CORROSION AND CORROSION PROTECTION FOR REACTOR PRESSURE VESSELS / PRIMARY CONTAINMENT VESSELS IN FUKUSHIMA DAIICHI UNIT 1-3 NUCLEAR POWER PLANT

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ABSTRACT

Fukushima Daiichi nuclear power plants (1F) were damaged by unprecedented severe accident in the great east Japan earthquake on 11th, March, 2011, and seawater had been injected as an emergency countermeasure for the core cooling. Although, the RPV and PCV were not supposed to be exposed to diluted seawater, the RPV and PCV have been exposed to such environment. Therefore, corrosion caused by seawater has become an important issue. Immersion corrosion tests were performed for low alloy steel of RPV material and carbon steel of PCV material in simulated 1F cooling water environment. As a result, the mass loss by corrosion was reduced with decreasing temperature and chloride ion concentration. Moreover, the effects of nitrogen deaeration on corrosion protection were remarkable. In addition, the preliminary attempt evaluation of structural integrity of RPV and PCV were performed in considering plate thickness reduction based on corrosion test data and load condition based on earthquake response analysis results. It has been confirmed that primary stresses for RPV and PCV equipment satisfied with allowable values until at least 15 years after the accident.

Keywords: corrosion, low alloy steel; carbon steel; general corrosion; localized corrosion; integrity assessment

1. INTRODUCTION

Because of the great east Japan earthquake on 11th, March, 2011, Fukushima Daiichi nuclear power plants (1F) were damaged by unprecedented severe accident which included station blackout and core cooling function decline. As an emergency countermeasure for core cooling, the seawater injection had been performed. Then, temperature and chloride ion concentration have been decreasing after injection of freshwater and purified water. Although, the RPV and PCV were not supposed to be exposed to diluted seawater, the RPV and PCV have been exposed to such environment. Since such conditions are considered to be continued for more than 15 years before fuel removal, it is required that the structural integrity of the RPV and PCV should be ensured.
There are many reports about the corrosion behavior of low alloy steel and carbon steel in neutral environment including chloride ion such as seawater. Kado and Watanabe have reported the influence of temperature on the corrosion of steel in seawater containing dissolved oxygen [1]. According to this report, corrosion rate increases with temperature up to about 353 K in an open container, but corrosion rate is going to decrease to a very low value near the boiling point. This may be due to increase in reaction rate with increasing temperature and significant decrease in oxygen solubility at a time. Whitman et al. reported the effects of pH and temperature on the corrosion of carbon steel in water [2]. Corrosion rate of carbon steel shows almost constant in the range of pH4-10. Corrosion rate of carbon steel is increased in acidic range, and is decreased in alkaline range due to passivation. In addition, there is a tendency that the corrosion rate increases with temperature when compared at the same pH. Miyasaka reported the influence of NaCl concentration on the corrosion of cast iron [3]. In this paper, the influence of NaCl concentration on the corrosion rate of carbon steel is small. Honda et al. reported the effect of dissolved oxygen concentration on the average corrosion rate of carbon steel. The corrosion rate is increased with dissolved oxygen concentration [4]. Thus, the individual factors to affect corrosion of steels have been studied to some extent and may be roughly used for corrosion evaluation of components made of steels. However, since the corrosion data have large variation and synergetic effects of such factors on corrosion behaviors of steels are not well known, it is difficult to evaluate corrosion loss of the RPV and PCV in 1F specific condition with sufficient accuracy under the actual condition from the present knowledge. Therefore, in order to evaluate the structural integrity of the RPV and PCV in consideration of corrosion loss in the core cooling water environment in 1F, it is necessary to evaluate the corrosion behaviors of related materials in a simulated actual environment.

The purposes of this study are to investigate corrosion behaviors of low alloy steel and carbon steel in the 1F cooling water simulated environment. In addition, preliminary attempt evaluation of structural integrity of the RPV and PCV equipment in 1F is performed in consideration of corrosion behavior of these materials.

