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Effect of applied stress on passivation kinetics and passivation modelling of 304L stainless steel in acidic medium

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1. Passivation Role in Stress Corrosion Cracking (SCC):

- Film rupture-dissolution model (FRM): describes intergranular SCC, shown in Fig.1, as repetitive cycles of local surface activation, dissolution, and passivation near the crack tip [1]. Fig. 2 illustrates one of these cycles as described next:

   - To develop a model quantifying the passivation kinetics and parameters of passive films constructed in acidic medium.
   - To use this model to check the influence of stress on stainless steel passivation and passive film quality.

2. Quantification of Passive Film Thickness and Quality in Acidic Medium:

   a. Atomic Emission Spectroscopy and Conventional Electrochemical Experiments

      - Stressed and non-stressed samples of 304L stainless steel are subjected to potential cycling tests as shown in Fig. 2. These serve as an electrochemical simulation of SCC activation/passivation cycle as described by FRM.
      - Experiments were performed in 2 M H2SO4 solution at room temperature.
      - In-situ atomic emission spectroelectrochemistry (AES/EC) [3] was coupled with conventional electrochemistry during the tests. By this, the passivation current due to metallic dissolution can be quantified, as shown by Fig. 4.

   b. High Field Ion Conduction Model and Passive Film Thickness (HFIC)

      - HFIC was adapted to describe the current evolution during passivation [2], as given by eq.1.
      - Passive film thickness can be calculated using Faraday’s law as indicated by eq.2.
      - The term $q_{\text{film}}$ represents the charge exchange responsible for passive film formation.
      - The external current during passivation is due to three components as explained by eq.3.
      - In acidic solutions, the major part of anodic current measured during passivation is due to metallic dissolution [2,3], where $q_{\text{dissolution}}$ is negligible [3].
      - The portion of charge due to film formation can be calculated by quantifying the dissolution component as given by eq.4.
      - A particular approximation is taken as $i_{\text{external}} = i_{\text{dissolution}}$ to calculate the current fraction due to iron dissolution $i_{Fe}$. By this, $i_{Fe}$ can be calculated as shown by eq.7.
      - The calculation of $i_{Fe}$ is based only on the metallic mass fractions in the alloy and their oxidation numbers in such acidic medium as derived from Faraday’s second law [8].
      - Using AES/EC quantified data for metallic dissolution during passivation (region C in Fig.4), $i_{Fe}/i_{Fe}$ can be calculated. This ratio can be used in eq.9 to calculate $i_{Fe}$ for the concerned metals. By eq.2, passive film thickness is calculated as shown in Fig.5.

3. Influence of Stress on Passive Film Thickness and Quality:

   - For stressed samples, Fig. 6 shows a direct measure of the passivation rate and the film Ionic conductivity.
   - $i_{Fe}$ is inversely proportional to the film quality [4]. Fig. 7 shows this factor for stressed and unstressed cases.

References: