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Facility for stress corrosion cracking of irradiated specimens in supercritical water

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Abstract

This paper presents the capability to conduct stress corrosion cracking experiments on neutron-irradiated material in supercritical water for application to the GenIV supercritical water reactor concept. The Irradiated Materials Testing Laboratory (IMTL) is dedicated to performing stress corrosion cracking experiments of neutron-irradiated specimens in supercritical water in both content extension rate tensile mode and crack growth rate mode and to conduct analysis of the morphology and composition of the fracture surface via scanning electron microscopy with energy dispersive X-ray analysis.

1. Introduction

A comprehensive study of the feasibility of the supercritical water reactor concept must include the assessment of the effect of irradiation on stress corrosion cracking in supercritical water. Techniques such as proton irradiation are appealing surrogates for neutron-irradiation in assessing its effect of stress corrosion cracking (SCC) initiation, and can be used for screening of various material and environmental conditions. However, neutron-irradiation is required to confirm the role of in-core irradiation on crack growth and in performing final verification of the effect of alternative irradiation on candidate alloys. The GenIV supercritical water reactor (SCWR) concept requires extensive materials testing in a supercritical water (SCW) environment in order to assess the susceptibility of materials to corrosion and stress corrosion cracking in supercritical water at temperatures as high as 600 °C. The effect of irradiation on corrosion and stress corrosion in SCW is completely unknown, and no facilities exist to test irradiated materials under these conditions. With over 40 years of experience in handling radioactive materials at the Phoenix Memorial Laboratory (PML), and extensive experience in stress corrosion cracking capability and testing in supercritical water environment in the High Temperature Corrosion Laboratory, the University of Michigan undertook to develop the capability to conduct stress corrosion cracking experiments on neutron irradiated materials. This paper describes the capabilities of the facility.

2. Description and capabilities

The Irradiated Material Testing Laboratory was designed to provide constant extension rate tensile

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(CERT) experiment and crack growth rate (CGR) capabilities of neutron-irradiated specimens in supercritical water, and for specimen analysis by scanning electron microscopy (SEM). The facility was designed to minimize the occupation time of the hot cell by making both the load frame and the SEM column mobile. Hot cell #1 in PML is adjacent to the Irradiated Material Testing Laboratory (IMTL) and is used for specimen loading into the autoclave, autoclave closure and pressure testing in preparation for the experiment, and application of shielding. The autoclave is then rolled into IMTL, a 1000 sq ft laboratory located next to the hot cell, where the CERT or CGR experiment is conducted. Once the experiment is completed, the autoclave is rolled back in the hot cell for specimen unloading. Then, the SEM is installed in the hot cell for post-test analysis of fracture and gage sections. The schematic in Fig. 1 shows the two-laboratory complex along with the general procedure used to perform experiments.

2.1. Description of the SCC facility

The SCC facility provides the capability to perform stress corrosion cracking experiments in pure supercritical water, up to 30 MPa of pressure and 600 °C, in a controlled, refreshed environment. The environmental control consists of dissolved oxygen control and monitoring and conductivity monitoring. Oxygen levels below 10 ppb and inlet conductivity of 0.07 μS/cm are obtainable. The make-up and control on the environment is performed in the *water loop*, described in detail below. Constant strain rate, constant load and constant K experiments can be conducted, in addition to fatigue pre-cracking and programmed loading sequences. Those loading modes are applied and controlled by the loading unit described later. The crack propagation measurement technique will also be detailed in this section.

