



The Adhesion Characteristics of Protective Coating Materials for the Containment Structure in Nuclear Power Plants

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ABSTRACT

Protective coating materials used in the containment structures should be durable for the designed 30 to 40 year lifetime of a nuclear power plant. At the present, these materials have not yet been developed. Therefore it is very important to keep the durability of the protective coating materials through persistent maintenance, and in order to achieve this, understanding the adhesion characteristics of the coating materials is of utmost importance. Therefore, this study attempts to find any methods for durability maintenance of these protective coating materials. To accomplish these aims, this study applied an experimental deterioration environment condition relevant to Loss of Coolant Accident (LOCA) and Main Steam Line Break (MSLB), categorized as of Design Basis Accident (DBA), onto steel liner plate specimens covered with protective coating materials. Adhesion tests were performed on these deteriorated coating materials to characterize the physical properties and through these tests, the quantitative adhesion characteristics according to the history of deterioration environment were found.

KEY WORDS: protective coating, containment, nuclear, adhesion, deterioration, environment, LOCA, MSLB, design basis accident (DBA), liner plate, adhesion tests, zinc primer, epoxy paint, adhesive strength, accelerated deterioration test.

INTRODUCTION

The steel liner plate is located in the inner side of the containment structure to contain radioactive substances. The surface of the steel liner plate is applied with protective coatings to control corrosion and radioactive contamination levels, as well as to protect its surfaces from wear. The coating materials applied should be capable of withstanding high temperature, pressure, and radioactivity caused at the simulated design basis accident such as LOCA and MSLB. For that reason, the protective coating materials are classified into the safety related item and produced under the strict quality control.

When Reg. Guide 1.54, related to protective coating materials of nuclear power plant was issued in 1973, it was expected that the protective coating materials that met this guide would keep its function for the designed durability period. But, examining the operation history of N.P.P.s, we can find many types of deteriorations such as unexpected cracks and delamination from substrata in the qualified protective coating materials.

Ideally, protective coating materials should be durable for the designed lifetime of a nuclear power plant, which is generally around 30 to 40 years. At this present, these materials have not yet been developed, so therefore it is very important to keep the durability of the protective coating materials through persistent maintenance. In order to achieve this, understanding the adhesion characteristics of the coating materials is of utmost importance.

But, it is not easy to grasp the state of deterioration only by measuring material properties, because the existing coating materials have a big deviation according to the location of application and its conditions and circumstances. The

appropriate measures could not be taken in advance due to the lack of understanding in the characteristics of deterioration.

Therefore, we conducted the accelerated deterioration experiment in which we exposed the coating material in deteriorating environments. Physical adhesion test about the deteriorated materials were carried out to find a solution to these problems and a way to insure the quality of the coating materials. From this experiment, the quantitative adhesion characteristics according to the history of deteriorating environment were comprehended. Furthermore, methods to improve durability and quality were suggested. In conclusion, the key factors in improving the durability and quality of the coatings are selecting the most suitable coating system for its purpose and performing the initial curing thoroughly under construction.

TEST SPECIMENS PREPARATION

The test specimens used in this study were manufactured according to the ASTM D5139 standard. We conducted physical tests on adhesion, abrasion, and impact to examine the conformity of the test specimens. The test specimens were manufactured in two ways. The first group specimens were made in a construction site with poor working conditions during a humid rainy season. In contrast, the second group specimens were manufactured in the sound laboratory situation where the temperature and humidity can be controlled. The first group specimens were cured at normal temperature for 5 days, while the second group specimens were cured for 14 days at a constant temperature of 65°C according to the standard.

Manufacturing of the First Group Specimens

Coating System Code 1

Inorganic zinc of company A was used as the material for the primer, and epoxy paint of company B was used as the material for the top coat. We made the test specimen from ASTM A36 carbon steel after the pre-treatment of the coating surface considering testing conditions (radiation, DBA) according to the methods and thickness in the CP-A3 specification and the ASTM/ANSI standard. Table 1 shows the specification of coating system code 1.