2. EXPERIMENTAL

2.1 Survey of Plant Data and Test Condition

To determine the corrosion test conditions, the plant data of 1F unit 1-3 (temperature of the RPV / PCV, events, etc.) were surveyed on the basis of published information [5-6]. Figure 1 shows temperature change of the RPV of unit 3 based on temperature measurement data [5-6]. In 1F unit 3, the temperature of the RPV had increased to a maximum value of about 623 K (350 °C) from the results of Modular Accident Analysis Program (MAAP) analysis [5]. The temperature behavior for 1-3 months after the accident was unstable, and then it was gradually decreased due to the starting of the circulation cooling system. The period of 373 K or more (>100 °C) was continued for about 6 months. After that, the temperature has become 373 K or less (<100 °C). The period of 323-373 K (50-100 °C) was continued for about 12 months, and then the temperature was had maintained 323 K or less (<50 °C).

The seawater injection was performed for core cooling during about two weeks after the accident. Then, it had been switched to freshwater. Further, the use of filtered water and treated water by water treatment facilities, nitrogen gas bubbling to water source tank, and nitrogen injection into the RPV and the PCV were started. In each unit, the borate injection (Na2B10O16 or H3BO3) had been done as appropriate in order to prevent recriticality.

Table 1 shows the test matrix that was determined based on the plant data. After the accident, the period was classified in accordance with the temperature as follows; the temperature of 373 K or more (>100 °C) was called as "short-term", the temperature of 323-373 K (50-100 °C) was called as "medium-term", and 323 K or less (<50 °C) was called as "long-term". Moreover, the chloride ion concentration from seawater which was injected immediately after the accident has been reduced by the operation of the purification system. Therefore, it was defined that Cl\textsuperscript{−} concentration and testing temperature in the medium-term and
the long-term conditions were 1000 ppm (20-fold diluted seawater) at 353 K and 100 ppm (200-fold diluted seawater) at 323 K, respectively. In addition, the influence of borate which was injected for reactivity prevention and nitrogen deaeration for the purpose of reducing dissolved oxygen concentration in the environment were examined.

2.2 Immersion Corrosion Test

In this study, low alloy steel (RPV equivalent material) and carbon steel (PCV equivalent material) were used in immersion corrosion test. Table 2 shows chemical compositions of test materials. Figure 2 shows the schematic of corrosion test specimen. The dimension of the specimen was 40 mm L×10 mm W×2 mm T, the longitudinal direction was set to be the rolling direction. Before the test, the surface of the specimen was polished #600, and ultrasonically cleaned in acetone. On all the test condition, three specimens were tested.

The schematic of the corrosion test equipment is shown in Figure 3 [7]. The specimen was placed using an attachment jig made of insulated material. The test solution was heated with a heater and held to constant temperature using a temperature controller. During the test, in order to maintain dissolved oxygen concentration in solution constant, it was introduced air or nitrogen gas into the test equipment. The gas mass flow rate was controlled at about 10 mL/min.

The artificial sea water (YASHIMA PURE CHEMICALS CO., LTD., Aquamarine) diluted with distilled water was used for the test solution. Na2B10O16 or H3BO3 was used in borate addition conditions. In the case of Na2B10O16, it was added so that the boron concentration could serve as 1200 ppm. In the case of H3BO3, it was added to a pH of 5. The ratio of test solution volume to specimen area was set to be about 40 mL/cm².

The mass of the corrosion test specimen was measured before and after the immersion corrosion test. After the test, the surfaces of the specimens were covered with corrosion products. Thus, in order to estimate net mass loss, the mass of the specimen was measured after removing corrosion products by 5 % hydrochloric acid with inhibitor.