2.1.1. Hot cell description

The hot cell facility is provided by PML is the cave No. 1 presented in Fig. 2. The cell consists in a 3 m wide by 1.8–2.5 m deep room. Shielding is provided by 0.9 m of high density concrete and two 35 cm thick iron doors at the rear. The cell contains three leaded glass, viewing windows with a viewing area of 78 cm (tall) by 90 cm (wide). It is negatively pressurized through a HEPA or

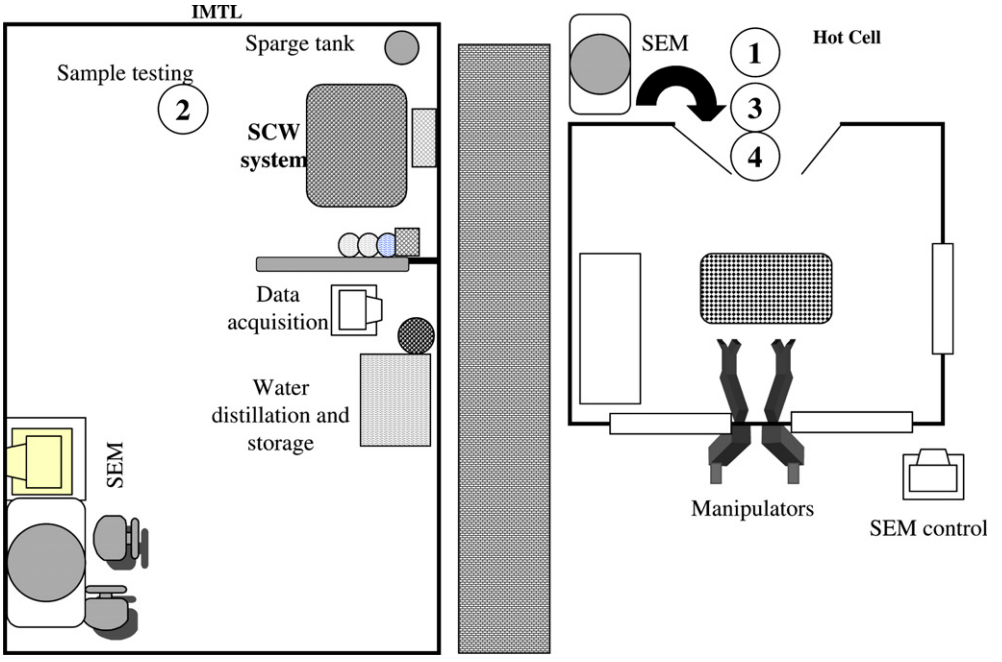

Fig. 1. Schematic of the irradiated materials testing facility, consisting of the Irradiated Materials Testing Laboratory and hot cell #1, and the arrangement of instrumentation for conducting CERT and CGR tests and post-test fractographic analysis on neutron-irradiated materials via SEM. The specimens are loaded on the test facility in the hot cell (1), then the facility is shielded and is moved to IMTL, where the test is performed (2). After completion of the test, the specimens are unloaded in the hot cell (3), and a SEM is rolled in the hot cell for specimen analysis (4).

