current study on fatigue behavior of Zr-2.5Nb pressure tube material. Proposed future experiments include:

- Fatigue tests in simulated water environment under fast strain rate (FSR) cycling to eliminate the strain rate effect are needed to develop a better understanding of the influence of the aqueous corrosion on the fatigue resistance of Zr-2.5Nb.
- Fatigue tests in simulated water environment at lower temperatures, where the dynamic strain aging (DSA) effect is suppressed, are important to understand the possible role of the DSA process and its coupling effect with corrosion in the fatigue response of Zr-2.5Nb.
- Fatigue crack growth rates in simulated water environment are crucial information for the structural integrity of Zr-2.5Nb pressure tubes. Fatigue crack growth experiments in simulated CANDU-reactor water environments are highly recommended.
- Further examination of the strain rate dependence of the fatigue response is needed by applying a slow strain rate in compression, and a slow strain rate in both tension and compression.

- Fatigue tests at a wide range of temperatures and strain amplitudes to fully understand the temperature dependence and the role of DSA process in the fatigue performance.
- Detailed microstructure characterization to understand the deformation modes and crack initiation and propagation mechanisms.
- The role of texture of the material in the fatigue performance needs evaluation by developing data on specimens in both longitudinal and transverse directions.
- Perform fatigue tests on specimens taken from various locations of a pressure tube to evaluate the effect of variations in grain structure and crystallographic texture along the pressure tube.
- Fatigue data of Zr-2.5Nb with surface roughness and surface flaws to assess the fatigue resistance in the pressure tubes with flaws produced during manufacturing and/or operation.
- Fatigue tests of Zr-2.5Nb pressure tube material with oxidized surface layer to evaluate the effect of surface oxidation on fatigue life.
- Determine the effect of hydrogen concentration and/or hydrides and their orientation on the fatigue properties of the Zr-2.5Nb pressure tubes.
- Develop a fatigue life prediction model that encompasses the effects of aqueous environment, temperature, strain rate, texture, oxidation, and hydride distribution on fatigue performance.

This page is intentionally left blank

Table of Contents

Executive Summary	iii
1. Introduction.....	1
2. Experimental Procedure.....	2
2.1 Materials and Specimens	2
2.2 Test Systems.....	3
2.3 Fatigue Tests.....	10
2.4 Scanning Electron Microscopy (SEM).....	11
3. Experimental Results	12
3.1 Fatigue Response in Air	12
3.1.1 Effect of Temperature on Fatigue Behavior	14
3.1.2 Effect of Strain Rate on Fatigue Behavior	15
3.2 Fatigue response in Water	17
3.3 SEM Examination.....	20
3.3.1 Fractography	20
3.3.2 Surface Cracking.....	37
4. Discussion	43
5. Conclusions.....	48
6. Recommended Future Work	49
References	51

This page is intentionally left blank

Fatigue Behavior of Pressure Tube Material Zr-2.5Nb in Air and in Simulated CANDU-Reactor Water Environments

Meimei Li, W. K. Soppet, and K. Natesan
Nuclear Engineering Division
Argonne National Laboratory
Argonne, IL 60439

1. Introduction

Zirconium alloys are one of the most important engineering materials in nuclear technology. These alloys have low neutron absorption cross section, adequate corrosion resistance, excellent compatibility with uranium fuels, and good high temperature properties. Zr-2.5Nb is the pressure tube material in CANDU Pressurized Heavy Water Reactor (PHWR). Zr-2.5Nb pressure tubes and Type 403 stainless steel end fittings constitute the primary pressure boundary in a modern CANDU PHWR such as the Wolsong Unit-2 Reactor. The seamless pressure tubes have the dimensions of 640 mm in length, 103 mm in outside diameter (OD), and 4.1 mm in wall thickness, and contain fuel assemblies and heavy water coolant pressurized to 10.3 to 11.1 MPa and maintained at inlet and outlet temperatures of 266 and 312°C, respectively. Assurance of long-term integrity of the pressure tubes, end fittings, and their rolled joints within the design life span is crucial to safe, reliable, and economic operation of the reactor.

Zr-2.5Nb is susceptible to degradation of mechanical properties from reactor service environments (i.e., corrosive media, high stress, high temperature, etc.). During reactor operation, small defects and subcritical-size flaws are present in pressure tubes, either as a result of hydride blistering during operation or fabrication flaws. Some flaws can be produced during tube handling at the time of reactor construction or as a result of fuel assembly vibration and fretting during operation. When a crack initiates and propagates, several crucial information must be known for safe operation of a reactor, namely, crack growth rate under cyclic loading, effect of water chemistry on the fatigue properties, subcritical crack growth rate, size of the crack when it penetrates through wall and a leakage occurs, whether or not the leakage can be detected, and whether a sufficient time is available to shut down the reactor safely before the crack has grown to a critical size leading to an unstable fast fracture. The concern of structural integrity in pressure tubes in PHWRs requires systematic assessment of fatigue performance and accurate prediction of lifetime of Zr-2.5Nb in reactor environments.

Very little information is currently available on the fatigue properties of Zr-2.5Nb pressure tube material. The available fatigue data on Zr-2.5Nb were reported by Scarth et al [Scarth et al 1995] and by Hosbons and Wotton [Hosbons and Wotton 1996]. Scarth et al performed fatigue tests under stress-controlled conditions in tension with $R = \sigma_{\min}/\sigma_{\max} = 0$. Unnotched ring specimens were tested at 303°C using unirradiated and irradiated Zr-2.5Nb material and notched ring specimens were tested at 20°C using irradiated Zr-2.5Nb material. These fatigue data were not considered for developing the fatigue design curve due to concerns of stress-controlled conditions and the assumption of elastic behavior. In the study by Hosbons and Wotton, a limited number of fatigue tests were carried out on

unirradiated and irradiated Zr-2.5Nb in air and in water at 300°C. Fatigue tests were carried out under constant load control either in tension-compression with $R = -1$ or in tension-tension with $R = 0.1$. The results showed that the fatigue properties of Zr-2.5Nb are anisotropic and are affected by both neutron irradiation and water chemistry. A fatigue design curve was generated from these test data. However, the design curve was not adopted because of concerns with the stress-controlled method and the water chemistry control. Strain-controlled conditions and the strain-life approach are regarded more appropriate for developing the fatigue design life, particularly at high load levels where the material response is elastic-plastic.

An experimental program was initiated at Argonne National Laboratory (ANL) to develop fatigue data under strain-controlled conditions for proper assessment of the fatigue behavior of Zr-2.5Nb pressure tube material in support of CANDU reactors for Atomic Energy of Canada Limited (AECL)/Chalk River Laboratories (CRL). Fatigue tests were carried out on specimens fabricated from as-received pressure tube material to determine the fatigue behavior in air and in simulated CANDU-reactor water environments. Fatigue tests in air were carried out under constant total strain amplitudes in fully-reversed ($R = -1$) strain control. Fatigue behavior of Zr-2.5Nb was examined at 300°C over a range of total strain amplitudes from 0.4% to 0.75% at a strain rate of 0.4%/s. The temperature effect on fatigue performance was investigated by conducting tests at 20 and 100°C. The strain rate dependence of the fatigue response was investigated by applying a slower strain rate in tension (0.004%/s). Fatigue tests in water environments were carried out under the stroke control mode. The axial stroke response was correlated with the axial strain control from the air tests, and was used to provide control for the fatigue tests in the autoclave system. Fatigue behavior of Zr-2.5Nb pressure tube material was examined in low dissolved oxygen (DO) LiOH water over a range of total strain amplitudes of 0.45 - 0.75% at 300°C with a slower strain rate of 0.004%/s in tension and a faster strain rate of 0.4%/s in compression. Fatigue data were analyzed to obtain the strain-life relation, the cyclic stress response during fatigue cycling, half-life hysteresis loops, and half-life cyclic stress-strain curves. The cross-sectional fracture surfaces and longitudinal surfaces of the gage section of tested specimens were examined by scanning electron microscopy (SEM) to understand the fatigue crack initiation and propagation behavior. This report summarizes the fatigue data in air and water, and discusses the effects of several parameters including water environment, strain amplitude, strain rate, and temperature on the fatigue behavior of Zr-2.5Nb pressure tube material.