3. RESULTS AND DISCUSSION

3.1 Corrosion Behavior of Low Alloy Steel and Carbon Steel

The results of the surface observation on the low alloy steel and carbon steel specimens in the medium-term condition (1000 ppm Cl⁻, 353 K) for before test and after 500 h are shown in Figure 4 [7]. As shown in Figure 4 (c) and (d), the entire specimen surface of each low alloy steel and carbon steel was covered with corrosion products. The difference in the surface condition of each material was little. It was considered that corrosion form of low alloy steel and carbon steel was general corrosion. Figure 5 shows the relationship between mass loss and testing time in the medium-term condition (1000 ppm Cl⁻, 353 K) and the long-term condition (100 ppm Cl⁻, 323 K). The mass loss due to general corrosion of low alloy steel and carbon steel was increased with time. In addition, the mass loss of long-term condition was reduced when compared to the medium-term condition. This tendency was consistent with the results of Kado and Watanabe [1], and Miyasaka [3] as mentioned above. Therefore, the reduction of corrosion rate is expected by mainly performing the cooling of the RPV and PCV in 1F. Although 1F is an irradiated environment, the immersion corrosion tests in this study were carried out on non-irradiated condition. Nakano et al. reported the weight loss in the steels with a 200 Gy/h dose rate were comparable with those without irradiation [8]. In addition, the result of dose measurement inside of PCV at 1F-2 was up to 72.9 Gy/h [9]. The influence of radiation on the corrosion behavior of low alloy steel and carbon steel is mainly attributed to generation of the hydrogen peroxide accompanying the radiolytic decomposition of water. Since the test solution was aerated, it is considered that corrosion behavior of low alloy steel and carbon steel under non-irradiation environment is similar to that under irradiation environment.
For recriticality prevention, injection of borate such as Na₂B₁₀O₁₆ and H₃BO₃ was performed after the accident in 1F. Therefore, in order to evaluate the influence of borate on corrosion behavior, corrosion tests with borate were performed under the medium-term condition. The influence of borate on the corrosion behavior of low alloy steel and carbon steel is shown in Figure 6 [7]. As compared with the medium-term condition without borate, the mass loss increased about 3-4 times with H₃BO₃ addition, and it was considered that general corrosion occurred from the surface morphology of specimen. On the other hand, when Na₂B₁₀O₁₆ was added, the mass loss of low alloy steel and carbon steel decreased about one-half. From the result of surface observation, it was considered that the specimen surface was passivated and localized corrosion was occurred. Thus, it was considered that the reduction of mass loss with Na₂B₁₀O₁₆ addition due to surface passivation. In the neutral borate solution, it is known that carbon steel is passivated easily. Ohba et al. reported that passive region was observed from the result of anodic polarization measurement in boric-borate buffer solution of pH 8.45 [10]. Nishimura and Sato reported that boron was presented in the precipitation film which formed at neutral pH borate solution, and the passive film exhibited anion permeability [11]. Also, Tokunaga reported that the precipitation film consisted of complex of ferric iron such as Fe(OH)(BO₂)₂ and FeOHBO₄ [12]. Generally, it is known that breakdown of passive film caused by chloride ion occurred easily in the case of the anion permeable film rather than the cation permeable film [12]. Therefore, while corrosion prevention by passivation is expected by the addition of Na₂B₁₀O₁₆, there is a possibility that localized corrosion occurs.

The surface photograph of carbon steel after the corrosion test in the long-term condition (100 ppm Cl⁻, 323 K) with nitrogen deaeration is shown in Figure 7 [7]. The specimens surface was changed color to light brown. It was considered that the specimens were covered with thin surface film. The time dependence of the mass loss with and without nitrogen deaeration is shown in Figure 8 [7]. As compared with the conditions without nitrogen deaeration (aerated long-term condition), the significant decrease in mass loss was observed in each test materials in the case of nitrogen gas deaeration. Because measurement results of dissolved oxygen concentration with nitrogen deaeration were 0.1 to 0.3 ppm, it is considered that this result has been attributed to cathodic reaction rate which was decreased remarkably. For this reason, it seems that nitrogen deaeration is effective in corrosion suppression.

