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A Study on Ageing of a Canister during Long-term Storage: Evaluation of the Influence of Temperature Difference between Canister Surface and Environment on Corrosion

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Abstract

From the viewpoint of flexibility in spent fuel management, it is important to provide options of dry storage technology. The Central Research Institute of Electric Power Industry has been developing a concrete cask technology specifically for application in Japan. The most concerning issue in Japanese application is the stress corrosion cracking (SCC) of a canister since the dry storage facilities are usually located near the coasts in Japan.

For the evaluation of the SCC of canisters, it is important to collect data such as the initiation of corrosion and SCC, pitting depth, and crack growth rate because it is difficult to understand SCC simply by theoretical procedures. Numerous corrosion tests have been performed worldwide not only in the nuclear power field but also in many fields.

In the default method of corrosion test, test specimens with sea salt are placed inside a chamber with constant temperature and humidity conditions, which are close to that of the canister surface. However, considering the actual conditions, the hot canister surface is exposed to different temperatures and humidities. In this study, to examine this influence of the temperature difference between the canister surface and the environment, test specimens with heater were placed in a chamber. As a reference, test specimens without heater were also placed in the chamber. The test parameters were the temperature and humidity in the chamber and the surface temperature of the test specimens. The test concept was basically to compare the corrosion area between the two test conditions. The result for the test with a heater of 50°C in a chamber of 30°C, 100%RH was equivalent to that for the test without a heater and chamber conditions of 50 °C, 35%RH. The test specimens were made of type 304L stainless steel. The test duration was about 7900 h. During the tests, the test specimens were taken out from the chamber at certain intervals, and their photos were taken. The photos were binarized, and the corrosion area was obtained. It was found that there was no large difference between the test results of specimens with and without heater. Furthermore, after completing the test, the pitting depth was measured.

Introduction

Considering the uncertainties in the roles of nuclear power generation in Japan, the necessity of introducing interim storage facilities for spent nuclear fuel at or away from reactor sites is increasing. Although the use of metal casks for spent fuel storage has been practically employed to date, the utilization of an alternative storage method such as the concrete cask is also needed from the perspective of cask procurement risk management and economic benefit. Since Japan is an island nation, the corrosion of multipurpose canisters in the concrete cask storage system should be evaluated and tackled [1-5]. In general, general corrosion and pitting first occur, and then chloride-induced stress corrosion cracking (SCC) occurs at the bottom of the pits. From viewpoint of containment function, SCC is the most severe phenomena because its propagation rate is greater than those of pitting and general corrosion. In the past, we performed SCC tests and obtained the corresponding data. In this study, we focus on pitting corrosion as well as on the corrosion test condition. In the default method of corrosion test using constant temperature and a humidity chamber, the test specimen with corrosive substances is placed into the chamber until corrosion occurs, and in this method, the test specimen and environment have the same temperature. However, in the actual condition of the canister, the temperature of the canister surface is higher than that of environment because the canister is heated by the decay heat of the spent nuclear fuel. To evaluate the influence of temperature difference between the test specimen and the environment on the corrosion condition, a test specimen heated by an electrical heater is placed into the chamber.

The data obtained in our study were used to evaluate the containment function of multipurpose canisters during long-term interim storage to promote the concrete cask system in Japan.

Corrosion Test Method

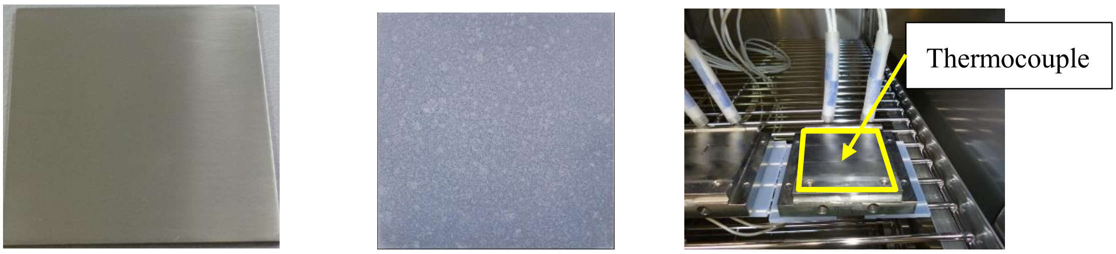
Corrosion Test Method

To evaluate the influence of temperature difference between the test specimen and the environment on the corrosion condition, we performed a corrosion test. We used two types of test specimens; the first was a square plate (75*75 mm, 2 mm thickness) made of SUS304L material (Fig. 1), and the second was a test specimen made of SUS304L material subjected to constant-load using a spring (Fig. 2). The test specimens were cut from the non-welded plate and welded plate whose surface was grinding, taking the actual condition into account. Artificial seawater was intermittently supplied to the surface of the test specimen using a nozzle that could spray fine particles. Heaters for the plate test specimen and constant-load specimen were prepared. A thermocouple was pressed against the back side of the test specimen for temperature control. The test specimens were placed into a constant temperature and humidity chamber (Fig. 3). Six constant chambers were used for the test.

