



ELSEVIER

Contents lists available at ScienceDirect

Corrosion Science

journal homepage: www.elsevier.com/locate/corsci

Effects of dissolved hydrogen on the crack-initiation and oxidation behavior of nickel-based alloys in high-temperature water

Taeho Kim, Kyoung Joon Choi, Seung Chang Yoo, Ji Hyun Kim*

School of Mechanical and Nuclear Engineering, Ulsan National Institute of Science and Technology (UNIST), 50 UNIST-gil, Eonyang-eup, Ulju-gun, Ulsan 44919, Republic of Korea

ARTICLE INFO

Article history:

Received 10 October 2015
Received in revised form 7 January 2016
Accepted 5 February 2016
Available online xxx

Keywords:

A. Alloy
A. Nickel
B. Raman spectroscopy
B. TEM
C. High temperature corrosion

ABSTRACT

The effects of dissolved-hydrogen concentration on the formation of oxide films on Alloy 182 were investigated, along with the crack-initiation time. Results from both in-situ Raman analyses and ex-situ electron microscopy analyses indicated that NiO formed at lower dissolved-hydrogen concentrations as the NiO Raman peak decreased, whereas Cr₂O₃ peak and spinel oxide peaks appeared as the dissolved-hydrogen concentration increased. Crack-initiation testing showed that the crack-initiation time decreased as the dissolved-hydrogen concentration increased. The stable and thick NiO could act as a barrier to ion diffusion, and may increase the crack-initiation time of nickel-based alloys.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Nickel-based alloys and their weld metals are widely used as key materials for critical structural components in high-energy systems. In fossil and nuclear power plants and turbine generators, for example, they are used to construct steam generator tubes and high-energy piping nozzles. In these applications, nickel-based alloys and their weld metals are normally exposed to high-temperature water, which may lead to cracking initiated by the oxidation process. Further, such cracking may compromise the integrity of the entire system. This problem has been investigated in several studies [1–10], and it has been determined that primary water stress corrosion cracking (PWSCC) is one of the most important degradation modes of nickel-based alloys applied in the primary cooling circuits of pressurized water reactors in nuclear power plants [11–18]. Although such nickel-based alloys are well protected against rupturing by design-based safety margins, PWSCC is a significant concern as regards nuclear power plant safety, because the steam generator tubing or nozzles may rupture as a result of this phenomenon.

Earlier studies [19–22] have shown that the concentration of dissolved hydrogen in high-temperature water can affect the PWSCC susceptibility of the nickel-based alloy, Alloy 600, and

those of the Alloy 82 and Alloy 182 weld metals. The characteristics of oxide films play an important role in the crack-growth mechanism, and it is well known that the crack-growth rate and crack-initiation time of nickel-based alloys are also influenced by the Ni–NiO equilibrium electrochemical potential [23]. Further, it has been determined that the crack-growth rate and PWSCC initiation time vary as functions of the dissolved-hydrogen concentration in high-temperature water, which may constitute evidence that the oxide films present on the surfaces of nickel-based alloys in primary water play a key role in the PWSCC mechanism. Therefore, understanding of the oxide film behavior during the cracking process is important, and in-situ investigations of oxide film characterization may facilitate comprehension of PWSCC in a general sense.

Therefore, in order to establish the relationship between the crack-initiation time and the oxide characteristics on Alloy 182, a crack-initiation test and in-situ Raman spectroscopy are conducted on U-bend specimens in a high-temperature aqueous environment in this study. However, a limitation is encountered in that the results for the crack-initiation time obtained using optical microscopy images are measured in an ex-situ experiment. This is problematic as specimen oxide characteristics and structures are modified as soon as they are removed from a high-temperature corrosive environment. Therefore, Raman spectroscopy is used for in-situ investigation of the high-temperature and high-pressure aqueous environment in this experiment. As demonstrated in previous studies, Raman spectroscopy can measure the wavelength

* Corresponding author.

E-mail address: kimjh@unist.ac.kr (J.H. Kim).

and intensity of light scattered from a specimen irradiated by a high-energy laser, based on the energy emitted by the test-sample molecules [24,25].

The present work investigates the crack-initiation time of Alloy 182 U-bend specimens using optical microscopy, and also examines the oxide film characteristics of oxidized Alloy 182 samples using in-situ Raman spectroscopy, for different dissolved-hydrogen concentrations under primary-water conditions in a pressurized water reactor. This investigation facilitates an understanding of the relationship between the oxide film characteristics and the cracking process in this alloy. Along with in-situ Raman spectroscopy measurements, ex-situ surface analyses, including scanning electron microscopy (SEM), transmission electron microscopy (TEM), and energy-dispersive X-ray spectroscopy (EDS), are performed.

2. Experimental

2.1. Materials and specimen preparation

To simulate the nickel-based-alloy weld metals examined in this study, stainless steel 316L and an Alloy 182 weld deposit were used. Fig. 1 presents sketches of the stainless steel 316L and Alloy 182 weld deposit specimens, in which the dimensions in the longitudinal (L), transverse (T), and short transverse (S) directions are given. A weaved-bead welding technique was used, with the welding rod traveling in the L direction with sideward displacements of a few millimeters in the T direction. The shielded-metal arc-welding method was employed, using ENiCrFe-3 welding rods. Post-weld heat treatment was not conducted, but a certified material test report and defect inspection were completed. To confirm that the Alloy 182 weld metal characteristics satisfied the American Society of Mechanical Engineers composition specifications and conformed to the information provided in the certified material test report, chemical-composition analysis was conducted at the Korea Testing & Research Institute. Specimens were collected from various locations on the Alloy 182 samples used in this study, and the chemical composition results are summarized in Table 1. It was confirmed that no defects were caused by the sample-manufacturing process.