3.2 Estimation of plate thickness reduction and Preliminary Attempt Evaluation of Structural Integrity

Based on the corrosion test results and literature, the thickness reduction in each period after the accident was estimated. In this paper, the scale growth model of high-temperature oxidation and corrosion test data obtained in high temperature environment including chloride ion [13] was used for estimate of the short-term plate thickness reduction. In the case of the medium- and long-term conditions, the corrosion test results of this study were used. Since the injection of borate was temporary, the influence of the borate was not considered. In actual plant, it is expected that dissolved oxygen in refined water inside RPV and PCV may be decreased by nitrogen bubbling to injection water and nitrogen sealed RPV and PCV, so that the case of deaeration with nitrogen was considered for one of the corrosion protection countermeasures. In generally, because the corrosion behavior of the carbon steel in natural water environment is diffusion limited of dissolved oxygen through the surface film, the corrosion progress rate model with parabolic law shown in equation (1) is used [14].

\[ (\bar{a} + kr_0)^2 = 2kr_0^2t + k^2r_0^2 \]  

(1)

Here, \( r_0 \) is the initial rate (mm / year), \( k \) is the rate constant (year), \( \bar{a} \) is the average depth of corrosion (mm), \( t \) is the time (year). In this study, the literature [13] was quoted and the value of \( k \) was set 0.80. The initial rate \( r_0 \) was determined by the method of least squares. The value of the initial rate \( r_0 \) of low alloy steel and carbon steel based on the result of up to 500 h or 2000 h is shown in Table 3. The initial rate obtained from the results of up to 2000 h was found to be slightly increased as compared to that obtained from the results up to 500 h. However, as shown in Figure 9, it was found that there is little difference in
the approximation results of using these initial rates. In addition, the initial rate was found to be smaller in order of the medium-term condition, the long-term condition without deaeration, and long-term conditions with deaeration in each low alloy steel and carbon steel. Based on the corrosion rate of each period obtained from the result of up to 500 h, the plate thickness reduction in the course of the years up to 15 years in RPV and PCV was presumed. As an example, the estimated result of the plate thickness reduction in RPV of 1F unit-2 is shown in Figure 10 [7]. Estimation of plate thickness reduction at 15 years after the accident was 1.8 mm when corrosion countermeasure was not performed, and 0.7 mm when N₂ deaeration were performed in the long-term period. Subsequently, in consideration of thickness reduction, the integrity assessment of the major RPV and PCV equipment to maintain the stable condition with continual cooling were performed as preliminary attempt to estimate the structural integrity. The result of the structural integrity evaluation of with and without N₂ deaeration for 15 years 1F unit-1 is shown in Figure 11, and the results of 1F unit-2 and unit-3 are shown in Figure 12 [7]. The subject equipment for integrity was RPV support skirt, drywell shell and suppression chamber shell. The primary stress of these equipment, in consideration of the plate thickness reduction and load calculated by the earthquake response analysis for the standard seismic ground motion $S_s$ under the condition of the PCV filling with water, were evaluated below the allowable stress defined as “service condition $D_s$” in the “Technical Code for Seismic Design of Nuclear Power Plants (JEAC4601-2008)”, except for some equipment of PCV. However, the primary stress of all RPV and PCV equipment was below the ultimate tensile strength $S_u$ in the “Codes for Nuclear Power Generation Facilities; Rules on Design and Construction for Nuclear Power Plants (JSME S NC1-2005/2007)”. Moreover, the buckling of RPV support skirt was evaluated below the criterion. Therefore, the equipment of RPVs and PCVs in 1F unit 1, 2 and 3 has the potential to maintain the structural integrity in more than 15 years after the accident.

CONCLUSIONS

It is necessary to evaluate the corrosion behaviors of low alloy steel and carbon steel in simulated actual environment in 1F in order to assess the structural integrity of the RPV and PCV in consideration of corrosion loss in the diluted seawater environment in 1F specific condition. Therefore, this study evaluated the corrosion behavior of low alloy steel and carbon steel in consideration of the actual plant condition of 1F and corrosion inhibition measure applicable to 1F. The results are as follows;

(1) The corrosion form of low alloy steel and carbon steel was general corrosion. As compared with the medium-term condition (1000 ppm Cl⁻, 353 K), the mass loss of low alloy steel and carbon steel in the long-term condition (100 ppm Cl⁻, 323 K) decreased.