Coating System Code 2

Inorganic zinc of Company B was used as the coating materials for the primer and epoxy paint for the topcoat. All coating sequences and processes of manufacturing were same as the coating system Code 1. Table 2 shows the specification of coating system code 2.

Coating System Code 3

The suggested coating system with two epoxy layers was used in this study. We used the product of Company B as the coating material for the epoxy primer and the epoxy topcoat. All coating processes were conducted as same as the coating system Code 1. Table 3 shows the specification of coating system code 3.

Manufacturing of the Second Specimens

Coating Systems Code I-1, Code I-2

Inorganic Zinc of company A was used as the material for the primer and epoxy paint of company B was used as the material for the top coat. Tables 4 and 5 show the specifications of coating system code I-1 and code I-2, respectively.

Coating System Code •

The products of Company B were used as the coating materials for the inorganic zinc primer and epoxy top coat of the coating system Code •. Table 6 shows the specification of coating system code • used in this study. Additionally, the sequences and processes of manufacturing were the same as Code •.

Accelerated Deterioration Test

To make accelerated deterioration environments, we carried out the standard experimental deterioration tests about the environmental conditions such as possible temperature, humidity, radiation that can be occurred at the LOCA and MSLB. Table 7 shows the deterioration conditions applied for the accelerated deterioration test.

Radiation Test

The radiation test to make the deterioration environment was conducted according to the standard of ANSI N5.12. Total quantity of radiation was 2×10^8 rads and irradiation dose rate was $4 \cdot 5 \times 10^5$ rads/hr.

DBA Test

We simulated the accident environment by using DBA testing facility with the E.Q-Profile by DBA scenario. Concerning DBA scenario, LOCA scenario of the Younggwang units 3 and 4 and MSLB scenario of the Uljin units 5 and 6 were used respectively.

LOCA Environment

LOCA test was performed according to the related specification (CP-A3 No: 9-191-A610) and standard (ANSI N101.2). The required testing time was 12 days and the actual used maximum temperature and pressure were 329• and 59 psig(407 kPa), respectively. Figure 1 shows the E.Q-profile used in this test.

Examining the LOCA test history curve (Figure 2) from the test results, we can confirm that the coating systems experienced the LOCA environment of design base accident because the temperature and pressure curve envelop the required E.Q-Profile according to the related specification requirements.

MSLB Environment

MSLB test was performed according to the related specification (CP-A3 No: 9-191-A291) and standard (ASTM D3911). The required maximum temperature and pressure of E.Q Profile were 360°F and 57 psig (393 kPa), respectively. It took 30 days for the test, and the test history curve is in Figure 2. Compared with E.Q-Profile, it shows that MSLB test history curve envelop the required E.Q Profile. Therefore, we can confirm that the coating systems experienced MSLB environment of design base accident.

Adhesion Test and Results

Adhesion Test

Adhesive performance of film to a substrate is a very important property because even good coating materials cannot maximize its coating potentials if adhesion is low. Especially, in the protective coating materials of containment, the most important performance is adhesion. However, the mechanism of adhesion has not become clear since it has been explained on the basis of construction experience.

Adhesion test was performed to the specimens exposed to deterioration environment with Elcometer Adhesion

Tester according to ASTM D4541. We used a small tester, because we expected that the adhesion would not be great before the test. However, the same type of large tester was used with the small one since adhesion of some parts of test specimens were more than 1000 lb/in²(70.3 •/•). Epoxy adhesive was used as a bonding agent of Dolly and we measured the adhesive strength after drying and hardening at room temperature for more than 2 days.

Comparative Analysis of Results

Influence by Irradiation

In order to understand how irradiation influences the adhesive strength of test specimen, the results of the adhesive strength measured before and after irradiation are compared in Table 8.