2. Experimental Procedure

2.1. Material and Specimens

The material examined in this study was Zr-2.5Nb pressure tube material (Heat # 243981, Tube # RX213) with the ingot chemical composition (in wt%): Nb 2.67, O 0.128, Fe 0.1, C 0.007, and Zr balance. According to the literature [Coleman et al 1980, Bickel and Griffiths 2008, Holt 2008, Holt et al 1987, Holt et al 2003], Zr-2.5Nb has a two-phase ($\alpha + \beta$) structure with the elongated grains of hexagonal close packed (hcp) α -phase surrounded by a network of thin filaments of body centered cubic (bcc) β -phase. The α -phase is a slightly super-saturated solid solution containing 0.6-1.0% Nb. The α -grains have a high grain aspect ratio (varying from 5 to 20) with the average grain width of 300-400 nm. A thin film of β -phase (β -Zr or β -Nb) has a thickness of 20-50 nm. In Zr-2.5Nb alloy, a dislocation

Fatigue specimens were fabricated as per the specifications developed jointly by AECL and ANL in accordance with ASTM 606 specification. Specimens had a nominal gage diameter of 0.125 in and gage length of 0.5 in. Specimens were cut from longitudinal blanks extracted from a ≈ 0.165 in wall of 4.425 in diameter pressure tube and were then machined to shape and final dimensions shown in Fig. 1. Prior to fatigue testing, all specimen gage sections were mechanically polished to 1- μm finish using a diamond paste. Polishing was performed in the axial direction to prevent circumferential scratches that may lead to initiation of cracks.

Two systems were utilized for conducting the fatigue tests, one for air environment and the other for simulated water environment. Both test systems contain the following components: a MTS model 312 closed-loop hydraulic material test frame rated at a maximum force of 20 kip, MTS hydraulic power supply, manifold assembly, load cell, hydraulic load-train grips, Instron 8500+ Dynamic Materials Testing System, thermocouple scanner, X-Y chart recorder, and a computer with LabVIEW® software application for data acquisition. The system used for conducting air tests is shown in Fig. 2. The system utilizes a Lepel model T-2.5-1-KC1-BW induction heating system, an axial strain extensometer for feedback control, and an LVDT stroke extensometer for correlation with the axial strain transducer. The Instron 8500+ hydraulic control system consists of a frame transducer interface box (FIB), hydraulic manifold interface box (HIB), master hydraulic manifold, actuator control panel, model 8500 dynamic control tower (Tower) and operator control/display (Console). The axial strain extensometer was designed following

1. MTS Test Frame Base
2. Hydraulic Service Manifold
3. MTS Actuator & Servo Valve
4. MTS Test Frame Crosshead
5. Load cell
6. Hydraulic Grips
7. Lepel Induction Heater
8. Instron Frame Interface Box
9. Instron 8500+ Control Tower
10. Data Acquisition Computer
11. Master Hydraulic Control
12. Instron 8500+ Console
13. Thermocouple Scanner

1. MTS Hydraulic Grips
2. Pull Rod
3. Induction Heating Coils
4. Extensometer Alumina Probes
5. Extensometer LVDT Assembly
6. Extensometer Balance Weights
7. Stroke LVDT Assembly
8. Stroke Quartz Support Rods

Figure 2. Fatigue test facility for tests in air environment.

"ASTM-E606 Standard Practice for Strain-Controlled Fatigue Testing." It utilizes alumina knife-edge contact probes set around an Invar spring hinge that provides mechanical amplification to the LVDT assembly opposite the contact probe. Interfaced with the Instron 8500+ Dynamic Materials Testing System, the axial strain extensometer has a resolution of 0.002% strain.

The second fatigue test system is equipped with a resistance-heated tube autoclave and a recirculating or once-through water system for controlling the aqueous environment, as shown in Fig. 3. The figure illustrates the MTS load frame with the hydraulic actuator and servo-valve below the frame platen and adjustable crosshead at the top. The load train consists of the specimen and the pull rods surrounded by the tube autoclave, hydraulic specimen grips and load cell transducer secured to the crosshead. Fatigue tests performed with the autoclave system use only an LVDT stroke extensometer attached to the pull rods outside of the autoclave for feedback control. The autoclave preheater and ECP cell can be seen to the left and right of the autoclave. The high-pressure pump and water-system pressure gage are visible at the bottom front of the MTS load frame. An instrument cabinet with temperature controllers for the autoclave, preheater and ECP cell is partially visible opposite the test system. The instrument cabinet also contains the power switch for the high-pressure pump and a high-temperature limit control. Above the Instron 8500+ control system is a thermocouple scanner system that monitors the autoclave thermal profile, ECP cell, preheater, and heat exchanger temperatures. Next to the thermocouple scanner is a console for monitoring the following in-line water quality transducers: pressure, flow, conductivity, and pH. Both temperature and water quality systems are interfaced to the computer data acquisition system (DAS) for synchronized recording with fatigue test data.

Figure 4 shows a schematic of the autoclave with the aqueous recirculating system used for fatigue tests with simulated water chemistry. The system consists of a 132-liter closed feedwater supply tank, Pulsafeeder™ high pressure pump, regenerative heat exchanger, autoclave preheater, tube autoclave, electrochemical potential (ECP) cell, back-pressure regulator, ion-exchange bed, 0.2 micron particulate filter, and a return line to the supply tank. Water is circulated through the autoclave at a rate of ≈ 10 ml/min. A recirculating pump on the ion-exchange cleanup system maintains water quality and in-line sensors monitor system pressure, flow rate, conductivity, and pH. Water from the back-pressure regulator in the once-through flow system is released to the drain, and in the recirculating flow system into an ion-exchange cleanup system that is part of the storage/supply tank. The tube autoclave, manufactured by ANL Central Shop, is constructed of Type 304SS and contains a press-fit titanium liner. The supply tank and most of the low-temperature piping are made of Type 304SS. Titanium tubing is used in the heat exchanger, connections to the autoclave, and to the ECP cell. The feedwater storage tank has either a nitrogen/oxygen or hydrogen cover gas to maintain a desired dissolved gas concentration in the water. For fatigue tests in low dissolved-oxygen (DO) environments, the ECP cell and the ion-exchange filter system in the return line from the autoclave to the water supply tank are by-passed during recirculation.

1. Test Frame Crosshead
2. Hydraulic Actuator
3. System Pressure Gage
4. High Pressure Pump
5. Recirculation Pump
6. Supply Tank
7. Instron Control Tower
8. Data Acquisition Computer
9. X-Y Plotter
10. Instrument Cabinet
11. Thermocouple Scanner
12. Water Quality Monitor

1. MTS Test Frame Crosshead
2. Test Frame Base/ Platen
3. Load Cell
4. Hydraulic Grips
5. Pipe Autoclave
6. Autoclave Preheater
7. ECP Cell
8. Heat Exchanger
9. High Pressure Pump
10. Instron Frame Interface Box
11. Master Hydraulic Control
12. Instron Control Console
13. Instron 8500+ Control Tower
14. Data Acquisition Computer
15. Thermocouple Scanner
16. Water Quality Monitor Console

Figure 3. Fatigue test facility for tests in simulated water environment.

Figure 4. Schematic of autoclave water recirculation system.

Both fatigue test systems are capable of being operated in load, actuator position, axial strain, and axial stroke control modes. For tests conducted at elevated temperature in the high-pressure autoclave, an axial extensometer could not be attached to the specimen gage section and therefore, an axial stroke transducer is utilized. Axial stroke operation produces specimen deformation by controlling the displacement of two fixed points on the load train surrounding the specimen outside of the autoclave and gage length regions. The control mode response for servo-hydraulic fatigue test systems is a function of frame/actuator stiffness, compliance of the specimen load train, hydraulic pressure, servo valve flow rate and frequency response. Following transducer calibration, the hydraulic closed-loop control system feedback parameters proportional gain, integral reset and derivative rate (PID) were tuned for each of the four test control modes. These PID parameters are necessary to ensure that system response tracks programmed input command LCF waveforms with minimal error.

The Instron 8500+ Dynamic Materials Testing System control firmware provides the ability to input known constants for hydraulic pressure and frame/actuator stiffness used in the digital control algorithm, but the PID parameters for each control mode were tuned empirically for the operating ranges stated for the test plan. ANL specimen ZNB25-5 was used for the PID tuning process.

Prior to fatigue testing, calibration was performed on primary transducers integral to both MTS/ Instron fatigue test systems. Load cells and hydraulic actuator position control LVDTs were calibrated on-site by Instron Corporation. The calibrations were performed to NVLAP, ANSI-NCSL Z540-1 and ISO 9001 accrediting standards. An MTS Model 650.03 extensometer calibration fixture, used for standardizing the axial extensometer and stroke transducers, was calibrated by ANL Technical Services Division Inspection Department to NIST traceable standards.