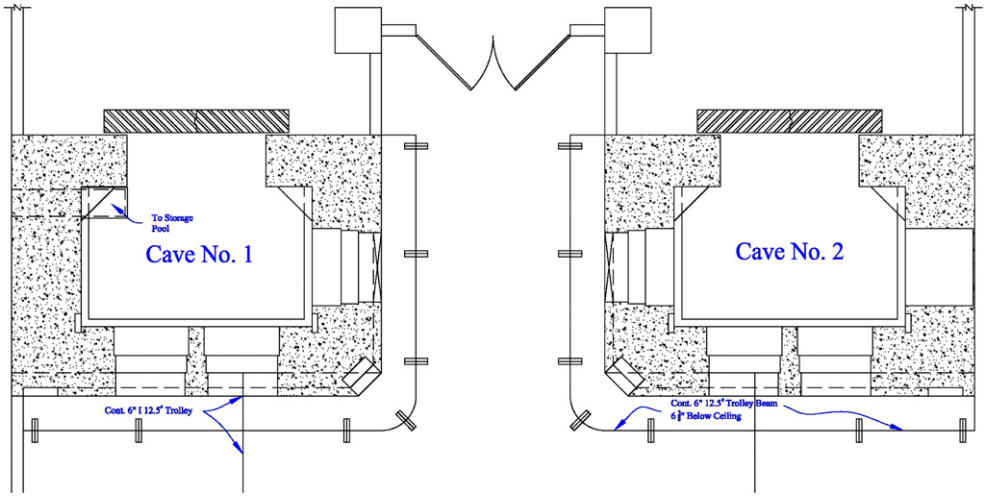

Fig. 2. Layout of the hot cells within the Phoenix Memorial Laboratory. Cave No. 1 is used for loading and unloading irradiated specimens for testing done at the Irradiated Material Testing Laboratory, and for observation of irradiated specimens with SEM.

charcoal (operator selectable) exhaust that can be truncated to provide localized collection of fumes and materials.

The hot cell has two central research model 8 manipulator arms that can transverse left and right across the operating face providing full access to most of the hot cell volume. It contains two half-ton hoists. One is installed on a trolley running along the operating face at the centerline of the hot cell. The other one is located between the manipulator arms.

2.1.2. Water loop

The testing facility consists of a closed-loop, flowing water system. The water chemistry is first prepared and controlled using a *water board*, where distilled water is stored in two glass columns. Gas is bubbled through the columns and a small positive gas pressure can be applied in order to control the dissolved gas content of the water. Both columns are connected to a recirculation loop that directs the water through an ion exchanger to maintain purity. As the primary column contains the water to be used for the experiment, a conductivity meter and an oxygen meter are installed in the recirculation loop to permit continuous monitoring of the conductivity and dissolved oxygen content of the water at room temperature and atmospheric pressure.

When the desired conditions are reached, the water is pressurized and heated up to a temperature close to the target temperature before it flows into the autoclave. A high-pressure liquid chromatographic pump pushes the water from the primary column to the autoclave through the preheater and controls the flow rate for the experiment up to 200 ml/min. The pressurized water will pass through the thermodynamic critical point of water as it is heated to a value that is close to the testing temperature. This is achieved by flowing the water through a regenerative heat exchanger that takes its heat from the water leaving the autoclave. The water then flows through a coil heated by four heating cords controlled by two temperature controllers. The preheater (heat exchanger plus heated coil) is enclosed in a $40 \times 23 \times 36$ cm ceramic insulating box. The water temperature after the heat exchanger and at the exit of the preheater (before the water flows in the autoclave) is continuously recorded.

Once inside the autoclave, the water is maintained at the desired temperature by a set of heating bands clamped to the autoclave body, and an insulating jacket. The temperature is measured by a thermocouple located inside the autoclave.

After flowing through the autoclave the water is cooled down to room temperature before it returns to the water board. The temperature decrease is achieved using a regenerative heat exchanger described previously, followed by a non-regenerative heat exchanger. The water finally goes through a 2 μm filter and a back pressure regulator (BPR), where the pressure is reduced. The section of the loop between the pump and the BPR is at the system pressure, controlled by the BPR, and measured by a pressure gage and two pressure transducers

(located both at the inlet line and the outlet line). After the BPR, the water is at atmospheric pressure and flows through a conductivity meter and an oxygen meter before returning to the primary column.

All water system parameters such as temperature, pressure, dissolved oxygen content, conductivity are continuously monitored and recorded. The temperature inside the autoclave is controlled with a variation ±0.2 °C and the pressure fluctuation are below ±0.07 MPa. A schematic of the system is provided in Fig. 3 and the assembled system is shown in Fig. 4.

