



Short communication

The repassivation kinetics study of Alloy 800 in high-temperature pressurized water

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ARTICLE INFO

Article history:

Received 16 July 2015

Received in revised form 19 August 2015

Accepted 20 August 2015

Available online 28 August 2015

Keywords:

Repassivation

Alloy 800

High-temperature water

Stress corrosion cracking

Nuclear power

ABSTRACT

The repassivation kinetics of Alloy 800 is studied in high-temperature pressurized water with different dissolved hydrogen (DH) and dissolved oxygen (DO) at 300 °C. The results demonstrate that under DO conditions, the *cBV* value increases with the rise of DO concentration; and under DH conditions, the *cBV* value passes through a local maximum at DH = 1.0 ppm. In addition, the repassivation results in the present work confirm the applicability of the slip-dissolution model for explaining stress corrosion cracking in high-temperature pressurized water by means of electrochemical measurement. Compared with other methods of evaluating stress corrosion cracking susceptibility of engineering materials, studying the repassivation kinetics by rapid scratch technique is a more economical and time-efficient evaluation method for nuclear materials in high-temperature pressurized water.

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1. Introduction

In order to explain the stress corrosion cracking (SCC) susceptibility of stainless steels (SSs) and nickel-based alloys in high-temperature pressurized water, several theories and models have been postulated, such as the slip dissolution/film rupture mechanism, the enhanced surface mobility theory, coupled environment fracture model, internal oxidation mechanism and hydrogen-induced cracking model. Among these theories and models, the slip-dissolution model [1–4] is one of the most widely used models to explain the occurrence of SCC for SSs and nickel-based alloys in high-temperature water. According to this model, cracks grow through the repetitive processes of film rupture, dissolution and repassivation. Since external factors such as loading conditions (e.g. stress, strain) and geometrical dimensions are excluded, repassivation kinetics has been considered as a key factor which determines the intrinsic SCC susceptibility of alloys [5–7]. The crack initiation and growth processes can be well reproduced by studying repassivation kinetics of materials. The rapid scratch method is most often used for measuring film repassivation kinetics [8]. However, due to the difficulty in achieving in situ scratch and measuring electrochemical signals during repassivation in the autoclave, currently there is no literature studying repassivation kinetics of alloys by in situ rapid scratch technique in high-temperature pressurized water.

It is well known that the water chemistry environment in a nuclear power plant has an obvious effect on the corrosion and stress corrosion

behavior of alloys in high-temperature water. A number of researchers reported that dissolved hydrogen (DH) and dissolved oxygen (DO) in high-temperature water influenced the corrosion rate and SCC susceptibility of nickel-based alloys [9–16]. Since the SCC of alloys in high-temperature water is essentially an electrochemical process, the purpose of this work is to study the repassivation kinetics of Alloy 800 in high-temperature pressurized water with different DO and DH water chemistry environments based on the in situ rapid scratch electrode system built in an autoclave.

2. Experimental

2.1. Samples and solutions

Nuclear-grade commercial Alloy 800 tubes were provided by the Atomic Energy of Canada Limited (AECL, Chalk River, Ontario, Canada). The chemical compositions of Alloy 800 (in wt.%) were 32.76 Ni, 21.90 Cr, 43.10 Fe, 0.49 Mn, 0.46 Ti, 0.45 Si, 0.28 Al, 0.015 N, 0.017 C, 0.010 Co, 0.015 Cu, 0.013 P and 0.001 S. The arc angle and length of scratch samples were 90° and 2.0 cm, respectively. The scratch samples were gradually grounded with silicon carbide papers up to 2000 grit and then point welded to the pure Ni wires.

The samples were immersed in simulated primary water (1500 ppm B and 2.3 ppm Li). Five different water chemistry environments were used: DO = 2.0 ppm (about 1.4 cm³ STP O₂/kg H₂O), DO = 0.1 ppm, DO and DH < 0.01 ppm, DH = 1.0 ppm (about 11 cm³ STP H₂/kg H₂O) and DH = 2.3 ppm. The DH and DO in the water loop were continuously monitored and controlled automatically by LabView software. The test temperature and pressure were 300 °C and 10.0 MPa, respectively.

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2.2. Electrochemical measurements and rapid scratch test

After the scratch samples were immersed in autoclave for 120 h at 300 °C, a Gamry Reference 600 potentiostat was used to measure the open circuit potential and potentiodynamic polarization curves. The three-electrode system including working, counter and external Ag/AgCl reference electrodes was used. The counter electrode was a platinum foil with a size of 20 × 20 mm², and the concentration of KCl in the Ag/AgCl reference electrode was 0.1 mol/L. The potentiodynamic polarization measurements were performed at a scan rate of 0.5 mV/s. In order to form a stable passive film on the surface of Alloy 800, the scratch samples were polarized at the preset potential under potentiostatic condition for 1200 s. Then, the actuating device was switched on and the Al₂O₃ scratch pin began to scratch the surface of the samples rapidly. The rapid scratch system used in the present work has been detailed in Ref. [8]. The scratch speed and time of each sample were about 2 m/s and 0.01 s, respectively. The data-collecting frequency was 1000 Hz in order to obtain precise data during the scratch process. The scratch width of each sample was 0.05 ± 0.01 mm.

All the potentials in the present work have been converted to standard hydrogen electrode (SHE) using the following equation [17]:

$$E_{\text{SHE}} = E_{\text{obs}} + 0.2866 - 0.001 \times (T - T_r) + 1.745 \times 10^{-7} \times (T - T_r)^2 - 3.03 \times 10^{-9} \times (T - T_r)^3 \quad (1)$$

where E_{SHE} is the electrode potential vs. SHE, E_{obs} is the measured electrode potential, T is the experimental temperature, and T_r is room temperature (25 °C).