Corrosion Test Conditions

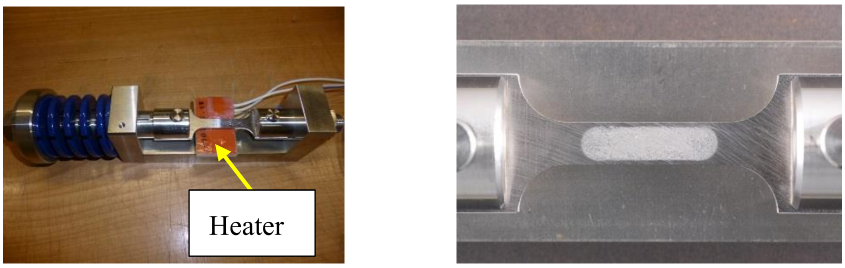
Table 1 presents the test conditions. The test temperature and relative humidity were determined by considering the actual environment based on the domestic climate observation data. As the environment was conservative, we assumed 30°C and 100% relative humidity (RH). In this condition, absolute humidity was 30.4 g/m^3. When air of 30°C and 100%RH went into the concrete cask, the temperature and relative humidity of the air changed because of the heat from the canister surface. We chose 50°C, 60°C, and 70°C as the chamber temperatures. We excluded the condition of over 70°C, because no corrosion occurs when RH is less than 15%.

Figure 4 shows the schematic view of a concrete cask, where T0 and H0 are temperature and RH of canister surface, respectively, and T1 and H1 are temperature and RH of cooling air, respectively. In the case of simulating this condition with the general test method, test specimens were placed into the chamber of T0, H0. Applying more actual conditions to the test method, the test specimens of T0 were put into the chamber of T1, H1. Considering this setting, we performed two types of tests and compared the results. Test specimens with heater were put into the chamber. The difference cases are presented in Table 1. The cases of using specimens without heater and a chamber of T0 and humidity H0 were compared with the cases of using specimens with a heater of T0 and a chamber of T1 and humidity H1. Hence, Case 1 was compared with Case 4. The deposition amount of chlorine ion was set to 5 g/m^2 as Cl for all test specimens. The applied tensile stress for constant-load specimens was about 1.25 σy. The test specimens were taken out from the chamber at certain intervals, and their photos were taken to observe the corrosion conditions (Fig. 5). The corrosion test lasted for about 7900 h.



(a) Before salt spraying (b) After salt spraying (c) Heater

Figure 1. Square plate test specimen



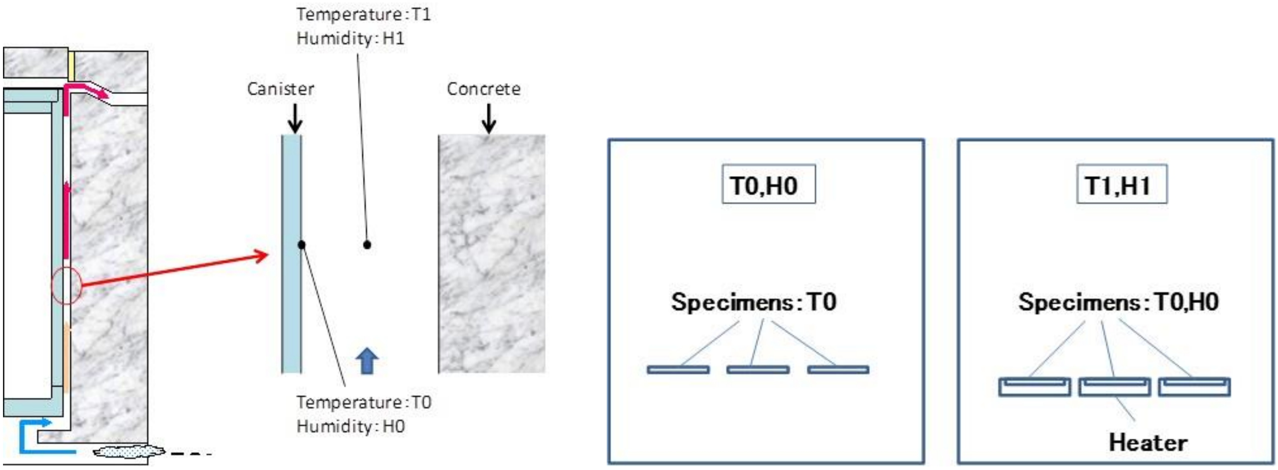
(a) Before salt spraying (b) After salt spraying

Figure 2. Constant-load test specimen

Figure 3. Test chamber

Table 1. Test Conditions

Case No	Conditions in the Chamber		Heater	Temp. of Specimen	Number of Specimen
1	30°C, 100%RH	Absolute Humidity 30.4 g/m^3	With	50°C	Square Plate Base Metal: 3 Weld Part: 2 Constant Load Weld Part: 2
2	30°C, 100%RH		With	60°C	
3	30°C, 100%RH		With	70°C	
4	50°C, 35%RH		Without	—	
5	60°C, 25%RH		Without	—	
6	70°C, 15%RH		Without	—	



(a) Schematic view of concrete cask (b) Test conditions in the chamber

Figure 4. Outline of test conditions

(a) Specimens with heater (Case 1, 2, 3) (b) Specimens without heater (Case 4, 5, 6)

Figure 5. Test specimens in the chamber

Corrosion Test Results

General Corrosion Area

The general corrosion areas of the test specimens in the test chamber were changed with time. The test specimens taken out from the chamber were photographed. Figure 6 shows the photos of Cases 1 and 4 at the end of the tests. The corrosion conditions were similar between Cases 1 and 4. To calculate the corrosion area, the photos were binarized. The corrosion area was obtained by image processing. The corrosion conditions of test specimens with and without a heater were compared (Fig. 7).

Figure 8 illustrates the comparison result of the corrosion area ratio variation with time between Cases 1 and 4. The data are the averaged value of three specimens for base metal, and two for weld part. The lines of Case 1 are close to those of Case 4. The corrosion area of the weld part was bigger than that of the base metal. For Cases 3 and 5 (Fig. 10), the four lines are close to each other. Compared with Cases 1 and 4 (Fig. 8), the corrosion area was very small. Fig. 9 (Cases 2 and 5) shows a different tendency from Figs.8 and 10. The corrosion area of Case 5 was very small compared with those of

Cases 2, 3, 6. The reason for this is difficult to explain. One of the possibilities is that the conditions in the chamber were not well controlled.

Figure 11 illustrates the influence of the temperature and humidity in the chamber on the general corrosion. The corrosion areas for the cases with lower temperatures and higher relative humidities were larger than those for the cases with higher temperatures and lower relative humidities.

Concerning the constant-load specimens, no SCC was observed. The results of the corrosion area will be evaluated in future.

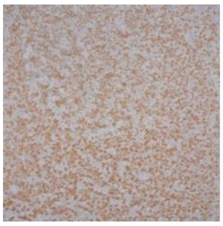
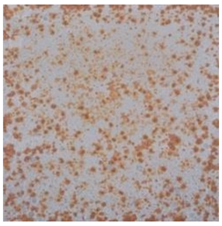
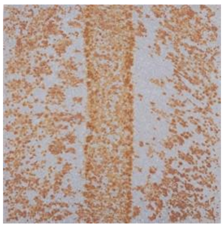
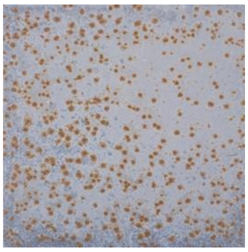
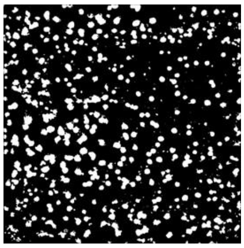
Case 1		**Case 4**	
Chamber: 30℃, 100%RH **With Heater: 50℃**		**Chamber: 50℃, 35%RH** **Without Heater**	
Base Metal	**Weld Metal**	**Base Metal**	**Weld Metal**
			

Figure 6. The photos of test specimen (7900 h)