After tuning the hydraulic control system feedback parameters, the Lepel induction heating system was setup for the air test system to determine the temperature profile for the specimen and load train assembly for operation at 300°C. The temperature profile data is used to ascertain gage length thermal uniformity and correlate axial stroke response with axial strain control that was subsequently used to provide control for the fatigue tests in the autoclave system. Fifteen Type-K thermocouples, 0.010-in dia gage, were spot welded at several positions above and below the gage length center and along the load train. A schematic depicting thermocouple placement locations is shown in Figure 5.

Figure 5. Thermocouple placement locations on load train for temperature profile measurement.

For fatigue tests conducted at elevated temperature, only two (TC+2 and TC-2 positions) of five thermocouples are spot welded to the specimen to avoid imparting undesirable flaws along the gage length. These two thermocouple locations are also used for temperature set-point control and over-temperature limit protection. Establishing a uniform temperature profile along the specimen gage length entailed an iterative process of induction coil design, variable inductance oscillating tank coil tap setting, grid coil/power triode bias setting, and RF output frequency. The temperature profiling process was conducted under load control mode with a zero-stress set point. Figures 6 and 7 show typical temperature profiles for the overall load train and the specimen region. Temperature control obtained along the specimen gage length is within $\pm 3^{\circ}\text{C}$.

Figure 6. Temperature profile for overall load train and fatigue specimen by induction heating.

Figure 7. Temperature profile for fatigue specimen obtained by induction heating.

Fatigue Tests

Fatigue behavior of Zr-2.5Nb in the longitudinal direction was examined in air at 300°C over a range of total strain amplitudes from 0.4% to 0.75%. Fatigue tests were carried out under constant total strain amplitudes in fully-reversed ($R = -1$) strain control with a balanced triangular wave form (equal strain rates in tension and compression) at a strain rate of 0.4%/s (referred to as “fast strain rate (FSR) cycling”). The effect of strain rate on the fatigue response was investigated by applying a waveform with a slower strain rate in tension (0.004%/s) and the original strain rate in compression (0.4%/s) as in the fast strain rate cycling. This type of loading is referred to as “slow strain rate (SSR) cycling”. The waveforms of FSR and SSR cycling are schematically shown in Fig. 8. Fatigue tests were also carried out at 20 and 100°C at total strain amplitude of 0.6% to examine the effect of test temperature on fatigue behavior.

Figure 8. Waveforms of fast strain rate (FSR) cycling and slow strain rate (SSR) cycling.

Fatigue behavior of Zr-2.5Nb was examined in simulated water environment at 300°C in a high-pressure autoclave under the pressure of 1500 psi. The simulated CANDU-reactor water environment consisted of high-purity water containing <10 ppb DO achieved by a N_2+H_2 gas mixture for cover gas and with the addition of ≈ 2 ppm LiOH with a pH value of ≈ 10.5 and conductivity of 70-80 $\mu S/cm$. Fatigue tests were conducted over a range of total strain amplitudes under FSR and SSR cycling conditions. A summary of fatigue test conditions is given in Table 1. The water chemistry conditions for various tests are listed in Table 2.

The fatigue life was defined as the number of cycles when a load drop was greater than 25%. Fatigue properties were measured from half-life hysteresis loops. The elastic strain amplitude, $\Delta\epsilon_e/2$ was calculated using the following equation:

$$\Delta\epsilon_e/2 = \Delta\sigma/E \quad (1)$$

where $\Delta\sigma$ is the stress amplitude at half fatigue life, and E is the Young's modulus. The room temperature Young's modulus of 95 GPa was used in the calculations [Hosbons and Wotton 1996], and the temperature-dependence of the Young's modulus was not considered in the calculations. The plastic strain amplitude, $\Delta\epsilon_p/2$, was measured as the half of the maximum width of the half-life hysteresis loop.

Table 1. Summary of fatigue test conditions.

Test No.	Specimen ID	Total strain amplitude (%)	Strain rate in tension/compression (%/s)	Test type	Test temperature (°C)	Environment
AECL-7	A10A	0.4	0. 4/0.4	FSR	300	air
AECL-9	A14A	0.45	0. 4/0.4	FSR	300	air
AECL-4	A5A	0.5	0. 4/0.4	FSR	300	air
AECL8	A12A	0.5	0. 4/0.4	FSR	300	air
AECL-5	A7A	0.6	0. 4/0.4	FSR	300	air
AECL-10	A16A	0.65	0. 4/0.4	FSR	300	air
AECL-11	A18A	0.75	0. 4/0.4	FSR	300	air
AECL-16	A28A	0.5	0.004/0.4	SSR	300	air
AECL-14	A24A	0.6	0.004/0.4	SSR	300	air
AECL-15	A26A	0.75	0.004/0.4	SSR	300	air
AECL-12	A20A	0.6	0. 4/0.4	FSR	100	air
AECL-20	A36A	0.6	0.004/0.4	SSR	100	air
AECL-13	A22A	0.6	0. 4/0.4	FSR	20	air
AECL-18	A32A	0.6	0.004/0.4	SSR	20	air
AECL-21	A1B	0.75	0.004/0.4	SSR	300	HP water
AECL-22	A2B	0.75	0.004/0.4	SSR	300	LiOH water
AECL-23	A4B	0.6	0.004/0.4	SSR	300	LiOH water
AECL-24	A5B	0.5	0.004/0.4	SSR	300	LiOH water
AECL-25	A7B	0.45	0.004/0.4	SSR	300	LiOH water
AECL-26	A9B	0.65	0.004/0.4	SSR	300	LiOH water

Table 2. Water chemistry details monitored during water tests.

Test #	Dissolved oxygen concentration (ppb)	Dissolved hydrogen concentration (cc/kg @STP)	Pressure (MPa)	Flow rate (cc/min)	pH _{RT} Inlet	Conductivity Inlet (μS/cm)	pH _{RT} Effluent	Conductivity Effluent (μS/cm)
AECL-21	≤ 10	≈ 0	10.3	15	6.55	≤ 0.6	-	-
AECL-22	≤ 10	9.7	10.3	13	10.59	69	10.35	80
AECL-23	≤ 10	3.4	10.3	13	10.45	80	10.28	81
AECL-24	≤ 10	5.33	10.3	14	10.50	80	10.48	80
AECL-25	≤ 10	5.45	10.3	16	10.50	80	10.43	79
AECL-26	≤ 10	5.45	10.3	15	10.40	80	10.38	79

AECL-21 test was conducted in high -purity water;

All other tests were conducted in LiOH-containing (2 ppm Li) water.

2.4 Scanning Electron Microscopy

The cross-sectional fracture surfaces of fatigue-fractured specimens were examined by using a Hitachi S-4700 scanning electron microscope (SEM) operated at 15kV. The longitudinal surfaces of the gage section were also examined by SEM to understand the fatigue cracking behavior.

3. Experimental Results

3.1 Fatigue Response in Air

The strain-life curve for Zr-2.5Nb tested at 300°C in air under FSR cycling is shown in Fig. 9 along with contributions of the elastic and plastic strain components. The elastic strain component dominates the fatigue response over the examined strain ranges. The elastic strain-life relation can be represented by:

$$\frac{\Delta \varepsilon_e}{2} = 0.454 (N_f)^{0.033} \quad (2)$$

The plastic strain-life relation can be represented by:

$$\frac{\Delta \varepsilon_p}{2} = 10.467 (N_f)^{-0.504} \quad (3)$$

Figure 9. Strain- life response in terms of total, elastic and plastic strain amplitudes for Zr-2.5Nb tested under FSR cycling at 300°C in air.

Representative hysteresis loops at half fatigue life are shown in Fig. 10. At higher strain amplitude and shorter fatigue life, the hysteresis loop is wider, representing more plastic strain. Cyclic strain hardening behavior is evident.

The cyclic stress response during fatigue cycling at different strain amplitudes is shown in the linear-log plot in Fig. 11. The initial cyclic stress was increased as the total strain amplitude increased. The stress-cycle profiles were characterized by successive stages of transitional cyclic softening, cyclic saturation, and secondary cyclic hardening (the terminology is used to differentiate from initial cyclic hardening), depending on the total strain amplitude. Significant cyclic softening was observed at the initial fatigue stage and was persistent up to more than 1000 cycles. This transient cyclic softening was followed by cyclic saturation manifested by a plateau in the stress response, and subsequent secondary cyclic hardening. The secondary cyclic hardening is well illustrated in Fig. 12 when the stress-cycle curves are plotted in the linear-linear scale. At high strain amplitudes

and short fatigue lives (e.g. 0.75% and 0.65%), only cyclic softening was observed before failure.