The adhesive strength of irradiated test specimens are improved more than that of basic test specimen before irradiation. It suggests that irradiation helps to increase the adhesive strength of the film, although it acts on protective coating materials as a deterioration factor.

It is thought that the improvement in the adhesive strengths of all the coating systems are due to irradiation's tendency to strengthen the contact between layers of steel plate and coating materials rather than damaging chemical chains in coating materials. This also suggests that we cannot regard the strength of adhesion as an indicator of deterioration degree, contrary to the existing expectation that irradiation will decrease the adhesive strength.

Influence by LOCA Environment

To understand the influence of LOCA environment on the adhesive strength of test specimen, the adhesive strengths before and after LOCA test are compared in Table 9.

The adhesion of the test specimens exposed to LOCA environment is improved after the test than before the test except the Coating System Code 2. The results counter our expectations that LOCA environment would obstruct the adhesive strength in the performance qualification of the protective coating materials. In case of Coating System Code 2, the measured adhesive strengths are less than 100 lb/in²(7.0 •/•²) since the materials that lie on the surface of the steel plates corroded. The reason of corrosion is that Code 2 specimens were manufactured in a relatively poor environment. In addition, they are made with a thick inorganic zinc layer, it seems to be severely influenced by deterioration environment.

Influence by MSLB Environment

To understand how MSLB environment influences on the adhesion of test specimens, the adhesive strengths before and after MSLB test are compared in Table 10.

All of the adhesive strengths have decreased except for Coating System Code 3, which exceeded the measurement range. The initial curing of the entire specimens helped to create sufficient adhesion although they do not have adequate margins of additional improvement. The decline in adhesion is believed to be caused by the 30 days deterioration environment period, with humidity particularly affecting on it negatively.

Influence according to Coating System

Regardless of experiment condition for manufacturing and curing, the adhesive strength of coating system Code 3 is of best performance. The normal system (Code •-1) with inorganic zinc primer and epoxy top is of the next performance, and the repair systems (Code 1(repair), Code 2(•), Code I-2) with inorganic zinc primer and epoxy top are of inferior performance.

The repair systems with poor adhesive strength have twice thicker inorganic zinc for primer than the normal system. It suggests that the thickness of inorganic zinc used for primer has a direct influence on lowering of adhesion. Therefore, it is not desirable to use inorganic zinc as a protective coating system except when this kind of system is specially used for preventing steel from corroding. If the system is used inevitably, however, we need a special construction management to make sure that the inorganic zinc is not thicker than the one in the specification. In addition, replacing the currently employed zinc primer to epoxy primer is advantageous to improve the adhesion of repair coating system. In all the coating systems, the adhesive performance of the second group specimen is much better than that of the first one, and the adhesive performance depends on the initial curing condition more than on deterioration environment. It suggests that the initial curing condition is an important factor to secure the durability of protection coating materials in construction.

Therefore, the most important factor for improvement in durability of protective coating materials and better quality is selecting the coating system most suitable for the use and to provide a working environment where the initial curing condition can be performed thoroughly.

CONCLUSION

- 1) Concerning the adhesion of coating systems, the coating system whose top and the primer coat are composed of epoxy layers has the highest adhesion, the normal coating system intermediate, and the repair system has the lowest.
- 2) The repair system with double the inorganic zinc has the lowest adhesion, indicating adhesion drop is directly related to the thickness of inorganic zinc.
- 3) Irradiation generates relaxations to the chemical bonds of the network of coating materials, but the benefits outweigh the detriments, improving the adhesion between the coating materials and steel liner plates. Therefore, both of the previously known facts that the irradiation reduces the overall adhesion of the coating materials and that the degree of deterioration depends solely on the strength of adhesion should be reconsidered.
- 4) Adhesion for all the specimens largely depends on the initial curing condition rather than the deterioration of the environment, suggesting that the initial curing condition is an important factor for the durability of protective coating materials.
- 5) Coating system with two epoxy layers, suggested in this study, resulted in providing adhesion far better than those currently used in domestic nuclear containment structures, and furthermore, show an outstanding resistance under deterioration conditions. Consequently, this study suggests replacing the current coating system with the new one.
- 6) In the future, research should be carried out in terms of the effect of the humidity upon durability and adhesion of protective coating materials. This will deliver a better and more systematical method for the evaluation of deterioration characteristics.

