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Experimental and boundary element method study on the effect of stress on the polarization curve of cast aluminum alloy in sodium chloride solution

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ABSTRACT

The effect of stress on the polarization curve of high pressure die cast aluminum alloy in sodium chloride solution was evaluated by mechano-electrochemical experiments and boundary element electrostatics simulations. The polarization curve was measured under a constant stress condition, and a decrease in natural potential due to stress was observed. A periodic arrangement of intact and damage surfaces was assumed in the mechanically damaged oxide film, and the polarization curve of the damage surface was identified. The simulation results showed that the decrease in natural potential is explained by the nucleation of the damage surface by plastic strain.

1. Introduction

Aluminum alloys are widely used in the automobile and aerospace industries owing to its light weight, high strength, and high corrosion resistance. Their corrosion resistance arises from the passive film made of hydrous aluminum oxide, which is stable in a neutral environment, but soluble in strong acidic or alkaline environments as a form of general corrosion [1–3]. Moreover, when the solution contains aggressive anions such as chlorine, localized corrosion or crevice corrosion occurs even in a neutral environment. In this case, cathodic protection does not effectively work because the cathodic reaction of water and oxygen makes the pH increase and consequently helps an alkaline dissolution [2]. When the oxide film is damaged electrochemically or mechanically and the raw aluminum surface is partially exposed to the electrolyte, the repassivation or localized corrosion starts depending on the local environment at the damaged site. Aggressive anions prevent the repassivation and often lead to localized corrosion and pitting associated with micro galvanic corrosion. Localized corrosion occurs mainly around geometrical defects like scratches, second phase particles, impurity inclusions, or grain boundaries [2,4–6]. These sites are also a stress concentration site where the oxide film is most likely to be damaged by the mechanical stress applied to the material. The stress concentration causes damage of the oxide film and generates a new surface without passive film, especially in the case of high stress, leading to a plastic strain. In the environment containing chlorine ions, the damaged surface is prohibited from the repassivation and micro

galvanic corrosion occurs due to the potential difference between the damaged surface and surrounding intact oxide film. Once the corrosion pit grows, repassivation is more severely prevented and also the metal dissolution is accelerated by the acidified environment inside the pit. The pit growth causes extreme increase in stress concentration and finally nucleates a crack which causes the stress corrosion cracking (SCC) or corrosion fatigue (CF) [2].

For the safety of industrial products, the prediction of corrosion in its type and rate is important in the stage of design but quantitative prediction is difficult to yet execute. For material failures caused by SCC or CF, the evaluation of crack growth rate based on linear fracture mechanics and life time prediction using the crack growth rate are the most popular and have been studied by many researchers [1,2,7]. However, since this approach often relies on an empirical relationship between the crack growth rate and the stress intensity factor in some limited environments, the contributions of corrosion, embrittlement and stress are almost unknown and cannot be evaluated individually. Moreover, the crack initiation mechanism has been studied only qualitatively but not quantitatively, therefore there is no evaluation method for the crack initiation life in which the local corrosion and local plastic deformation play an important role. Consequently, numerical approximation methods are required to evaluate the local corrosion rate in the initial stage of SCC because of the complexity of multi-physics phenomena consisting of the electrochemical reactions and mechanical damages.

Corrosion is a chemical reaction and its rate can be detected by the

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Table 1
Chemical composition of JIS ADC12 (wt%).

Cu	Si	Mg	Zn	Fe	Mn	Ni	Sn	Pb	Ti	Al
1.5–3.5	9.6–12.0	≤ 0.3	≤ 1.0	≤ 1.3	≤ 0.5	≤ 0.5	≤ 0.2	≤ 0.2	≤ 0.30	Bal.

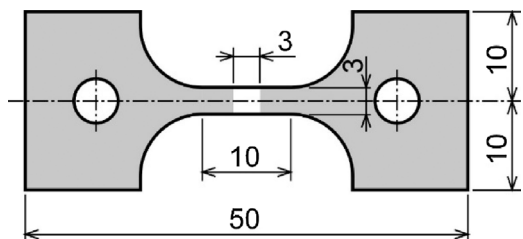


Fig. 1. Tensile specimen. Grey region was covered by the insulation coating.

electric current. If the electric field can be evaluated exactly around the surface of materials, the corrosion rate can also be determined from the electric current in spite of a nonuniform current density, that is, local corrosion. From this viewpoint, the electric field of galvanic corrosion has been studied for a long time [8,9], and the boundary element method (BEM) has been applied to practical corrosion problems by many researchers [10–15]. The advantage of BEM is not only its computational efficiency that comes along with its reduced integration dimension but also the ease of handling of a singularity or discontinuity of the field variables [16–18]. In the corrosion problem, the electric potential and current density are discontinuous on the border between different materials. If this discontinuity is considered in the finite element method (FEM), it intrudes into the analysis domain of electrolyte solution which, in practice, has to have a continuous electric field inside. By contrast, BEM can naturally account for this discontinuity only on the boundary without any difficulties.