Figure 10. Representative hysteresis loops at the half fatigue life for Zr-2.5Nb tested under FSR cycling at 300°C in air.

Figure 11. Variation of stress amplitude with number of cycles in the linear-log scale for Zr-2.5Nb tested under FSR cycling at 300°C in air.

Figure 12. Variation of stress amplitude with number of cycles in the linear-linear scale for Zr-2.5Nb tested under FSR cycling at 300°C in air.

3.1.1 Effect of Temperature on Fatigue Behavior

The effect of temperature on the strain-life response for Zr-2.5Nb is shown in Fig. 13. Though only one data point (at total strain amplitude of 0.6%) for each temperature is available to examine the temperature dependence, a strong effect of temperature on the fatigue life is evident. Fatigue life increased significantly as temperature decreased. The fatigue life at 20°C is more than three times longer than that at 300°C. The elastic strain component was considerably higher, and the plastic strain component was lower at the lower temperatures. The plastic strain data at 20 and 100°C fall on the plastic strain-life curve of the 300°C fatigue tests. It seems that the elastic strain component dominated for a greater portion of the fatigue life at lower temperatures than at higher temperatures at total strain amplitude of 0.6%. Note that the calculations of the elastic strain at 100 and 300°C were not corrected by the temperature-dependent Young's modulus.

The effect of temperature on the cyclic stress response over fatigue cycles is shown in Fig. 14 for the total strain amplitude of 0.6%. The stress level was much higher at lower temperatures. Transitional cyclic softening was evident at all temperatures. Moderate secondary cyclic hardening was observed at 300°C, and gradually vanished as temperature decreased. No secondary cyclic hardening was observed at 20°C, even though the fatigue life at 20°C was significantly longer than that at 300°C.

Figure 13. Effect of temperature on the strain-life response for Zr-2.5Nb, tested under FSR cycling between 20 and 300°C in air.

Figure 14. Effect of temperature on the cyclic stress response for Zr-2.5Nb tested under FSR cycling between 20 and 300°C in air.

3.1.2 Effect of Strain Rate on Fatigue Behavior

Figure 15 shows the effect of strain rate on the strain-life response in Zr-2.5Nb tested between 20 and 300°C in air. Reduction in fatigue life was observed when a slower strain rate was applied in tension during cycling. The effect was more pronounced in the low strain amplitude and long fatigue life regime. A 60% reduction in the fatigue life was observed at total strain amplitude of 0.5%. The effect of strain rate on the elastic strain-life curve was insignificant, while the plastic strain-life curve under SSR cycling shifted toward shorter fatigue lives compared to that under FSR cycling. The elastic strain dominated for a greater portion of the life under SSR cycling than under FSR cycling. The half-life hysteresis loops at total strain amplitudes of 0.5% and 0.75% are shown in Fig. 16 for tests under FSR and SSR cycling at 300°C. The elastic strain-life relation for the SSR cycling at 300°C can be represented by:

$$\frac{\Delta\epsilon_e}{2} = 0.667(N_f)^{-0.085} \quad (4)$$

The plastic strain-life relation for the SSR cycling at 300°C can be represented by:

$$\frac{\Delta\epsilon_p}{2} = 10.375(N_f)^{-0.539} \quad (5)$$

Figure 15. Effect of strain rate on the strain-life response for Zr-2.5Nb tested between 20 and 300°C in air.

Figure 16. Half-life hysteresis loops at total strain amplitudes of 0.5% and 0.75% for Zr-2.5Nb tested under FSR and SSR cycling at 300°C in air.

The effect of strain rate on the cyclic stress response during fatigue cycling at 300°C is shown in Fig. 17. While the stress-cycle profiles of the SSR and FSR cycling showed similar characteristics of transitional cyclic softening, cyclic saturation, and secondary cyclic hardening, the stress amplitudes at a given number of cycles were higher at the lower strain rate than those at the higher strain rate, except that the stress amplitudes in the first few cycles were not affected by the strain rate.

Figure 17. Effect of strain rate on the cyclic stress response for Zr-2.5Nb tested under FSR and SSR cycling at 300°C in air.

The effect of strain rate on the fatigue life was insignificant when Zr-2.5Nb was tested at 20 and 100°C, as shown in Fig. 15. The half-life elastic strain amplitude was similar, while the half-life plastic strain component was slightly higher under SSR cycling than under FSR cycling. The effect of strain rate on the cyclic stress response is different as temperature varies (shown in Fig. 18). The stress amplitude in the first few cycles decreased by applying a slow strain rate in tension at 20 and 100°C, while the initial stress amplitude was not affected at 300°C when the strain rate was reduced. As cycling progressed, the cyclic stress profile was relatively unchanged in fatigue tests at 20°C at two different strain rates, while the decrease in stress amplitude diminished when the number of cycles achieved $\approx 60\%$ of the fatigue life at 100°C. The extent of overall cyclic softening was less at the lower strain rate than that at higher strain rate at 300°C.

Figure 18. Effect of strain rate on the cyclic stress response for Zr-2.5Nb tested under FSR and SSR cycling at total strain amplitude of 0.6% between 20 and 300°C in air.

3.2 Fatigue Response in Water

Figure 19 shows the strain-life curve for Zr-2.5Nb tested in LiOH water under SSR cycling at 300°C. The fatigue data obtained in water tests are compared with the data developed in air tests. The fatigue life of Zr-2.5Nb was dramatically reduced when it was tested in LiOH water. When compared with the fatigue data in air under the same type of cyclic loading (SSR cycling), the effect of LiOH water environment on the fatigue life is more pronounced in the low fatigue life regime than in the high fatigue life regime. When compared with the fatigue data in air under FSR cycling, the fatigue life in LiOH water was considerably shorter in both low- and high-fatigue life regimes than the fatigue life in air. The synergetic effect of the aqueous environment and low strain rate is more detrimental to the fatigue resistance of Zr-2.5Nb. A fatigue life reduction factor of ≈ 4 was observed due to the combined effect of LiOH water and low strain rate. One fatigue test was conducted in pure water at strain amplitude of 0.75% at 300°C. No significant difference was observed between the pure water test and the LiOH water test.

Figure 19. Strain- life response for Zr-2.5Nb tested under SSR cycling at 300°C in water. The fatigue data obtained in water tests are compared with the data in air tests.

The strain-life curves for Zr-2.5Nb tested in LiOH water with contributions of the elastic and plastic strain components are illustrated in Fig. 20, together with the strain-life relations in air. The elastic strain component dominates the fatigue response over the examined strain ranges. The elastic strain-life relation in water can be represented by:

$$\frac{\Delta \varepsilon_e}{2} = 0.453(N_f)^{0.034} \quad (6)$$

The plastic strain-life relation in water can be represented by:

$$\frac{\Delta \varepsilon_p}{2} = 6.809(N_f)^{0.625} \quad (7)$$

The influence of the aqueous environment on the elastic strain-life curves is minimal, while the plastic strain-life relations between the water tests and the air tests are dramatically different. The fatigue ductility exponents of the plastic strain-life relations obtained from three types of fatigue tests are similar, in the range of 0.5-0.6, while the fatigue ductility coefficient decreased dramatically in the presence of the low strain rate and the aqueous environment.

Figure 20. Strain- life response in terms of total, elastic and plastic strain amplitudes for Zr-2.5Nb tested under SSR cycling at 300°C in water. The fatigue data obtained in water tests are compared with the data in air tests.

The half-life hysteresis loops at total strain amplitudes of 0.5% and 0.75% for Zr-2.5Nb tested in water and in air at 300°C are shown in Fig. 21. It is evident that the maximum width (the plastic strain range) and the loop area (the plastic strain energy per unit volume) were significantly smaller in the water tests conducted with a slow strain rate in tension.

Figure 22 shows the cyclic stress-strain curves for Zr-2.5Nb tested 300°C under SSR cycling in water, and under FSR and SSR cycling in air. The cyclic stress-strain curves were constructed by using the stress and plastic strain amplitudes of half-life hysteresis loops from fatigue tests at various strain amplitudes. It is found that the

application of a low strain rate in tension led to cyclic hardening, and the combination of water environment and low strain rate in tension resulted in significantly greater cyclic hardening. It is also noted that a linear cyclic stress-strain behavior exists for Zr-2.5Nb when tested in air, while the tests in water showed a power law relationship for the cyclic stress-strain curve.