2.1.3. Load frame and motor assembly

The autoclave is supported by a mobile load frame that also contains the servo motor and the

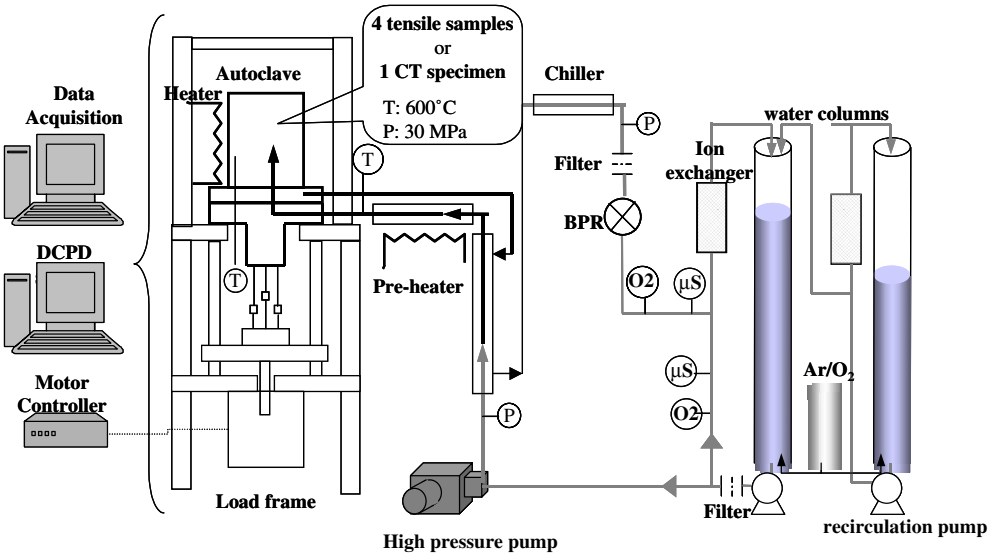

Fig. 3. Schematic of the stress corrosion cracking facility in IMTL.

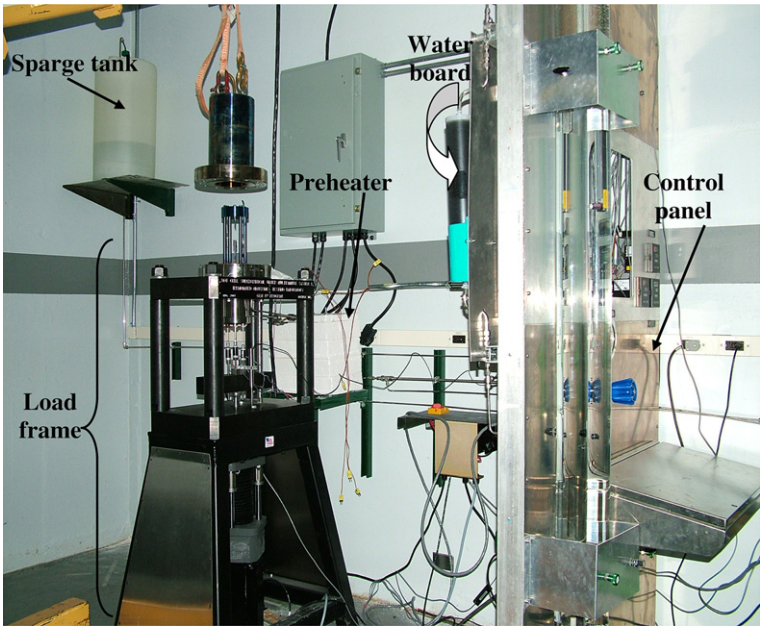

Fig. 4. Photo of the SCC facility installed in IMTL. In this configuration, the loading unit is equipped with an autoclave head designed for CERT experiments.

pull rod system. This assembly is described here as the load frame and motor assembly.

The autoclave is modular, in that it is designed to support separate heads for CERT and CGR tests. The CERT head accommodates four specimens for testing in parallel, whereas the CGR head is designed for one compact tension (CT) specimen. Each head contains a thermocouple to measure the temperature of the water next to the specimen. In addition, openings are provided on the CGR head for the leads for direct current potential drop (DCPD) measurement, and two extra openings were added to each head for future electrochemical measurements.