References

1. NRC Information Notice 97-13, "Deficient conditions associated with protective coatings at nuclear power plants", Mar. 1997
2. NRC Generic Setter 98-4, "Potential for degradation of the emergency core cooling system and the containment spray system after LOCA because of construction and protective coating deficiencies and foreign material in containment", 1998

Table 1 Specification of the coating system (Code 1)

No.	The Front Side Name of Product/ Thickness	The Back Side Name of Product/ Thickness
1st	Inorganic Zinc Primer CZ 11SG (3-5)	Inorganic Zinc Primer CZ 11SG (3-5)
2nd	Epoxy Topcoat ET 562 (3-5)	Inorganic Zinc Touch-up IZ197 (3-5)
3rd		Epoxy Topcoat ET 562 (3-5)
Total	(6-10)	(9-15)

Table 2 Specification of the coating system (Code 2)

No.	The Front Side Name of Product/ Thickness	The Back Side Name of Product/ Thickness
1st	Inorganic Zinc Primer IZ197 (3-5)	Inorganic Zinc Primer IZ197 (3-5)
2nd	Epoxy Topcoat ET 562 (3-5)	Inorganic Zinc Touch-up IZ197 (3-5)
3rd		Epoxy Topcoat ET 562 (3-5)
Total	(6-10)	(9-15)

Table 3 Spec. of the coating system (code 3)

No.	The Front Side Name of Product/ Thickness
1st	Epoxy Primer ET 562 (3-5)
2nd	Epoxy Topcoat ET 562 (3-5)
Total	(6-10)

Table 4 Spec. of the coating (Code •-1)

No.	The Front Side Name of Product/ Thickness
1st	Inorganic Zinc Primer CZ 11SG (3-5)
2nd	Epoxy Topcoat ET 562 (3-5)
Total	(6-10)

Table 5 Spec. of the coating system (Code •-2)

No.	The Back Side Name of Product/ Thickness
1st	Inorganic Zinc Primer CZ 11SG (3-5)
2nd	Inorganic Zinc Touch-up IZ197(3-5)
3rdl	Epoxy Topcoat ET 562 (3-5)
Total	(9-15)

Table 6 Spec. of the coating system (Code •)

No.	The Front Side Name of Product/ Thickness
1st	Inorganic Zinc Primer IZ197(3-5)
2nd	Inorganic Zinc Touch-up IZ197 (3-5)
3rdl	Epoxy Topcoat ET 562(3-5)
Total	(9-15)

Table 7 Comparison of deterioration environment conditions

	Deterioration condition	
	LOCA	MSLB
Maximum Tem./Duration	302°F(150•) / 10 ⁴ sec	360°F(182•) / 2×10 ² sec
Maximum Pressure/Duration	54psig(372kPa) /10 ⁴ sec	57psig(393kPa) /10 ³ sec
Total test time	12 days	30days
Other condition	Humidity condition	Humidity condition

Table 8 Comparison of adhesion test results (Irradiation)

System		Deterioration Environment	Before irradiation (lb/in ²)/(•/• ²)	After irradiation (lb/in ²)/(•/• ²)	Others
Code 1	Normal		380 / 26.7	520 / 36.6	
	Repair		140 / 98	190 / 13.4	
Code 2			230 / 16.2	240 / 16.9	
Code 3			Over 1500/105	Over 1500/105	Excess of measurement range
Code •	I-1		Over 1500/105	Over 1500/105	Excess of measurement range
	I-2		890 / 62.6	930 / 65.4	
Code •			850 / 59.8	920 / 64.7	

Table 9 Comparison of adhesion test results (LOCA)