(2) The mass loss of low alloy steel and carbon steel increased about 3-4 times in the condition of H₂BO₃ addition, and decreased about one-half in the condition of Na₂B₁₀O₁₆ addition. Although the corrosion feature in the case of H₂BO₃ addition was general corrosion, and localized corrosion was occurred when Na₂B₁₀O₁₆ was added.

(3) As a result of performing preliminary attempt to estimate the structural integrity, in consideration of the plate thickness reduction by corrosion and the load calculated by the earthquake response analysis for the standard seismic ground motion $S_s$ under the condition of the PCV filling with water, the equipment of RPVs and PCVs in 1F unit 1, 2 and 3 has the potential to maintain the structural integrity in more than 15 years after the accident.

ACKNOWLEDGEMENT

The authors are grateful to Central Research Institute of Electric Power Industry for their provision of immersion corrosion test data obtained in a high temperature diluted seawater environment which simulated “short period” for the purpose of integrity evaluation.

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REFERENCES


[13] Central Research Institute of Electric Power Industry provided data (unpublished data)

Table 1. Immersion corrosion test condition

<table>
<thead>
<tr>
<th>No.</th>
<th>Period</th>
<th>Temperature (K)</th>
<th>Cl⁻ ion (ppm)</th>
<th>Borate</th>
<th>Corrosion protection</th>
<th>Time (h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Medium</td>
<td>353</td>
<td>1000</td>
<td>—</td>
<td>—</td>
<td>50, 100, 500, 2000</td>
</tr>
<tr>
<td>2</td>
<td>Medium</td>
<td>353</td>
<td>1000</td>
<td>Na₂B₁₀O₁₆ (B: 1200 ppm)</td>
<td>—</td>
<td>50, 100, 500</td>
</tr>
<tr>
<td>3</td>
<td>Medium</td>
<td>353</td>
<td>1000</td>
<td>H₃BO₃ (pH5)</td>
<td>—</td>
<td>50, 100, 500</td>
</tr>
<tr>
<td>4</td>
<td>Long</td>
<td>323</td>
<td>100</td>
<td>—</td>
<td>—</td>
<td>50, 100, 500, 2000</td>
</tr>
<tr>
<td>5</td>
<td>Long</td>
<td>323</td>
<td>100</td>
<td>—</td>
<td>N₂ deaerated</td>
<td>50, 100, 500</td>
</tr>
</tbody>
</table>

Medium: Corresponding to the period which temperature 323-373 K (50~100 °C) in 1F  
Long: Corresponding to the period which temperature 323 K or less (<50 °C) in 1F

Table 2. Chemical composition of specimens (mass %)

<table>
<thead>
<tr>
<th>Specimen</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Ni</th>
<th>Mo</th>
<th>Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low alloy steel (SQV2A*1)</td>
<td>0.17</td>
<td>0.25</td>
<td>1.4</td>
<td>0.004</td>
<td>0.011</td>
<td>0.62</td>
<td>0.53</td>
<td>0.02</td>
</tr>
<tr>
<td>Carbon steel (SGV480*2)</td>
<td>0.18</td>
<td>0.23</td>
<td>1.15</td>
<td>0.007</td>
<td>0.003</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>

*1: Japanese Industrial Standard G3120, equivalent to ASTM A 734  
*2: Japanese Industrial Standard G3118, equivalent to ASTM A 516
Table 3. Initial rate $r_0$ and rate constant $k$ in medium-term and long-term period