The load controller is a motor that can be operated in constant strain, constant load, constant K and cyclic modes. The motor is capable of cyclic loading ($\geqslant 1$ Hz) required to fatigue pre-crack specimens for CGR experiments. It is permanently fixed to the load frame beneath the autoclave. The motor and pull rod(s) are connected via load cells that permit the measure of the load applied to each specimen. For CERT tests, a displacement gage is installed to measure the specimens extension rate. For CGR tests, the motor is controlled by the DCPD system. Parameters such as applied strain and load are continuously monitored and recorded for each specimen.

The autoclave and the motor are installed on a sturdy load frame that could also carry the load of a shield when radioactive specimens are used. The load frame is movable and can be disconnected from the water loop to be moved to the hot cell.

2.1.4. The direct current potential drop system for measuring crack extension

The DCPD system for measuring crack extension plays a key role in CGR experiments. In this technique, a current is passed through the specimen, measurement of the probe potential are taken from different parts of the specimen, the direction of the current is reversed and the probe potential is measured again. The average of the two absolute values recorded constitute a reading. By repeating this sequence a number of times, and averaging the readings to form a single data point, the crack growth can be estimated with high precision [1].

The CT specimen is instrumented with platinum current and potential probe leads that are believed to be necessary to ensure secure connections to the specimen in supercritical water. The Pt current and potential probe leads are connected to the CT specimen by spot welding. The DC source supplies a stable current to the CT specimen, which is reversed periodically through solid-state relays in order to correct for thermocouple effects. The potential drop resulting from crack extension in the specimen is measured by a nanovolt meter. A dedicated software program run on a personal computer controls the nanovolt meter, the DC source and the relays through IEEE-488 interfaces and a parallel port, for the purpose of achieving data acquisition, current reversal, crack extension calculation and load control for constant stress intensity factor (K) CGR experiments. The computer also supplies 0–5 volt signals needed to control the relays.

2.1.5. Conduction of SCC experiments

SCC experiments are conducted in either of two modes; constant extension rate tensile (CERT) or crack growth rate (CGR). The CERT tests are conducted using an autoclave head with four penetrations to load all four specimens simultaneously and independently. Each pull rod is connected to a load cell so independent stress–strain curves are obtained for each specimen. Specimens are strained under a constant extension rate until all specimens fail.

A separate head with a feed through for a single, 6 mm diameter pull rod is used for crack growth rate tests. In these tests, the specimen is connected to the DCPD system to record the crack length, which is used to control the K. For a CGR experiment, the crack is initiated and extended in air by fatigue. The specimen is then immersed in the water environment and once the environment has stabilized, crack growth is reactivated by successive loading steps in order to transform the cracking mode from transgranular to intergranular before switching to fully constant K loading. Following the completion of the crack growth rate test, the specimen is broken by fatigue in air at room temperature.

2.2. Safety system

The facility has several safety features included in this design. The hazards to be considered are high-pressure, high temperature, presence of active material and potential contamination of the facility.

The pressure is controlled in three ways. First, the pump has a limiting pressure above which the pump will not operate. A relief valve is installed after the pump that limits the pressure to a preset value by depressurizing the water and returning it to ambient pressure. Finally, a rupture disk is installed in the

outlet line, after the preheater-cooling unit and just before the water enters back in the water board. If the preset limit is exceeded, the disk ruptures and the pressurized water is drained into a sparge tank. The use of a sparge tank in the laboratory was required as the water could be contaminated.

Each temperature controller is equipped with an alarm that would turn-off the power, if the temperature were to exceed a preset value.

The system was designed so that the operator has access to each control of the system without having to be in the immediate proximity of the autoclave when experiments are in progress. This was achieved by building a control panel outside the restricted area, where the facility stands as shown in Fig. 4.

In addition to these safety features, the program used to continuously record all data during experiments has the capability to send a message to the operator via a pager whenever the temperature or pressure exceed a preset range for the experiment.