The T–L direction was chosen as the variable dimension of the U-bend specimen. The oxidized specimen designs are illustrated in Fig. 2. The specimens subjected to in-situ Raman spectroscopy had a U-bend shape, and two different designs were used in order to simulate different stress conditions, in accordance with the ASTM G30-97 standard. The high- and low-stress specimens had U-bend radii of $R=8.3$ and 12.5 mm, respectively. Further, the high- and low-stress specimen deflections were 12% and 8% for fixed strain, respectively. The strain was maintained during the corrosion experiment using a horizontal bar and spring.

2.2. Control of water chemistry

In order to simulate the primary water used in pressurized water reactors, a loop was used to generate both high temperature and pressure, along with an autoclave; the experimental temperature and pressure were set to 325 °C and 180 MPa, respectively. Water with a resistivity of 18 M Ω cm was mixed with chemicals to produce a solution containing 1200-ppm (mg/kg) boron(III) cations (from boric acid) and 2-ppm (mg/kg) lithium ions (from lithium hydroxide); this chemical composition is identical to that of typical pressurized-water-reactor primary water.

The dissolved-hydrogen concentration was strictly controlled by bubbling a mixture of hydrogen and argon gas into the water reservoir. Three different dissolved-hydrogen concentration values (5 , 25 , and 50 cm³/kg, at standard temperature and pressure) were selected in order to investigate the effect of the dissolved hydro-

gen on the oxide film on Alloy 182 at high temperature. Note that Xu et al. [26] have estimated that the dissolved-hydrogen content at the Ni–NiO transition at 320 °C is approximately 9.16 cm³/kg. Therefore, this indicates that Ni is stable at dissolved-hydrogen concentrations of 25 and 50 cm³/kg, whereas NiO is stable when the dissolved-hydrogen concentration is at 5 cm³/kg. Finally, in this study, the dissolved-oxygen concentration was maintained below 5 ppb (μ g/kg) in order to eliminate the effects of oxygen on the specimen surface.

2.3. In-situ Raman spectroscopy apparatus

In-situ Raman spectroscopy measurements were conducted using a RamanRXTM, manufactured by Kaiser Optical Systems, Inc., which uses a 532-nm-wavelength krypton-ion laser with a maximum power of 100 mW. To prevent damage to the specimens, the power density at the specimen surface was maintained below 10 mW/cm², and the specimen-surface irradiation area was set to approximately 100 μ m in diameter. The in-situ Raman system consisted of four parts, including the immersion optics used to move the laser toward the sample, a notch filter for signal processing, a band-pass filter, and a charge-coupled device detector. Detailed descriptions of the Raman system are available in the literature [24,25,27]. The probe optics for the in-situ Raman investigation was installed in the autoclave within 1 mm of the sample, which is within the maximum focal length of the laser system.

2.4. Experimental procedure

In-situ Raman spectroscopic measurements and crack-initiation tests were performed for Alloy 182 U-bend specimens in a primary-water environment. The U-bend-shaped Alloy 182 specimens were exposed to primary-water chemistry conditions until crack initiation was observed, or until the exposure time reached 1000 h. At each dissolved-hydrogen concentration, a total of four specimens was used for the crack-initiation test; among these specimens, two had $R=8.3$ mm and the other two specimens had $R=12.5$ mm. The tests were conducted in an autoclave system that was specially constructed for this study. Extremely rigorous control of the water chemistry was exercised and near-theoretical water conductivity was routinely achieved. The dissolved-hydrogen concentration of the inlet water in the loop was monitored continuously using an OrbisphereTM dissolved-hydrogen sensor, so as to control the dissolved-hydrogen concentration in the high-temperature water. In addition, the solubility of hydrogen in water decreases with increased temperature; however, it increases with increased pressure [28]. Therefore, it was possible to “cancel out” the effect of the temperature on the dissolved hydrogen. The dissolved-hydrogen concentration was controlled using the overpressure method, which is the same technique used in previous studies [20,29]. The apex of the oxidized specimen surface was observed using an optical microscope after periods of 24 , 100 , 500 , and 1000 h, so that any cracks could be identified. After the optical microscope analysis, a high-resolution transmission electron microscope (HRTEM) was used for microstructural characterization. Ga-ion milling with a focused ion beam (Quanta 3D Dual-Beam) was employed to prepare the TEM samples. The specimen surface was coated with carbon before the focused ion beam milling, so as to prevent both surface contaminations from impurities and ion-beam damage.

3. Results and discussion

3.1. Crack initiation at different dissolved-hydrogen levels

Observation of crack initiation at the apex of the oxidized specimen surface was conducted after the specimens were exposed to

Download English Version:

<https://daneshyari.com/en/article/7894708>

Download Persian Version:

<https://daneshyari.com/article/7894708>

[Daneshyari.com](https://daneshyari.com)