The cyclic stress responses during fatigue cycling for Zr-2.5Nb tested at 300°C in water and in air are compared in Fig. 23. The cyclic stress behavior in water environment is similar to that in air under SSR cycling.

Figure 21. Half-life hysteresis loops at total strain amplitudes of (a) 0.5% and (b) 0.75% for Zr-2.5Nb tested in water and in air at 300°C.

Figure 22. Cyclic stress-strain curves for Zr-2.5Nb tested at 300°C under SSR cycling in water. The fatigue data obtained in water tests are compared with the data in air tests.

Figure 23. Cyclic stress response for Zr-2.5Nb tested at 300°C in air and in water.

3.3 SEM Examination

3.3.1 Fractography

Fatigue Tests in Air under FSR Cycling

The macroscopic appearance of the fracture surfaces for Zr-2.5Nb fatigue tested under FSR cycling at 300°C in air at total strain amplitudes of 0.5 (specimen ID: A12A), 0.6% (specimen ID: A7A), and 0.75% (specimen ID: A18A), are shown in Fig. 24. All fracture surfaces show typical fatigue features such as surface crack initiation, fatigue zone of crack growth, and fast fracture zone. Fatigue crack initiation occurred at the edge (specimen surface) in all tests. A single site for crack initiation was observed in the lower strain amplitude and longer fatigue life specimen (e.g. A12A), while multiple crack initiation sites and ratchet marks were observed in the higher strain amplitudes and shorter fatigue life specimens (e.g. A18A). Fatigue zones tended to be rougher as strain amplitude increased.

Figure 24. Macroscopic appearance of fracture surfaces of Zr-2.5Nb tested at 300°C in air at a total strain amplitude of (a) 0.5% (ID: A12A), (b) 0.6% (ID: A7A), and (c) 0.75% (ID: A18A).

A series of micrographs were taken (at a magnification of 200-300X) from the crack initiation point at the edge to final fast fracture along the crack propagation direction. An overview of the fracture surface is shown in Fig. 25 for specimen A12A tested at strain amplitude of 0.5%, in Fig. 26 for specimen A7A tested at strain amplitude of 0.6%, and in Fig. 27 for specimen A18A tested at strain amplitude of 0.75%, respectively. The crack propagation direction is indicated by an arrow. Enlarged views at a higher magnification show fatigue fracture mode in the “stage I” early crack propagation region and in the “stage II” faster crack propagation region. The fractographic morphology of fracture surfaces was quite similar for the three specimens with an apparent earlier transition to stage II in the specimens of higher strain amplitudes. The stage II fracture surfaces didn’t show well-defined fatigue striations due to heavy laminated secondary cracking perpendicular to the main crack propagation direction. Laminated secondary cracking is more severe in the high strain amplitude specimens than in the low strain amplitude specimens. Surface oxidation was more severe in the lower strain amplitude specimens that had longer fatigue lives and thus longer exposure to air during fatigue cycling.

Figure 25. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested under FSR cycling at 300°C in air at total strain amplitude of 0.5% (ID: A12A).

Figure 26. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested under FSR cycling at 300°C in air at total strain amplitude of 0.6% (ID: A7A).

Figure 27. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested under FSR cycling at 300°C in air at total strain amplitude of 0.75% (ID: A18A).

Figure 28 compares the macroscopic features of fracture surfaces of the specimen tested at 20°C (A22A) and the specimen tested at 300°C (A7A) at the same strain amplitude (0.6%) under FSR cycling. The major difference is the multiple crack initiation sites shown in the specimen A7A with a shorter fatigue life and a single crack initiation site shown in the specimen A22A with a longer fatigue life. An overview of the fracture surface of specimen A22A is shown in Fig. 29 with enlarged views at a higher magnification showing details of fatigue fracture modes. Compared to the micrographs of specimen A7A shown in Fig. 29, there was no significant difference in fatigue fracture modes. The striations and laminated secondary cracking are better presented in specimen A22A likely due to much less surface oxidation at room temperature.

Figure 28. Comparison of macroscopic appearance of fracture surfaces for Zr-2.5Nb at a total strain amplitude of 0.6% under FSR cycling (a) tested at 20°C (specimen ID: A22A) and (b) tested at 300°C (specimen ID: A7A).

Figure 29. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested under FSR cycling at 20°C in air at total strain amplitude of 0.6% (ID: A22A).

Fatigue Tests in Air under SSR Cycling

The effect of strain rate on macroscopic fracture features of fatigue-failed specimens is shown in Fig. 30(a) for specimen A28A (0.5% SSR cycling) and in Fig. 30(c) for specimen A26A (0.75% SSR cycling) tested at 300°C in comparison with Fig. 30(b) for specimen A12A (0.5% FSR cycling) and Fig. 30(d) for specimen A18A (0.75% FSR cycling). Noticeable differences of surface features between specimens in SSR cycling and specimens in FSR cycling were observed. At total strain amplitude of 0.5%, river marks in the fast-growing fatigue zone were evident in specimen A28A, while at total strain amplitude of 0.75%, heavy ratchet marks and ridges are clearly seen in specimen A26A.

(a) $\Delta\epsilon_t / 2 = 0.5\%$ SSR cycling (ID: A28A) (b) $\Delta\epsilon_t / 2 = 0.5\%$ FSR cycling (ID: A12A) (c) $\Delta\epsilon_t / 2 = 0.75\%$ SSR cycling (ID: A26A) (d) $\Delta\epsilon_t / 2 = 0.75\%$ FSR cycling (ID: A18A)

Figure 30. Macroscopic appearance of fracture surfaces of Zr-2.5Nb tested at SSR and FSR cycling at 300°C in air (a) and (b) at a total strain amplitude of 0.5% and (c) and (d) at a total strain amplitude of 0.75%.

Macroscopic fracture of fatigue-failed specimen (A32A) tested at 0.6%/20°C in air under SSR cycling is shown in Fig. 31. In comparison with specimen A22A tested at 0.6%/20°C under FSR cycling, fracture surface showed more roughness at a lower strain rate than at a higher strain rate.

(a) SSR cycling (ID: A32A) (b) FSR cycling (ID: A22A)

Figure 31. Comparison of macroscopic appearance of fracture surfaces for Zr-2.5Nb tested in air at 20°C at a total strain amplitude of 0.6% (a) under SSR cycling (specimen ID: A32A) and (b) under FSR cycling (specimen ID: A22A).

Figure 32 shows an overview with enlarged views at a higher magnification of the fracture surface of specimen A28A, Fig. 33 for specimen A26A, and Fig. 34 for specimen A32A. Compared with the specimens fatigue-cycled at a faster strain rate, the fracture modes did not change when a slower strain rate in tension was applied. Crack initiation sites showed some corrosion effect.

Figure 32. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested at 300°C in air at a total strain amplitude of 0.5% under SSR cycling (ID: A28A).

Figure 33. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested at 300°C in air at total strain amplitude of 0.75% under SSR cycling (ID: A26A).

Figure 34. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested at total strain amplitude of 0.6% under SSR cycling at 20°C in air (ID: A32A).

Fatigue Tests in Water under SSR Cycling

Macroscopic fracture features of fatigue-failed specimen tested in water under SSR cycling at 300°C are shown in Figs. 35(a) for specimen A7B (0.45%), (b) for specimen A5B (0.5%), (c) for specimen A4B (0.6%), (d) for specimen A9B (0.65%), and (e) for specimen A2B (0.75%). A single crack initiation site was observed in the lower strain amplitude fatigue tests (e.g. 0.45% and 0.5%), while multiple crack initiation sites were observed in the higher strain amplitude fatigue tests. Ratchet marks and rough fatigue zones were observed in the high strain amplitude test specimens, similar to the macroscopic fracture features observed in the air tests under SSR cycling.

Figure 35. Macroscopic appearance of fracture surfaces of Zr-2.5Nb tested under SSR cycling at 300°C in water over a range of total strain amplitude of 0.45-0.75%.

An overview of the fracture surfaces at a magnification of 200X from the crack initiation point(s) at the edge to the final rupture zone along the crack propagation direction is shown in Figs. 36-40 for specimens A7B (0.45%), A5B (0.5%), A4B (0.6%), A9B (0.65%), and A2B (0.75%), respectively. Enlarged views at a higher magnification of the fracture surfaces of specimens are also shown. Compared with the specimens tested in air at the same temperature and cyclic loading mode, there are no significant changes in the fatigue fracture modes by the water environment. Well-developed fatigue striations are seen in the late stage II of fatigue crack propagation in all the specimens tested in water.