(a) Photo



(b) Binarized data

Figure 7. Example of binarization

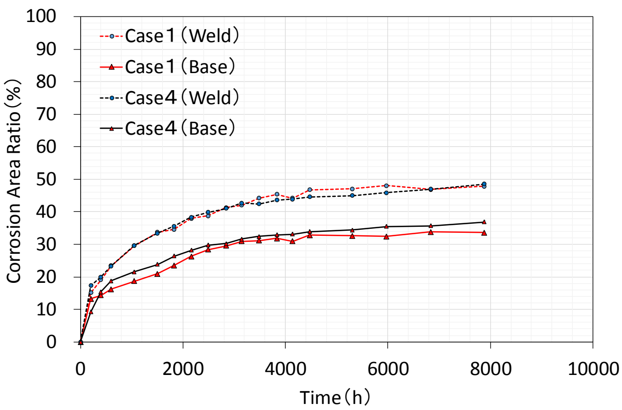


Figure 8. Test results of Cases 1 and 4

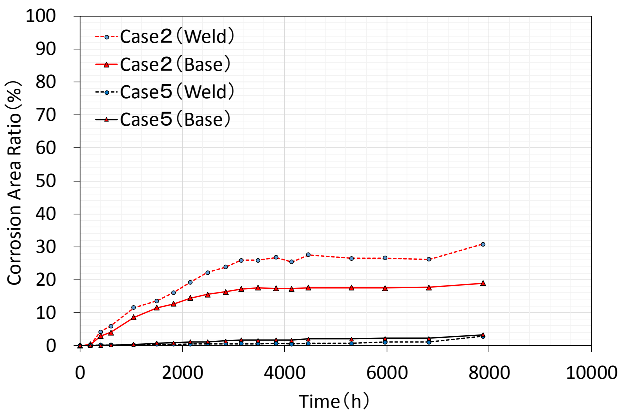


Figure 9. Test results of Cases 2 and 5

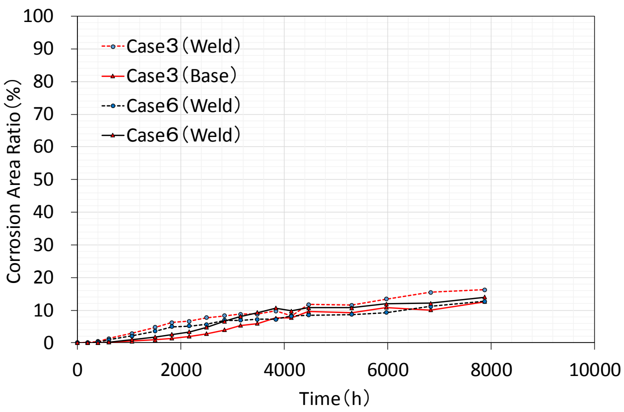


Figure 10. Test results of Cases 3 and 6

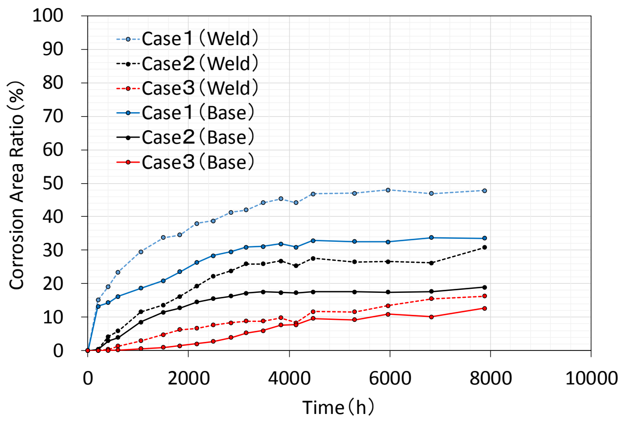


Figure 11. Test results of Cases 1, 2, and 3

Pitting Depth

After conducting the corrosion area measurements, all test specimens were taken out from the chamber, and the corrosion products were removed by chemical liquids, i.e., ammonium hydrogen citrate and nitric acid (Fig. 12). The pitting on the test specimen was observed, and the pitting depth was measured using 2D laser scanning microscope (Fig. 13).

Figure 14 shows the measurement results of the pitting depths of the square plate test specimens. Except for one irregular data, the pitting depths were all below 200 μm. The pitting depths at lower humidities tended to be greater than those at higher humidities. The numbers of pits at lower humidities also tended to be larger than those at higher humidities. Furthermore, the numbers of pits for Cases 1, 2, and 3 were larger than those of Cases 4, 5, and 6. There is a possibility that the pitting condition is different between the case with heater and that without heater.

