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Effects of flow, Si inhibition, and concurrent corrosion of dissimilar metals on the corrosion of aluminium in the environment following a loss-of-coolant accident

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ABSTRACT

Experiments were conducted to investigate the aluminium corrosion in the simulated containment pool environment following a loss-of-coolant accident at a nuclear power plant. The results show an identical power law correlation between the corrosion rate and mass transfer coefficient for different flow systems. Aluminium corrosion is inhibited by the silicon leached from the insulation materials, and the inhibition efficiency increases with the silicon concentration due to the increase of Al₂SiO₅ or SiO₂ content in the passivation layer. The concurrent corrosion of copper and iron has little influence on aluminium corrosion while the presence of zinc significantly retards the aluminium corrosion.

1. Introduction

Severe challenges to the safe operation of pressurized water reactors are posed by the loss-of-coolant accident (LOCA) [1], in which the hot and pressurized coolant is ejected from the primary cooling system into the reactor containment via a broken pipe. The emergency core cooling system is designed to pump the coolant collected in the containment sump back to the primary cooling and spray systems to assist in cooling the reactor after LOCA. However, the materials in the containment can be corroded by the hot coolant, and the resulting corrosion products with the formed chemical precipitates can block the sump pump screen, thus creating a head loss and a potential blockage for the circulation of coolant back to the reactor core. Previous studies have shown that the aluminium is the most corrodible metal and has the highest potential for mass release in the post-LOCA solutions [2–4]. The soluble corrosion products could be saturated to form precipitates such as aluminium hydroxide, and also react with other chemicals released from the calcium silicate and fibreglass pipe insulations to form solid precipitates like sodium aluminium silicate [2,5,6]. Experimental results from different groups have shown the raised head loss across a pump inlet strainer due to the dissolution and precipitation of aluminium [7–10].

The coolant contains a high concentration of boric acid (up to 16,000 mg/L) that is used as a neutron absorber in pressurized water reactors. Studies from several groups have found that the corrosion rate

of aluminium in the post-LOCA solutions with high concentrations of boron is 1–2 orders of magnitude higher than in a solution with the same pH without boron [11–13]. The strong dependences on temperature and pH were also observed. The Westinghouse Electric Company [2] and Edwards et al. [14] provided the nuclear industry with aluminium release equations as a function of temperature and solution pH based on their experimental data. In our previous research [15], the effects of temperature and solution pH solution were also investigated using a rotating cylinder electrode (RCE), and the results showed similar temperature and pH dependence with Westinghouse's and Edwards's equations. However, the predictions from Edwards's equation are about 2–6 times of the values predicted by Westinghouse's equation, while our previous data showed an even higher aluminium release rates than Edwards's predictions. The large discrepancy on the magnitude of the rate might be due to the variety of the hydrodynamic conditions. For example, Edwards's tests were performed in stirred solutions compared to Westinghouse's quasi-static solutions. Bahn et al. [9] also reported that the experimental corrosion rate of aluminium alloy 1100 under a velocity of 0.03 m/s in their flow loop is approximately two times higher than the prediction of Westinghouse's equation. Flow velocity is a well-known factor that could influence the corrosion rate [16,17]. Our preliminary results [15] also indicated the effect of flow velocity on aluminium corrosion in simulated post-LOCA solution using RCE system. However, corrosion rate at the same flow velocity can be

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varied in different flow systems as demonstrated by the investigation of Cu-Ni alloy corrosion using flow through pipe channel, annular flow channel, and rotating cylinder [16]. More importantly, it was found the corrosion rate measured in various hydrodynamic systems was independent of the flow geometry when plotted as a function of mass transfer coefficient [15]. This indicates the transfer of corrosion data from one flow system to another can be achieved. Thus, in this study, the dependence of aluminium corrosion with mass transfer coefficient in post-LOCA solution will be investigated using two different hydrodynamic systems.

In addition, some other factors influencing the aluminium corrosion have been little studied [18]. Insulation leaching tests performed by Westinghouse Electric Company [2], Los Alamos National Laboratory [4], and Argonne National Laboratory [19] showed that, depending on the simulated post-LOCA conditions (e.g., temperature, pH, and buffering agents), up to ~ 300 mg/L silicon and ~ 100 mg/L calcium could be released from the fibreglass and calcium silicate insulations into the solution. Chen et al. [20] reported that the presence of 88.7 mg/L silicon caused a $300 \times$ reduction in the aluminium corrosion rate while the introduction of 50 mg/calcium also reduced the aluminium corrosion rate 50%. Howe et al. [21] reported that the concurrent corrosion of zinc in the nuclear reactor containment significantly influenced the aluminium release rate in a trisodium phosphate-buffered chemical environment after a LOCA. The post-LOCA solution also contains some trace amounts of chemical constituents such as chloride, fluoride and sulfate anions [22], which are well known to influence the aluminium passivation and the resistance to pitting corrosion [23], but has not been assessed in post-LOCA conditions.

In this work, effects of flow velocity, amount of silicon leached from insulation materials, concurrent corrosion of other metals, and trace miscellaneous anions on the aluminium corrosion and release in the post-LOCA solution were investigated using electrochemical and inductively coupled plasma techniques in well-characterized hydrodynamic conditions.