Figure 36. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested at total strain amplitude of 0.45% under SSR cycling at 300°C in water (ID: A7B).

Figure 37. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested at total strain amplitude of 0.5% under SSR cycling at 300°C in water (ID: A5B).

Figure 38. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested at total strain amplitude of 0.6% under SSR cycling at 300°C in water (ID: A4B).

Figure 39. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested at total strain amplitude of 0.65% under SSR cycling at 300°C in water (ID: A9B).

Figure 40. SEM micrographs of the fracture surfaces for Zr-2.5Nb tested at total strain amplitude of 0.75% under SSR cycling at 300°C in water (ID: A2B).

3.3.2 Surface Cracking

Fatigue Tests in Air under FSR Cycling

Representative SEM micrographs showing surface secondary cracking along the specimen gage length are shown in Fig. 41 for specimen A18A (strain amplitude of 0.75%) and for specimen A12A (strain amplitude of 0.5%) tested at 300°C in air under FSR cycling. Higher strain amplitudes resulted in severe surface cracking (A18A) with cracks oriented in the direction normal to the applied loading direction, while at lower strain amplitudes, fatigue slip bands were evident (A12A).

(a) Specimen A18A (strain amplitude of 0.75%).

(b) Specimen A12A (strain amplitude of 0.5%).

Figure 41. Representative SEM micrographs showing longitudinal surface features in the gage section for Zr-2.5Nb tested at 300°C in air under FSR cyclic loading.

Figure 42 compares the surface features of the specimens tested at two different temperatures, 20 and 300°C, at total strain amplitude of 0.6%. Some surface cracking was observed in specimen A7A tested at 300°C, while the gage surface was free of cracking in specimen A22A tested at 20°C.

SEM micrograph of the gage surfaces of specimen A7A (tested at 300°C).

SEM micrograph of the gage surfaces of specimen A22A (tested at 20°C).

Figure 42. Representative SEM micrographs showing surface features in the gage section for Zr-2.5Nb tested at 20 and 300°C in air at strain amplitude of 0.6% under FSR cyclic loading.

Fatigue Tests in Air under SSR Cycling

The effect of strain rate on the surface secondary cracking behavior is shown in Fig. 43 for specimens tested at strain amplitude of 0.5% and 300°C and in Fig. 44 for specimens tested at strain amplitude of 0.75% and 300°C, respectively. For specimens tested at the lower strain amplitude (0.5%), no significant cracking was observed. The effect of strain rate is manifested by fatigue slip bands observed in specimen A12 tested under FSR cycling, whereas the specimen A28A tested under SSR cycling showed a clean surface. When strain amplitude increased to 0.75%, significant surface cracking was observed in both FSR and SSR cycling modes, and much more severe surface cracking occurred under SSR cyclic loading.

The effect of strain rate on the surface secondary cracking behavior is shown in Fig. 45 for specimens tested at strain amplitude of 0.6% and 20°C. The gage surfaces are nearly free of surface cracking.

SEM micrograph of the gage surfaces of specimen A12A (FSR cycling).

SEM micrograph of the gage surfaces of specimen A28A (SSR cycling).

Figure 43. Representative SEM micrographs showing surface features in the gage section for Zr-2.5Nb tested at 300°C in air at strain amplitude of 0.5% under FSR and SSR cycling.

SEM micrograph of the gage surfaces of specimen A18A (FSR cycling).

SEM micrograph of the gage surfaces of specimen A28A (SSR cycling).

Figure 44. Representative SEM micrographs showing surface features in the gage section for Zr-2.5Nb tested at 300°C in air at strain amplitude of 0.75% under FSR and SSR cycling.

SEM micrograph of the gage surfaces of specimen A32A (0.6% SSR cycling at 20°C).

SEM micrograph of the gage surfaces of specimen A22A (0.6% FSR at 20°C).

Figure 45. Representative SEM micrographs showing surface features in the gage section for Zr-2.5Nb tested at 20°C in air at strain amplitude of 0.6% under FSR and SSR cycling.

Fatigue Tests in Water under SSR Cycling

Representative SEM micrographs showing surface secondary cracking along the specimen gage length are shown in Figs. 46-50 for the specimens tested in water at 300°C under SSR cycling at strain amplitude of 0.45-75%. Surface cracking was observed in all the specimens. At the lowest strain amplitude (0.45%), surface cracks are oriented toward the maximum shear direction and concentrated near the fracture surface. As the strain amplitude increased, surface cracks are aligned to the direction normal to the applied uniaxial loading direction, and cracks prevail over the gage section. Compared with the specimens tested in air under SSR cycling, secondary surface cracking is much more severe in the water-tested specimens.

Figure 46. Representative SEM micrograph showing surface features in the gage section for Zr-2.5Nb tested in water at 300°C under SSR cycling at strain amplitude of 0.45%.

Figure 47. Representative SEM micrograph showing surface features in the gage section for Zr-2.5Nb tested in water at 300°C under SSR cycling at strain amplitude of 0.5%.

Figure 48. Representative SEM micrograph showing surface features in the gage section for Zr-2.5Nb tested in water at 300°C under SSR cycling at strain amplitude of 0.6%.

Figure 49. Representative SEM micrograph showing surface features in the gage section for Zr-2.5Nb tested in water at 300°C under SSR cycling at strain amplitude of 0.65%.

Figure 50. Representative SEM micrograph showing surface features in the gage section for Zr-2.5Nb tested in water at 300°C under SSR cycling at strain amplitude of 0.75%.

4. Discussion

Cyclic stress response and cyclic stress-strain behavior

The cyclic stress response of Zr-2.5Nb during fatigue cycling often showed three stages: transient cyclic softening, cyclic saturation, and secondary cyclic hardening. Each stage is affected by strain amplitude, test temperature, and strain rate. Cyclic softening occurred during the initial phase of cyclic deformation in all the fatigue tests. This transient cyclic softening is expected in Zr-2.5Nb pressure tube material where pre-deformed microstructure experienced dynamic recovery as a consequence of dislocation annihilation and rearrangement.

In many cases, the initial cyclic softening was followed by cyclic saturation where the stress amplitude remained constant and the material achieved a cyclically stable condition.

Only in very high strain amplitude and short fatigue life tests, cyclic softening was persistent until the final load drop, due to early fatigue cracking. An interesting feature was observed in the cyclic stress response at relatively low strain amplitudes and long fatigue lives that progressive cyclic hardening occurred after cyclic saturation. Secondary cyclic hardening has been observed in several Zr-based alloys including Zr-Sn-Nb alloy [Tan et al 2006] and Zircalloys [Armas et al 1992, Moscato et al 1997, Armas et al 1996]. The occurrence of secondary cyclic hardening in zirconium alloys was suggested to be associated with the development of secondary dislocations due to the activation of the dynamic strain aging [Alvarez-Armas and Herenu 2004].

Dynamic strain aging is the phenomenon of interaction between diffusing solute atoms and mobile dislocations during plastic deformation. It depends on deformation rate and temperature, which govern the velocities of mobile dislocations and diffusing solute atoms, respectively. The DSA regime for Zircalloys has been reported to be 300-500°C for both monotonic and cyclic loading [Derep et al 1980, Ahn and Nam 1990, Moscato et al 1997, Alvarez-Armas and Herenu 2004]. The DSA regime for Zr-2.5Nb under monotonic tensile loading was found to be in the temperature range of 250-350°C [Singh et al 2005]. No work has been done on the occurrence of DSA in Zr-2.5Nb during fatigue deformation. Since the test temperature of 300°C used in this study falls in the DSA regime for monotonic tensile loading, it is necessary to examine the DSA under cyclic loading and its influence on fatigue performance of Zr-2.5Nb.

The cyclic stress response of Zr-2.5Nb showed a higher stress at a given total strain amplitude at the lower strain rate than that at the higher strain rate at 300°C, as shown in Fig. 17. Some type of cyclic hardening mechanism must be activated to offset the cyclic softening effect when the strain rate was reduced. This inverse stress-strain rate dependence is one of the typical features of the DSA process [Dieter 1976]. The cyclic stress-strain behavior shown in Fig. 22 also exhibits a strain rate effect. For both fast and slow strain-rate fatigue tests in air, a linear strain hardening was observed in cyclic stress-strain curves. The linear strain hardening rate is strain-rate-dependent: the lower the strain rate, the higher the strain hardening rate. This is another indication of dynamic strain aging [Dieter 1976]. A similar strain rate effect on cyclic hardening rate was observed in Zircaloy-4 [Moscato et al 1997], while the temperature range at which the phenomenon was observed was different (300-500°C for Zircalloys). The data developed in water seems to indicate significant increase in DSA when compared to the data in air, under identical cyclic conditions. It is apparent that the DSA occurred during fatigue deformation in Zr-2.5Nb at 300°C. The activation of this atomic process can provoke a drastic increase in dislocation density, leading to additional cyclic hardening. The secondary cyclic hardening and the higher stresses at a lower strain rate during fatigue cycling may be explained by the DSA effect. Since the DSA is a thermally-activated process, cyclic hardening behavior in Zr-2.5Nb is dependent on test temperature and strain rate.