To perform the simulation of stress-assisted corrosion, we have to clarify the effect of stress on the electrochemical property and obtain a formulation of stress- or strain-dependent polarization curve to be used in the BEM electrostatics simulation. However, despite of a few experimental studies [19–22], the precise effect of stress on the electrochemical property is still mostly unknown. Butler et al. [19] have shown the effect of stress on the electrochemical property of austenitic stainless steel. They studied the variation of the polarization curve (PC) as the applied compressive stress increased within less than 90 MPa, and also showed the shift in open-circuit potential (OCP) due to the compressive stress up to 40 MPa with various stress rates. Darowicki et al. [20] have shown the effect of stress on the impedance of aluminum alloy surfaces. Krawiec et al. [21] have shown the effect of microcracks with breakage of precipitates under a few percent plastic strain on the local PC of cast aluminum alloy measured by the microcapillary technique [23,24]. Orlikowski and Darowicki [22] have shown the variation of impedance of aluminum alloy during tensile tests until the specimen breaking point and also showed the continuous variation of OCP with respect to the applied strain until the breaking point.

In this study, we first carried out a mechanical and electrochemical experiment to measure the PC of high-pressure die cast aluminum alloy under a constant tensile stress condition in a NaCl solution at room temperature. The effect of stress was quantified by the shift in natural potential (NP) determined from the PC which, in principle, is the same as the OCP. Next, we performed the BEM simulation with a discontinuous element discretization [25–28] to evaluate the mechanism of the shift in NP due to stress. The effect of stress was assumed to mainly arise from the mechanical damage of oxide film induced by plastic strain. To consider the plastic damage, the surface is divided into two portions: intact and damage portions. The damage surface is supposed not to be covered by the oxide film and to have a PC different

from the intact surface. The PC of the damage surface was identified from the comparison between experiments and simulations so as to equalize the NP when high stress was applied in the plastic regime. Periodicity of the electric field was assumed in the BEM simulation. Since the actual dimension of the damage surface was unknown, the effects of unit cell size and number of repetition were examined. The electric field in polarized conditions was also calculated to investigate the corrosion activity on the damage surface. Finally, we discuss the effects of stress and plastic strain on the PC and NP through comparison between the experimental and numerical results.

2. Experimental methods

2.1. Material

The material used was JIS ADC12 high pressure die cast aluminum alloy which is widely used in cast parts of automobiles. The chemical composition is listed in Table 1. Two types of specimen were prepared to examine the effect of cast surface. One was a specimen with an as-cast skin left and the other is a polished smooth specimen. Fig. 1 shows the shape of the specimen that was cut out from a cast plate of 3 mm in thickness. The specimen was coated by an electric insulation paint except for the test window of 3 mm × 3 mm as shown in Fig. 1. Moreover, the inside of the pin hole was covered by an adhesive cloth tape to reinforce the insulation. Fig. 2 shows the picture of the test window of as-cast and polished specimens. Many pores were exposed on the surface of the polished specimen. The cast surface was relatively rough with some scratches.

The mechanical property was obtained by tensile testing of another tensile specimen extracted from a cast plate of the same type. The tensile test was carried out in an air environment at room temperature and its result is shown in Fig. 3. The Young's modulus was 72.2 GPa, 0.2% proof stress was 162 MPa, tensile strength was 282 MPa, and elongation was 0.0246. The standard deviations for three samples were 0.591 GPa, 1.24 MPa, 13.1 MPa, and 0.0042, respectively. As it is natural for casting materials, the tensile strength and elongation strongly depend on the casting defects but the Young's modulus and proof stress are stable. The stress-strain curve in Fig. 3 is the result of the largest elongation in the three samples. This will be used to determine the plastic strain corresponding to the tested stress in the mechano-electrochemical experiment, i.e., the measured digital point of stress and strain nearest to the tested stress will be used to determine the corresponding plastic strain. Also in the simulation, these will be used to convert the simulated plastic strain to the applied stress.

2.2. Mechanical and electrochemical measurement

The PC was measured under constant stress. Fig. 4 shows the apparatus used in the experiment. The acrylic chamber was mounted on the round bar fixed by the grip of an MTS hydraulic fatigue test machine. The specimen was fully immersed in the electrolyte solution. The corrosive environment used was aerated 1.0% NaCl aqueous solution at room temperature. Three-electrode system and potentiodynamic mode were employed to measure the PC. The counter electrode was Pt wire and the reference electrode was Ag|AgCl installed in the Luggin capillary. The scanning range was from –1.5 to 0.5 V vs Ag|AgCl where the scanning rate was 1.0 mV/s and sampling rate was 1 Hz. The stress was held constant at prescribed stress values during the electrochemical

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