Precautions are taken in order to prevent the release of potentially contaminated liquid. As discussed, the exit of the rupture disk is connected to a sparge tank such as the potentially contaminated water would be collected and stored in IMTL. Also, the non-regenerative heat exchanger is connected to a closed-loop chiller in which the heat removed from the testing environment is transferred to the air.

2.3. Specimen loading procedure in the hot cell

Radioactive specimens are first transferred to a lead container before the loading assembly is rolled inside the hot cell. A *guideboard* equipped with cameras and removable guiding rods is installed on the loading assembly to assist the operator in the specimen loading operation. The specimens are then successively removed from the lead container and installed in clevises. In the current design, the clevises were designed to be compatible with a batch of neutron-irradiated specimens identified. The specimens are dog bone-type with a gage section of 2.4 mm wide by 0.5 mm thick (Fig. 5). Considering the small size of the specimens, the operation is done using a mounting jig and illuminated magnifier, specially made for the clevises. Once a specimen has been inserted into the clevis, it is loaded on the loading jig. The cameras installed on the guideboard assist the operator in mounting the specimens.

Once the specimens are installed, the guide rods are inserted in the guideboard, the hoist picks up the autoclave body and lowers it down over the head. The hoist then picks up a custom-made flange tensioner and installs it over the autoclave body. The use of a flange tensioner to seal the autoclave assures the application of uniform pressure around the entire seal and provides a reproducible process that greatly enhance success in achieving a watertight seal for very high-pressures.

The autoclave is connected to a high-pressure pump and is filled with pure water and pressurized. Once the autoclave was able to maintain pressure without leakage, the pressure is relieved and the tensioner can be removed along with the guideboard. The heating bands are installed with the thermal insulator. A shield is installed around the insulation as needed. After radiation survey, the loading unit is ready to be rolled outside the hot cell to be connected in IMTL.

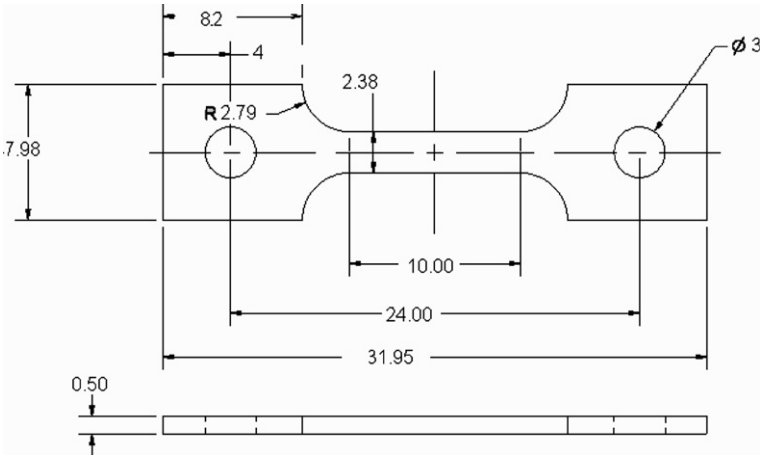

Fig. 5. Schematic of the specimens to be used to simulate irradiated specimens. Dimensions are given in mm.