No strong temperature dependence on the cyclic behavior could be observed in the temperature range of 20-300°C at the total strain amplitude of 0.6% in Zr-2.5Nb. There is an indication that the difference in the cyclic stress response at different temperatures resided in the secondary cyclic hardening at the late stage of fatigue cycling. Since the existence of secondary cyclic hardening depends on its occurrence before or after fatigue failure and the 300°C fatigue data showed evidence of secondary hardening only at lower strain amplitudes and longer fatigue lives, it is necessary to develop low-strain amplitude

fatigue data at lower temperatures to determine the temperature effect on the cyclic behavior of Zr-2.5Nb.

Limited fatigue data, developed in this program, showed that the effect of strain rate on the cyclic stress response is temperature-dependent in Zr-2.5Nb (shown in Fig. 18). The stress amplitude in the first few cycles decreased by applying a slow strain rate in tension at 20 and 100°C, while the initial stress amplitude was not changed at 300°C. As cycling progressed, the cyclic stress profile was relatively unchanged in fatigue tests at 20°C at two different strain rates, while the strain-rate-induced difference in cyclic softening diminished when the number of cycles achieved $\approx 60\%$ of the fatigue life at 100°C. The extent of cyclic softening was less at a lower strain rate even at the beginning of cycling at 300°C. The decrease of the initial stress amplitude at a lower strain rate at 20 and 100°C is expected, while similar values of the initial stress amplitudes at two different strain rates at 300°C are possibly due to activation of the DSA process. The diminishing cyclic softening difference at the late stage of fatigue cycling at 100°C implies that the DSA could occur during fatigue deformation even at 100°C. Tsuzaki et al [Tsuzaki et al 1991] concluded that the DSA during fatigue deformation can occur at lower temperatures in comparison with that during tensile deformation.

Effect of temperature on fatigue life

The fatigue life of Zr-2.5Nb was significantly improved as temperature decreased from 300°C to 20°C. The strain-life curves (Fig. 13) showed that the elastic strain component was considerably higher at lower temperature at a given total strain amplitude. The elastic strain component dominated a greater portion of the fatigue life at lower temperatures than at higher temperatures. Since the elastic strain component is related to material strength and the plastic strain component is related to material ductility, the effect of temperature on fatigue life may be explained by the temperature-dependent material strength and ductility. Table 3 summarizes the tensile properties of the Zr-2.5Nb specimens at 31°C and 299°C [Radford 2007].

Table 3. Tensile properties of Zr-2.5Nb.

Temperature (°C)	Specimen Orientation	0.2% offset yield stress (MPa)	Ultimate tensile strength (MPa)	Uniform elongation (%)	Total elongation (%)
31	Longitudinal	594	794	4.96	16.33
299	Longitudinal	415	561	3.77	16.86
31	Transverse	869	879	0.45	20.69
299	Transverse	596	635	1.26	17.44

It is noted that the yield stress and ultimate tensile strength decreased considerably as temperature increased from 31°C to 299°C, while the difference in tensile ductility at two temperatures is insignificant. The better fatigue resistance of Zr-2.5Nb at room temperature may be attributed to its high strength at room temperature.

Effect of strain rate on fatigue life

Strong strain rate dependence of fatigue life was observed at 300°C in the air tests. The fatigue life was significantly reduced when a low strain rate was applied in tension during cycling. The effect of strain rate was more pronounced at lower strain amplitudes and longer fatigue lives. Limited fatigue data showed an insignificant effect of strain rate on fatigue life at 20 and 100°C.

As shown in Fig. 22, the cyclic stress-strain curves show a linear strain hardening behavior under both FSR and SSR cycling in the fatigue tests in air. The linear strain hardening implies that planar slip is a dominant deformation mode during fatigue cycling in Zr-2.5Nb [Alvarez-Armas and Herenu 2004, Caceres and Blake 2007]. When DSA operates, planar slip is promoted. It is enhanced as the strain rate decreases [Dieter 1976]. Planar slip can lead to strong strain incompatibility among grains due to a limited number of slip systems, resulting in high intergranular stresses. The DSA also enhances the degree of inhomogeneity of plastic deformation, resulting in localized deformation within slip bands. Both mechanisms can lead to premature fatigue failure, which could explain why the fatigue life was shorter when a lower strain rate was applied at 300°C. The fact that the effect of strain rate on fatigue life was more pronounced at lower strain amplitudes than at high strain amplitudes implies that the strain rate effect was mostly in fatigue crack initiation.

Fatigue behavior in simulated water environments

The fatigue life of Zr-2.5Nb was significantly reduced when tested in simulated water environment at 300°C. When tested under the same type of waveform (SSR cycling), the influence of water environment on the fatigue life is more pronounced in the high strain amplitude and low fatigue life regime than in the low strain amplitude and high fatigue life regime. The combination of the water environment and the low strain rate in tension greatly reduced the fatigue life in the entire range of strain amplitudes examined. It is apparent that the synergistic effect of the aqueous environment and the strain rate is most detrimental to the fatigue resistance of Zr-2.5Nb. The deleterious effect of the aqueous environment on the fatigue resistance may be better understood by the coupling effects between corrosion and cyclic plasticity on fatigue crack initiation and propagation in Zr-2.5Nb.

The aqueous environment has little influence on the cyclic stress response and fatigue fracture modes, and however, significantly affects the plastic strain-life relation and the cyclic stress-strain behavior of Zr-2.5Nb. The fatigue life was remarkably shorter at a given plastic strain amplitude when tested in water than tested in air. The strain rate effect on the fatigue response, including both the plastic strain amplitude response and the cyclic strain hardening behavior was exaggerated in the aqueous environment. While the DSA process was suggested to be responsible for the additional cyclic hardening at a lower strain rate in the air fatigue tests, it is likely that the DSA effect is more pronounced in the aqueous environment, leading to much greater cyclic strain hardening under SSR cycling in Zr-2.5Nb.

Comparison with literature fatigue data

Fatigue data in the air tests are compared with the data reported by Hosbons and Wotton [Hosbons and Wotton 1996] in Fig. 51 for Zr-2.5Nb in the longitudinal direction tested at 300°C in air. Note that fatigue tests in this study were carried out in strain-controlled conditions, while the fatigue tests in reference [Hosbons and Wotton 1996] were conducted in load-controlled conditions. Figure 51(a) compares the strain-life response by determining elastic-strain amplitudes using Eq. (1) for the data in reference [Hosbons and Wotton 1996], and Fig. 51(b) compares the stress-life relation with the stress amplitudes in this study taken at the half fatigue life. It is seen that the data in reference [Hosbons and Wotton 1996] showed a large data scatter. The data in the strain-controlled tests in this

Figure 51. Comparison of strain-controlled fatigue data in this study and stress-controlled fatigue data reported by Hosbons and Wotton for Zr-2.5Nb tested at 300°C in air (a) strain-life response and (b) stress-life response.

study are in the low-cycle fatigue regime, and the fatigue data in the load-controlled tests in reference [Hosbons and Wotton 1996] are mostly in the high-cycle regime. In the intermediate fatigue cycle regime where the two experiments overlap, a better consistence exists in the relatively low cycle fatigue region, while a significant discrepancy occurs in the relatively high cycle fatigue region. This is somewhat surprising, as at long fatigue lives where load levels are low and the plastic strain is negligible, load-controlled and strain-controlled test results should be equivalent.