System		Deterioration Environment		Basic test specimen (lb/in ²)/(•/• ²)	LOCA test specimen (lb/in ²)/(•/• ²)	Others
Radiation						
Code 1	Normal	None		380 / 26.7	700 / 49.2	
		Irradiation		520 / 36.6	900 / 63.3	
	Repair	None		140 / 9.8	430 / 30.2	
		Irradiation		190 / 13.4	500 / 35.2	
Code 2	None		230 / 16.2	100 / 7.0*	*Steel corrosion	
	Irradiation		240 / 16.9	100 / 7.0*	*Steel corrosion	
Code 3	None		Over 1500/105	Over 1500/105	Excess of measurement range	
	Irradiation		Over 1500/105	Over 1500/105	Excess of measurement range	

Table 10 Comparison of adhesion test results (MSLB)

System		Deterioration Environment Radiation	Basic test specimen (lb/in ²)/(./• ²)	MSLB test specimen (lb/in ²)/(./• ²)	Others
Code •	I-1	None	Over 1500/105*	680 / 47.8	*Excess of measurement range
		Irradiation	Over 1500/105*	940 / 66.1	*Excess of measurement range
	I-2	None	890 / 62.6	640 / 45.0	
		Irradiation	930 / 65.4	800 / 56.3	
Code •	None	850 / 59.8	410 / 28.8		
	Irradiation	920 / 64.7	720 / 50.6		
Code 3	None	Over 1500/105	over 1500/105	Excess of measurement range	
	Irradiation	Over 1500/105	over 1500/105	Excess of measurement range	

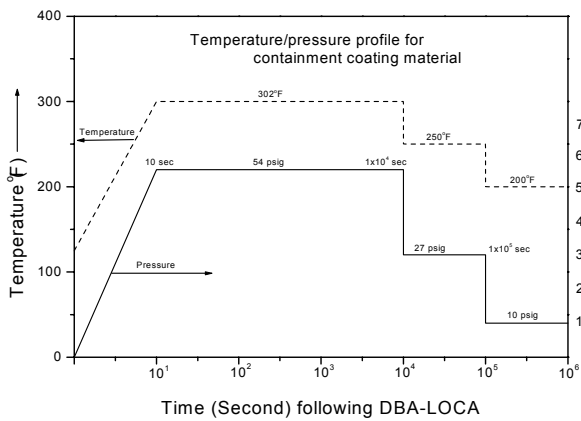


Fig. 1 E-Q-Profile for LOCA Test

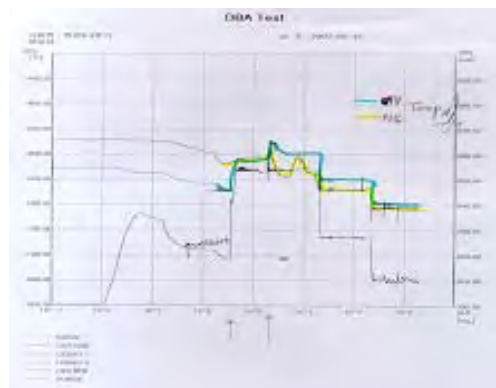


Fig. 2 Temp.-press. Curve by LOCA Test

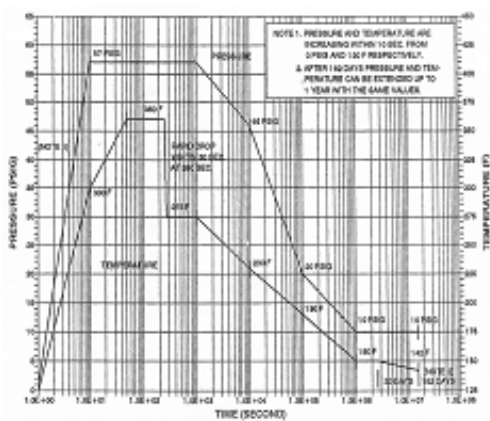


Fig 4