<table>
<thead>
<tr>
<th>Type of steel</th>
<th>Period</th>
<th>Temperature (K)</th>
<th>Cl$^-$ ion (ppm)</th>
<th>Corrosion protection</th>
<th>Test time (h)</th>
<th>Initial rate $r_0$ (mm/year)</th>
<th>Rate constant $k$ (year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low alloy steel</td>
<td>medium</td>
<td>353</td>
<td>1000</td>
<td>—</td>
<td>50, 100, 500</td>
<td>0.624</td>
<td>0.80</td>
</tr>
<tr>
<td>Carbon steel</td>
<td>medium</td>
<td>353</td>
<td>1000</td>
<td>—</td>
<td>50, 100, 500</td>
<td>0.532</td>
<td>0.80</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>50, 100, 500</td>
<td>0.594</td>
<td>0.80</td>
</tr>
<tr>
<td>Low alloy steel</td>
<td>Long</td>
<td>323</td>
<td>100</td>
<td>—</td>
<td>50, 100, 500</td>
<td>0.341</td>
<td>0.80</td>
</tr>
<tr>
<td>Carbon steel</td>
<td>Long</td>
<td>323</td>
<td>100</td>
<td>—</td>
<td>50, 100, 500</td>
<td>0.303</td>
<td>0.80</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>50, 100, 500</td>
<td>0.314</td>
<td>0.80</td>
</tr>
<tr>
<td>Low alloy steel</td>
<td>Long</td>
<td>323</td>
<td>100</td>
<td>N$_2$ deaerated</td>
<td>50, 100, 500</td>
<td>0.056</td>
<td>0.80</td>
</tr>
<tr>
<td>Carbon steel</td>
<td>Long</td>
<td>323</td>
<td>100</td>
<td>N$_2$ deaerated</td>
<td>50, 100, 500</td>
<td>0.030</td>
<td>0.80</td>
</tr>
</tbody>
</table>
Figure 1. Temperature behavior of PCV of 1F unit 3 (as an example)

Figure 2. Schematic of corrosion test specimen
Figure 3. An example of schematic of corrosion test equipment [7]
Figure 4. Result of appearance observation before and after immersion corrosion test in 1000 ppm Cl\textsuperscript{-}, 353 K (medium-term) [7]
Figure 5. Immersion corrosion test results of low alloy steel and carbon steel in medium-term and long-term condition
Figure 6. Influence of borate on the corrosion behavior of low alloy steel and carbon steel [7]
Figure 7. Result of appearance observation after corrosion test (500 h) in 100 ppm Cl\(^-\), 323 K (long-term) with N\(_2\) deaeration

Figure 8. Effect of nitrogen deaeration on the corrosion behavior of low alloy steel and carbon steel [7]
Figure 9 Results of parabolic law approximation of immersion corrosion test data.

(a) Medium term condition

(b) Long term condition
Figure 10 Estimation result of reduction of plate thickness with and without inhibition (RPV of 1F unit-2 as an example) [7]
1. In consideration of thermal history in short-term, the value figured with 0.667 times the value of $S_u$ provided by “Codes for Nuclear Power Generation Facilities; Rules on Design and Construction for Nuclear Power Plants (JSME S NC1-2005/2007)”

2. Left part of buckling inequality $= \alpha \frac{B(P/A)}{f_c} + \alpha \frac{B(M/Z)}{f_b}$

3. In consideration of thermal history in short-term, the value figured with 0.667 times the value of $S_u$ and $S_y$ provided by “Codes for Nuclear Power Generation Facilities; Rules on Design and Construction for Nuclear Power Plants (JSME S NC1-2005/2007)”

Figure 11. Result of structural integrity of 1F unit-1 [7]
1: In consideration of thermal history in short-term, the value figured with 0.667 times the Su value provided by “Codes for Nuclear Power Generation Facilities; Rules on Design and Construction for Nuclear Power Plants (JSME S NC1-2005/2007)”

2: Left part of buckling inequality = $\alpha_b(P/A)/fc + \alpha_d(M/Z)/fb$

3: In consideration of thermal history in short-term, the value figured 0.667 times the Su value and Sy value provided by “Codes for Nuclear Power Generation Facilities; Rules on Design and Construction for Nuclear Power Plants (JSME S NC1-2005/2007)”

4: The Su value.

Figure 12. Result of structural integrity of 1F unit-2 and unit-3 [7]