5. Conclusions

- 1) The fatigue life of Zr-2.5Nb was significantly reduced in simulated CANDU-reactor water environments at 300°C. The synergetic effect of the aqueous environment and the strain rate is most detrimental to the fatigue resistance of Zr-2.5Nb. The fatigue life of Zr-2.5Nb in water was reduced in both high cycle and low cycle regimes when compared with the fatigue data in air under the fast-fast cyclic loading (FSR cycling). The influence of aqueous environment on the fatigue resistance is more pronounced in the high strain amplitudes and low cycle regime than in the low strain amplitudes and high cycle regime when compared with the fatigue data in air with the slow-fast cyclic waveform (SSR cycling).
- 2) Significant reduction in fatigue life was observed at 300°C in air when a lower strain rate was applied in tension during cycling. The strain rate effect was more pronounced in the low strain amplitude and long fatigue life regime.
- 3) Fatigue performance of Zr-2.5Nb was strongly affected by test temperature. Fatigue life increased significantly as temperature decreased along with considerably higher elastic strain component and lower plastic strain component at lower temperatures. It is apparent that the fatigue performance of Zr-2.5Nb at low temperatures is dominated by the material strength.
- 4) The cyclic stress response during cycling at 300°C can be characterized by initial cyclic softening, cyclic saturation, and secondary cyclic hardening. Secondary cyclic hardening was more pronounced at lower strain amplitudes.
- 5) The cyclic stress response was insensitive to the applied strain amplitudes and test temperature, while it was strongly affected by the strain rate during cycling. The extent of cyclic softening in the cyclic stress response was less when a slow strain rate was applied in tension at 300°C; the cyclic stress-strain behavior showed an inverse strain-rate dependence of cyclic strain hardening rate. While the aqueous environment has little influence on the cyclic stress response as a function of cycles, the cyclic strain hardening at the half-fatigue life was much more pronounced under the coupling effect of the low strain rate and the aqueous environment.
- 6) All fatigue fracture surfaces showed typical surface crack initiation, crack propagation fatigue zone and fast fracture zone. A single crack initiation site was often observed in specimens tested at lower strain amplitudes and lower temperatures, while multiple crack initiation sites were observed in specimens tested at higher strain amplitudes and higher temperatures. Fatigue zones were covered with striation-like features and heavy laminated secondary cracking in all the tests. Early onset of stage II crack propagation was evident in specimens with shorter fatigue lives. Secondary surface cracking along the gage was more pronounced at higher strain amplitudes and higher temperature and at slow strain rates. The aqueous environment has little influence on the fatigue fracture modes of Zr-2.5Nb, while it was observed that surface cracking along the gage was more severe in the water tests than in the air tests.

6. Recommended Future Work

- It is apparent that the synergistic effect of aqueous environment and the strain rate is most detrimental to the fatigue resistance of Zr-2.5Nb. Fatigue tests in simulated water environment under FSR cycling are needed to develop a better understanding of the fatigue resistance of Zr-2.5Nb in water.
- The interaction between corrosion and cyclic plasticity assisted by the DSA may be responsible for accelerated fatigue crack initiation and propagation in Zr-2.5Nb. Fatigue tests in water at a lower temperature where the DSA effect is suppressed are important to understand the possible role of the DSA process and its coupling effect with corrosion in the fatigue response of Zr-2.5Nb.
- Fatigue crack growth rates in simulated water environment are crucial information for the structural integrity of Zr-2.5Nb pressure tubes. Fatigue crack growth experiments in simulated water environments are highly recommended.
- The strong strain-rate dependence of the fatigue life needs further investigation by applying a slow strain rate in compression, and a slow strain rate in both tension and compression. This set of experiments will allow determining the role of the DSA in fatigue initiation and propagation in Zr-2.5Nb. The effect of hold time on fatigue life at 300°C should also be considered.
- Fatigue tests at a wide range of temperatures and strain amplitudes are needed to fully understand the role of the DSA process in the fatigue performance and to determine the DSA regime during fatigue deformation in Zr-2.5Nb.
- Detailed microstructure characterization of fatigue-deformed specimens by transmission electron microscopy (TEM) is critical to understand the deformation modes (wavy slip vs. planar slip) leading to fatigue failure. Dislocation evolution during fatigue cycling and crack initiation and propagation mechanisms should be examined by performing interrupted fatigue tests.
- Fatigue studies of Zr-2.5Nb are complicated by strong texture developed during thermo-mechanical processes. The anisotropic properties of the hcp crystal structure and strong crystallographic texture in Zr-2.5Nb not only give anisotropic mechanical properties, but can also lead to shorter fatigue life due to limited plastic deformation ability. The tensile properties of Zr-2.5Nb showed significantly higher tensile strengths in the transverse direction than in the longitudinal direction, but very little strain hardening and ductility. It is critical to obtain fatigue data in the transverse direction to determine the effect of texture.
- There is a gradual change in grain structure and crystallographic texture from one end of the pressure tube to the other. Pressure tubes are stronger at the back ends than at the front ends of the tubes due to the finer grain size at the back ends. Fatigue specimens taken from various locations of a pressure tube should be tested to evaluate the variation in fatigue properties along the pressure tube.

- Fatigue data of Zr-2.5Nb with surface flaws are needed to assess the fatigue resistance in the pressure tubes with flaws produced during manufacturing and/or operation.
- Previous work showed that an oxide layer may reduce the fatigue limit of Zr-2.5Nb by up to a factor of two. Fatigue tests of Zr-2.5Nb pressure tube material with the oxide surface layer are suggested.
- Zr-2.5Nb pressure tubes are prone to hydrogen pickup during service. It needs to be determined whether the hydrides that are present in the tube during operation affect the fatigue properties of the Zr-2.5Nb pressure tubes.
- A fatigue life prediction model for Zr-2.5Nb is needed to account for the effects of aqueous environment, strain rate, temperature, texture, oxidation, and hydride on fatigue performance.

References

- J. S. Ahn and S.W. Nam, *Mater. Lett.* 9 (1990) 413.
- I. Alvarez-Armas and S. Herenu, *J. Nucl. Mater.* 334 (2004) 180.
- A.F. Armas, I. Alvarez-Armas, and G. Mansilla, *J. Mater. Sci.* 27 (1992) 1307.
- A.F. Armas, I. Alvarez, and G. Moscato, *Scripta Metall.* 34 (1996) 281.
- G.A. Bickel and M. Griffiths, *J. Nucl. Mater.* 383 (2008) 9.
- C.H. Caceres and A.H. Blake, *Mater. Sci. Eng. A* 462 (2007) 193.
- C.E. Coleman, R.W. Gilbert, G.J.C. Carpenter, and G.C. Weatherly, in: *Phase Stability during Irradiation*, the Metallurgical Society of AIME, Pittsburg, 1980, p. 857.
- J.L. Derop, S. Ibrahim, R. Rouby, and G. Fantozzi, *Acta. Mater.* 28 (1980) 607.
- G.E. Dieter, *Mechanical Metallurgy*, 2nd Ed., McGraw-Hill Book Company, 1976.
- R.A. Holt, *J. Nucl. Mater.* 372 (2008) 182.
- R.A. Holt, N. Christodoulou, and A.R. Causey, *J. Nucl. Mater.*, 317 (2003) 256.
- R.A. Holt, M. Griffiths, and R.W. Gilbert, *J. Nucl. Mater.* 149 (1987) 51.
- R.R. Hosbons and B.L. Wotton, "Fatigue Design Curves for Zr-2.5Nb Pressure Tubes," COG-93-318-I (Rev. 1), AECL EACL, March 1996.
- M.G. Moscato, M. Avalos, I. Alvarez-Arma, C. Petersen, and A.F. Armas, *Mater. Sci. Eng. A* 234-236 (1997) 834.
- D.D. Radford, "Results based on MTS load and extensometer data from tube #H850 longitudinal and transverse tensile tests," AECL Memo, June 2007.
- D.A. Scarth, D.H.B. Mok, and G.W. Newman, "Assessment of bearing pad fretting flaws in Candu Zr-Nb pressure tubes," *Service Experience and Design in Pressure Vessels and Piping (Including High Pressure Technology)*, the 1996 ASME Pressure Vessels and Piping Conference, Montreal, Quebec, Canada, The American Society of Mechanical Engineers, PVP, v. 335, July 21-26, 1995, p. 75.
- R.N. Singh, S. Mukherjee, R. Kishore, and B.P. Kashyap, *J. Nucl. Mater.* 345 (2005) 146.
- J. Tan, S. Ying, C. Li, and C. Sun, *Script. Mater.* 55 (2006) 513.
- K. Tsuzaki, Y. Matsuzaki, T. Maki, and I. Tamura, *Mater. Sci. Eng. A* 142 (1991) 63.

Nuclear Engineering Division

Argonne National Laboratory
9700 South Cass Avenue, Bldg. #212
Argonne, IL 60439

www.anl.gov

Argonne National Laboratory is a U.S. Department of Energy
laboratory managed by UChicago Argonne, LLC