3. Results and discussion

3.1. Potentiodynamic polarization for Alloy 800

Fig. 1 shows the potentiodynamic curves for Alloy 800 in high-temperature water with different DO and DH at 300 °C. It can be seen that the corrosion potential of Alloy 800 decreases gradually when the water chemistry environment changes from DO condition into DH condition. Besides, this picture reveals that passivation occurs in Alloy 800 with DO and DH < 0.01 ppm, DH = 1.0 ppm and DH = 2.3 ppm, and the potentials of −0.298 V, −0.266 V and −0.202 V are selected as applied potentials during the scratch test in high-temperature water. The reason is that the above three selected potentials are located in the stable passive region and respond to the minimum passivation current densities at each condition. Fig. 1 also reveals that Alloy 800 is passive at

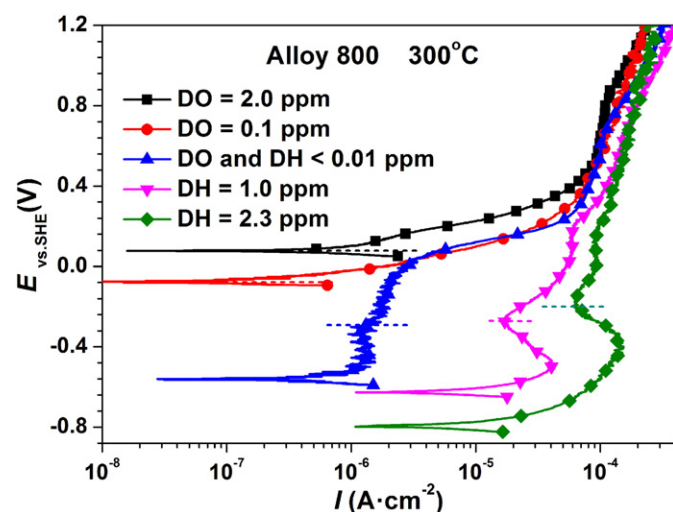


Fig. 1. The potentiodynamic polarization curves for Alloy 800 after 120 h immersion with different DH and DO.

the open circuit potential (OCP) in high-temperature water with DO = 0.1 ppm and DO = 2.0 ppm, respectively. Therefore, the potentials of +0.087 V and −0.089 V (+10 mV above OCP) are selected as the applied potentials. According to potential-pH diagrams for Ni, Cr and Fe calculated at 300 °C [18], the stable states of Ni, Cr and Fe at the above applied potentials are the oxidation states, i.e., NiO, Cr₂O₃ and Fe₂O₃, respectively. All the applied potentials under each condition are shown in Fig. 1 with dash lines.

3.2. Current transient measurement in each environment

The current transient curves under the preset potential at each condition are shown in Fig. 2a. Once the scratch pin breaks the oxide film on the surface of Alloy 800, the current transient increases suddenly to a peak due to anodic oxidation reaction, and then reduces as the repassivation process proceeds. Fig. 2b and c show the curves regarding the current transient density $i(t)$ versus the reciprocal of charged density $1/Q(t)$ for Alloy 800 with different DO and DH in high-temperature water, respectively.

Based on the high-field ion conduction model proposed by Cabrera and Mott [19], Burstein et al. [20] used scratch electrode technique and reported that $\log i(t)$ is linearly proportional to $1/Q(t)$ as shown in the following equation.

$$\log i(t) = \log A + \frac{cBV}{Q(t)} \quad (2)$$

where A and B are constant parameters associated with the activation energy for mobile ion migration, V is the potential drop across the film, c is a constant value equal to $zFp/2.3M$, $i(t)$ and $Q(t)$ are the current transient and charge density flowing from the scratch, respectively. Wang et al. [8] reported that the passive films of Alloy 690 in high-temperature water with DH < 0.01 ppm at 300 °C initially grew according to the place exchange model, and thereafter grew according to the high-field ion conduction model. Kwon et al. [7,21,22] studied the repassivation kinetics of stainless steels and Alloy 690 using rapid scratch technique below 100 °C and reported that the value of cBV was an effective parameter which demonstrated the repassivation rate and SCC susceptibility of alloys. They put forward that the SCC susceptibility of alloys increased with the rise of cBV . Fig. 3 shows the change of cBV in high-temperature water with different DO and DH at 300 °C. Under DO conditions, the cBV value increases with the rise of DO concentration; under DH conditions, the cBV value passes through a local maximum at DH = 1.0 ppm. The changing trend of cBV for Alloy 800 is consistent with that of the SCC growth rates for SSs and nickel-based alloys in high-temperature water with DO and DH [8,15,16,23–26]. It can be seen that the value of cBV obtained by studying repassivation kinetics in high-temperature pressurized water can demonstrate the SCC susceptibility of alloys under different water chemistry environments. Although the slip-dissolution model [1–4] is one of the most widely used models to interpret the occurrence of SCC for SSs and nickel-based alloys in high-temperature water, there is no direct theoretical evidence found to verify this model. The repassivation results in the present work confirm the applicability of the slip-dissolution model in high-temperature pressurized water by means of electrochemical measurement.

Compared with other methods of evaluating SCC susceptibility of engineering materials, including measuring the time-to-failure and the critical stress for cracking, the critical cracking potential under constant loading conditions and the strain-to-failure ratio using constant strain rate test, studying the repassivation kinetics using rapid scratch technique is a more economical and time-efficient evaluation method for nuclear materials in high-temperature water.

4. Conclusion

The repassivation kinetics of Alloy 800 was studied by the in situ rapid scratch electrode technique in high-temperature pressurized

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