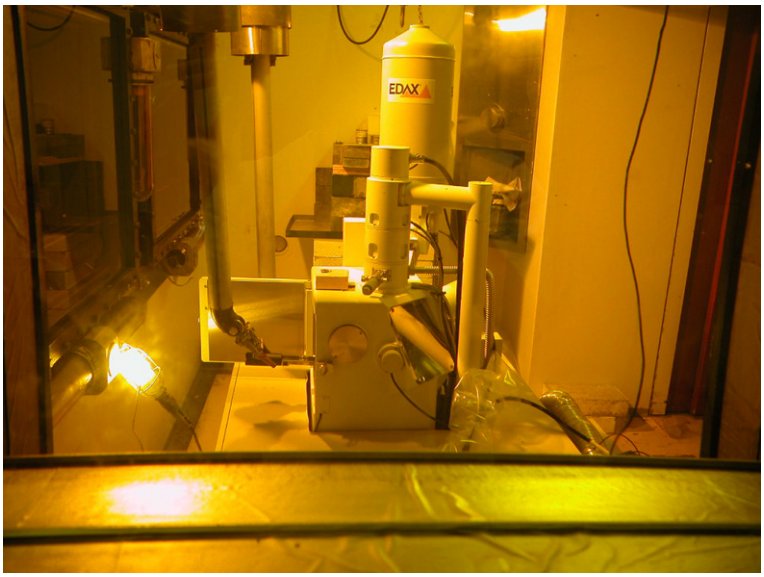

Fig. 6. SEM installed for observation of irradiated specimen in the hot cell. Note that the microscope is installed on a 'pan' that provides 'clean' air to cool the microscope.

2.4. SEM analysis

A JEOL Model JSM-6480 scanning electron microscope is used for post-test analysis. It is equipped with an Everhart Thornley detector for secondary electron imaging and a backscattered electron detector that provides compositional, topographic and shadow images. It also has a Genesis 2000 XMS system 60 energy dispersive spectrometer (EDS) from EDAX. The EDS has a sapphire detector with a resolution of 130 eV. For observation of irradiated specimens, the microscope is installed in the hot cell (Fig. 6) and connected to the control console outside the hot cell.

The column is pumped by a turbomolecular pump to eliminate the need for water recirculation and providing easy installation of the column in a hot cell. As the hot cell floor is a potential source of contamination, the microscope is connected to an external air source to assure that the air flowing through the assembly for cooling does not contain any radioactive particles.

3. Facility validation

3.1. Validation of facility for CERT experiments

The facility was validated in two steps. First, the facility demonstrated the capability to reproduce experiments previously performed in our cold-laboratory and produce similar results. Second, the feasibility of the procedures created for testing irradiated specimens and the control of their applications was demonstrated.

For the first step, a set of experiments previously performed at the University of Michigan in the High Temperature Corrosion Laboratory was repeated. The experiments were performed on an identical set of alloys: austenitic stainless steel alloys 304 (UNS S30400) and 316L (UNS S31603) and nickel-base alloys 625 (UNS N06625) and 690 (UNS N06690). The specimens were strained to failure at 3×10^{-7} s^{-1} in deaerated pure supercritical water (conductivity <0.1 μ S/cm, dissolved oxygen <10 ppb) at 500 °C. Straining was started after conditioning the system at 500 °C for about 139 h when the water chemistry was stable at target conditions. Before starting the test, it was verified that all parameters were at or very near the target values with acceptable oscillations. Other than the fact that the alloy 625 specimen failed outside of the gage section, the correlation between the results (crack density, average crack length, crack length per unit area, stress strain curves), presented in Table 1, confirms the reliability of the facility and procedure used at IMTL.

For the second step, the entire process of specimen loading in the hot cell, CERT experiment in 500 °C deaerated SCW performed in IMTL, specimen unloading and specimen observation by SEM,

Table 1
Summary of stress–strain and cracking results obtained in IMTL and in reference tests in 500 °C deaerated pure supercritical water

Tests	Alloys	Maximum strength (MPa)	Rupture strain (%)	Fracture mode	Crack density (#/mm^2)	Crack length (μm)	Maximum crack depth (μm)
Validation	304L	420	36.8	IG + ductile	59.3	46.8	49.9
	316L	370	36.6	Ductile	23.3	46.8	23.0
	690	475	41	IG + ductile	19.8	32.6	27.7
Reference	304L[a]	340[a]	25[a]	Did not fail	39.4	32.2	51.4
	316L	350	33	Ductile	38.1	28.5	24.9
	690	455	42	Granular + ductile	32	24.9	33.1

Table 2
Comparison between CGR result obtained at UoM and at GE

	GE	UoM
Alloy	21% Cold-work 316L	21% Cold-work 316L
Specimen	Side grooved 0.5T-CT,	Side grooved 0.5T-CT
Environment	288 °C, 2 ppm DO, pure water	288 °C, 2 ppm DO, pure water
CGR	2.7×10^{-7} mm/s	2.3×10^{-7} mm/s

has been validated by treating a set of unirradiated specimens as if they were irradiated.