Using these data, we will evaluate the pitting depth of canister after storage by the extreme value statistical method.

Figure 12. Removal of corrosion products

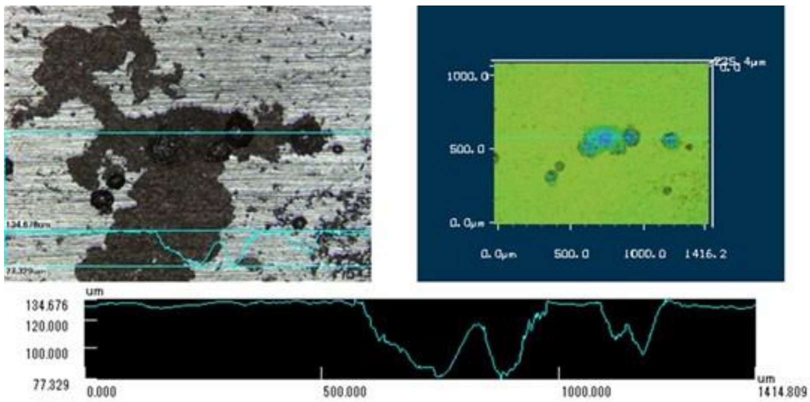


Figure 13. Measurement of pitting depth

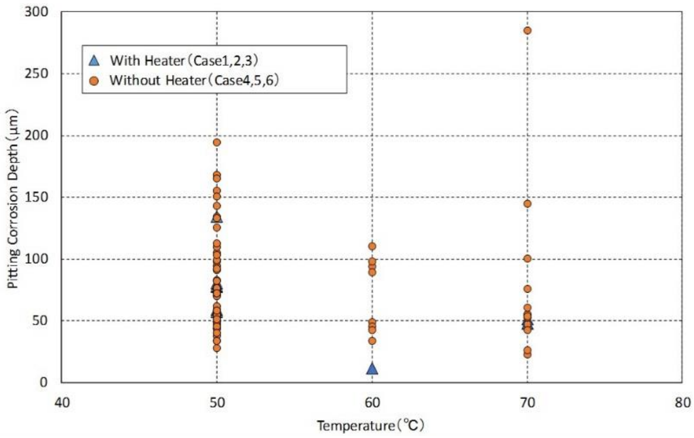


Figure 14. Pitting depth measurement result

Conclusions

To evaluate the influence of temperature difference between the test specimen and the environment on the corrosion condition, we performed a corrosion test using test specimens with and without a heater. From the tests, the following results were obtained:

Concerning the general corrosion, the result for the test using specimens without heater and a chamber of constant temperature (T0) and humidity (H0) was similar to that using specimens with a heater of temperature T0 and a chamber of constant temperature (T1) and humidity (H1).

T0, H0: canister surface temperature and humidity

T1, H1: cooling air temperature and humidity

Moreover, the pitting depths at lower humidities tended to be greater than those at higher humidities. Likewise, the number of pits at lower humidities tended to be greater than those at higher humidities. There is a possibility that pitting condition was different between the case with heater and that without heater.

The results obtained in this study can be utilized for the evaluation of canister integrity during long-term storage.

References

[1] Shirai K. et al., Study on Interim Storage of Spent Nuclear Fuel by Concrete Cask for Practical Use, – Feasibility Study on Prevention of Chloride Induced Stress Corrosion Cracking for Type304L Stainless Steel Canister –, CRIEPI Report N10035, 2010, (In Japanese)

[2] Wataru M., et al., SCC Tests of the canister for spent nuclear fuel storage using full scale lid model, Proceedings of PATRAM2016, 2016

[3] Goto M., et al., Study on Interim Storage of Spent Nuclear Fuel by Concrete Cask for Practical Use, – Feasibility Study on Prevention of Chloride Induced Stress Corrosion Cracking for Type304L, 316L Stainless Steel Canister –, CRIEPI Report N12023, 2013, (In Japanese)

[4] Wataru M., et al., Study on Commercial Realization of Concrete Cask for Interim Storage of Spent

Nuclear Fuel, – Proposal and Verification of Code Case on SCC Countermeasure of Canister –, CRIEPI Report N15014, 2016, (In Japanese)
[5] Wataru M., et al., Studies on the Long-term Integrity of the Canister for the Dry Storage of Spent Nuclear Fuel, Proceedings of RAMTRANSPORT2018, 2018