2. Experimental method

2.1. Materials and solution

The aluminium specimens used in this work were made of aluminium alloy 1100 with the following chemical composition (wt.%): Si 0.14%, Fe 0.43%, Mn 0.01%, Mg 0.02%, Cr 0.01%, Zn 0.01%, Ti 0.01%, V 0.01%, and Al balance. Rotating electrodes have been widely used for electrochemical measurement under flow conditions [24]. The aluminium washer shape specimen with an outer diameter of 15 mm and a height of 6.4 mm was mounted onto the rotating cylinder electrode (RCE) shaft for electrochemical measurement. In addition, a 5-mm diameter aluminium disk specimen was mounted on the bottom of the rotating disk electrode (RDE) shaft. Comparison of the RDE results with the RCE results allowed to investigate the aluminium corrosion in different flow systems with different flow patterns. The disk and washer specimens were successively ground with 360 and 600 grit silicon carbide papers, rinsed with deionized water and degreased in acetone. Since the presence of native oxide layer on aluminium could influence the corrosion and release rates in post-LOCA conditions, the aluminium specimens were exposed to atmospheric air for two weeks at 21 °C with a relative humidity of 50% before the tests.

The insulation materials used for the leaching tests were NUKON fibreglass (composition in wt.%: Si 29.2%, Ca 5.9%, Na 11.7%, Al 1.9%, Mg 2.1%, B 1.6%, O balance) and calcium silicate (composition in wt.%: Si 15.8%, Ca 24.8%, Na 1.7%, Fe 1.5%, Mn 0.1%, Mg 0.8%, K 0.3%, P 0.1%, Ti 0.2%, O balance) [20].

In post-LOCA conditions, the pH of the sump water is raised by adding buffering agents to minimize volatilization of radioactive iodine. In this work, the sodium tetraborate buffering agent is selected because it has a lower potential for precipitate formation than

trisodium phosphate [2]. The test solution to simulate the pool chemistry in the containment building of a pressurized water reactor under a large break LOCA scenario, consists of 236.2 mmol/L boric acid and 7.5 mmol/L $\text{Na}_2\text{B}_4\text{O}_7$, yielding to a pH around 7.2 at 21 °C. The values of the solution conductivity are 8.0 $\mu\text{S}/\text{cm}$ at 21 °C and 8.4 $\mu\text{S}/\text{cm}$ at 55 °C respectively, as measured by OAKTON PCSTestr 35.

After the LOCA, the solution temperature can be up to 130 °C initially [2] and then gradually decreases to room temperature in about 30 days or even longer. The temperature effect has been investigated in our previous study [15] and by other groups [2,14]. In this work, a moderate temperature, 55 °C, is selected to investigate the effect of mass transfer, Si inhibition, and other factors.

2.2. Glass cell tests

Experiments were conducted utilizing a typical three-electrode electrochemical system in a glass cell. The glass cell was filled with 1 L of test solution. An RDE or RCE with the Pine Research Instruments modulated speed rotator (0–4000 rpm) was used as the working electrode. A graphite rod seated inside a fritted glass tube was used as the counter electrode. An Ag/AgCl reference electrode was connected with a Luggin tube and inserted into the test solution in the cell, thus working at the test temperature rather than the room temperature. The chloride contamination from the reference electrode into the test solution, although we believe to be very minor in consideration of the short test duration and low seepage rate via the ceramic frit, should be prevented especially for the tests aiming at investigating the variability of corrosion caused by trace Cl^- additive. The chloride contamination was solved by using a double junction reference electrode which contains an inner filling solution made of 4 M KCl with AgCl and an outer filling solution composed of 1 M KNO_3 . Only the outer fill solution was in contact with the test solution via a ceramic frit, avoiding the chloride contamination. The contamination of the oxidising agent nitrate could also have an influence but again it should be limited considering the small quantity. The potential of the used reference electrode was calibrated periodically by an uncontaminated single junction Ag/AgCl (4 M KCl) reference electrode and referred to that value. The potential variations of the used reference electrode from test to test were less than ± 5 mV in a randomly scattered way. The solution temperature was maintained at 55 ± 1 °C by using a thermocouple and a controllable hot plate. The test solution was exposed to the atmosphere via a condenser, which was also used to avoid the evaporation of water. After the solution was heated to 55 °C, the working electrode was immersed in the solution, and then the desired rotation speed for working electrode was set. Based on our previous results [15], the dissolution of the native oxide layer and the reforming of the oxide layer in simulated LOCA solution can cause some instability of corrosion rate in the first 1–8 h, and the corrosion rate becomes stable afterward till 48 h as indicated by the measured film resistance. Hence, test duration of 48 h is selected which is also not too long to cause considerable concentration feedback effect.

All electrochemical measurements were performed using a Gamry Interface 1000 potentiostat controlled by the Gamry Framework software. The electrochemical impedance spectroscopy (EIS) measurements were carried out at the open circuit potential over a frequency range from 100 kHz to 10 mHz with a 10 mV amplitude sinusoidal voltage applied as the disturbance signal. After 48 h, the potentiodynamic polarisation curves were measured starting from a potential slightly more negative than the open circuit potential, and proceeding in the anodic direction at a potential sweep rate of 0.5 mV/s. As the simulated solution is dilute, the electrolyte resistances in the cell were estimated by the EIS measurement which are $\sim 54 \Omega$ for RCE (surface area 3.016 cm^2) and $\sim 620 \Omega$ for RDE (surface area 0.196 cm^2). Hence the IR compensation must be performed for the electrochemical measurement. During all polarisation measurements, the applied potentials were automatically corrected by the IR drop compensation function in

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