3.2. Validation of the facility for CGR experiments

In order to validate the experimental facility for crack growth rate (CGR) experiments, a CGR test of cold-worked 316 stainless steel in a simulated boiling water reactor (BWR) environment, previously performed at GE Global Research, [2] was reproduced. The specimen used for the test was a side grooved 0.5 T–CT specimen that was fabricated from 21% cold-worked 316 stainless steel. It was first pre-cracked in air followed by in situ pre-cracking and constant K stress corrosion cracking in the environment. The conditions and procedure of the benchmark CGR test are summarized in Table 2. The crack length was measured using the DCPD technique.

The experiment was conducted in constant K mode for 300 h during which the crack was grown by about 250 μm to ensure a straight crack front, several grain diameters deep. The crack growth rate was evaluated by performing a linear fit of the data provided by the DCPD measurement. Over 300 h, the correlation coefficient of the linear fit was 0.9989. The standard error in crack length: $\sigma_a = \sqrt{(a_m - a_f)^2/(n-2)} = 24.4$ μm and the error in CGR was 2.3×10^{-8} mm/s, which was about 10% of the measured CGR. Considering the error in crack length measurement, the error in K during the constant K test was calculated to be 0.077 MPa$\sqrt{m}$. These results indicate a well-controlled experiment from which the recorded CGR is reliable. Based on the DCPD measurement at constant K, the CGR was constant at 2.3×10^{-7} mm/s, Fig. 7. Result is to be compared with the value of 2.7×10^{-7} mm/s obtained with the same alloy in the same condition (cold-worked, same environment, same K applied) and published by

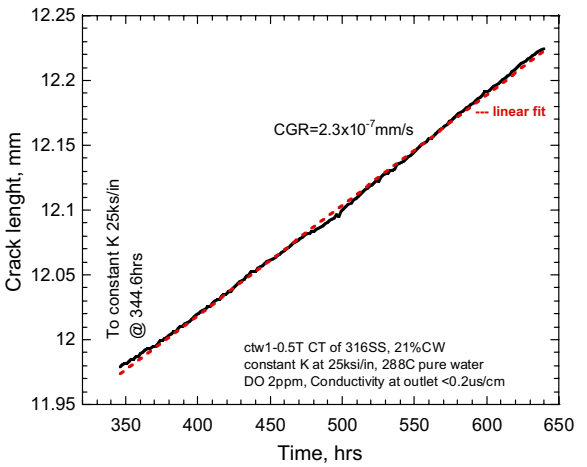

Fig. 7. The crack growth behavior of cold-worked 316 stainless steel, under constant K (25 ksi in$^{0.5}$) in 288 °C water containing 2 ppm DO.

Andresen et al. [2]. The good correlation between the results obtained in the two laboratories on the same alloy and for the same testing condition confirms the reliability of the facility and procedure used at IMTL.

4. Summary

A facility to conduct stress corrosion cracking experiments on neutron-irradiated specimens in supercritical water has been built at the University of Michigan. Experiments can be conducted in pure supercritical water, up to 30 MPa of pressure and 600 °C, in a controlled, refreshed environment. In this environment, four tensile specimens can be loaded under constant extension rate. Used with a DCPD system, the facility permits to load one CT under constant K and to measure the crack propagation rate. The facility has been validated for CERT experiments by reproducing a set of experiments previously performed at the University of Michigan in the High Temperature Corrosion Laboratory. Validation for CGR was done by reproducing an experiment which results were published by Andresen et al. [2]. Test procedures for radioactive specimens have been validated by treating a set of unirradiated specimens as if they were irradiated.

Acknowledgement

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