



Effective Use of Sacrificial Zinc Anode as a Suitable Repair Method for Severely Damaged RC Members Due to Chloride Attack

Pinta Astuti ^{1*}, Rahmita Sari Rafdinal ², Daisuke Yamamoto ³,
Volana Andriamisaharimanana ⁴, Hidenori Hamada ⁵

¹ Department of Civil Engineering, Universitas Muhammadiyah Yogyakarta, Yogyakarta, 55183, Indonesia.

² Maintenance Engineering Group, P.S. Mitsubishi Construction Co. Ltd., Harumi Center Building 3F, 2-5-24, Tokyo, 104-8215, Japan.

³ Department of Civil and Environmental Engineering, National Institute of Technology, Oita College, Maki 1666, Oita-shi, Oita, 70-0152, Japan

⁴ Nakabohtec Corrosion Protecting Co. Ltd., 2-5-2, Shinkawa, Chuo-ku, Tokyo, 104-0033, Japan

⁵ Department of Civil and Structural Engineering, Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka, 819-0395, Japan.

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Abstract

In many cases, the repair strategy by using sacrificial anodes for cathodic protection in real RC structures requires additional zinc anodes after several years due to the decreasing protective area. This experimental study evaluates the effectiveness of time lag application of sacrificial anode cathodic protection applied to RC beam specimens that deteriorated severely due to chloride attack. In the experiment, sacrificial anodes and cathodic protection (SACP) were applied to 41-year-old RC beam specimens exposed to natural marine environments in which the embedded steel bars were significantly corroded. The repair work was performed in three stages. Instant-off and rest potential tests of steel bars were conducted periodically to demonstrate the time-dependent depolarization value. In the first stage, a polymer-modified mortar as a patch repair material was cast to replace the concrete in the middle tensile part with small sacrificial anodes embedded in the mortar. After the protective current reaches an equilibrium state, the sacrificial anodes are disconnected from the steel bars for a year, defined as the second stage. During the one year in the second stage, the steel bar in the patch repair area remained passive, without any sign of corrosion. As for the third stage, additional sacrificial anodes were installed in the existing concrete part to protect the steel in it. From one year of observation after applying sacrificial anodes to old concrete parts, the time lag SACP application of both in patch and non-patch repair parts was clarified to be effective in stopping the corrosion of steel bar in both parts until 20–30 years based on the service life prediction.

Keywords: Patch Repair; Sacrificial Zinc Anode; Service Life; Steel Corrosion; Time Lag Cathodic Protection.

1. Introduction

The corrosion of steel bars due to chloride attack is one of the most common deterioration phenomena in reinforced concrete (RC) structures, especially those exposed to marine environments. The steel bar embedded in concrete is naturally protected from corrosion by the high alkalinity of the pore solution, which activates the passivity film on its surface [1, 2]. This passivity film prevents the development of an active corrosion agent [1, 3]. However, this protection can be destroyed by chloride ions or by the acidification of the environment in the vicinity of the rebar with carbonation [3]. The penetration of chloride ions into concrete activates the corrosion of rebar by destroying the passivity film when

* Corresponding author: pinta.astuti@ft.umy.ac.id

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the chloride ion concentration at the rebar surface reaches a critical value [4, 5]. When a sufficient number of chloride ions reach the surface of the rebar, along with enough oxygen and moisture, steel corrosion may be extremely severe, resulting in concrete cracking, spalling of concrete cover, and reduction of the steel reinforcement cross-section, ultimately leading to structural failure of the member [6]. The corrosion of reinforcement adversely affects the safety and serviceability of concrete structures, hence shortening the service life [7-10].

There are several well-known electrochemical repair methods for rehabilitation of the RC structures deteriorated by chloride-induced corrosion, i.e., electrochemical chloride removal, electro-deposition, cathodic protection, corrosion inhibitor, etc. [1, 11]. Among the methods, cathodic protection is the most effective for suppressing corrosion [12, 13]. The principle of cathodic protection is the corrosion control of the reinforcement by applying a cathodic current to the reinforcement such that the reinforcement is cathodically polarized and the anodic reaction is inhibited. There are two main systems of cathodic protection, i.e., impressed current cathodic protection and sacrificial anode cathodic protection [3]. The principle of an impressed current system is the protection of the steel bar by applying electrical current from an external power source connected between an anode and the steel bar [12, 14]. The principle of the sacrificial anode system is the electrical coupling of reinforcement to a sacrificial anode. It has been reported that both the sacrificial anode cathodic protection system and the impressed current cathodic protection system are appropriate repair options that can be used in the right circumstances [1, 15]. The cathodic protection was also used to combine with the structural strengthening; combined impressed current cathodic protection-structural strengthening (ICCP-SS) [16]. This technique was able to provide effective cathodic protection as well as shear stress transfer behavior to RC structures, leading to an improvement with respect to structural durability. The ICCP-SS was also used to rehabilitate sea-sand concrete columns and the results showed that the loading capacities of the columns retrofitted by the ICCP-SS method were up to 40% greater than those of the corroded columns without any protection [17]. Impressed current cathodic protection is preferred to be implemented in wide and severe RC structures exposed to the marine environment. Hybrid cathodic protection technology using sacrificial anodes and impressed current methods was developed by researchers [18–21]. For the impressed current cathodic protection, regular monitoring in order to assess the levels of cathodic protection being afforded to the structure is required. In addition, cabling and control boxes associated with the impressed current cathodic protection are required to be strategically placed in order to avoid the risk of vandalism, especially in developing countries. Therefore, the impressed current cathodic protection system may still not be appropriate for repairing small infrastructures in local areas or developing countries where the security system is not good enough [1].

A common approach to rehabilitate the deteriorated RC structures is the patch repair method. The British Standard of Design Manual for Road and Bridge [22] recommends that the sections showing a chloride ion contamination above 0.3% by weight of cement and half-cell potential value below -350 mV should be removed. Nonetheless, the concrete replacement work on chloride contaminated structures is exceptionally arduous and expensive [23, 24]. Sacrificial anodes have been selected as a method to limit the extent of concrete replacement and to extend the service life of patch repairs in corrosion-damaged RC structures [25]. Sacrificial anode is the most valuable choice for repairing. Zinc and its alloys have been experienced mostly among the sacrificial anode materials; however, the zinc alloys is limited in term of protection capability and anode implementation has evolved to promote sustained metal activity [26]. Zinc-Aluminium hot-dip galvanized coating improves corrosion protection of steel bars [27]. Alkali-activated galvanic anodes can protect the steel rebars from corrosion for at least 12 years based on long term observation [28]. New activating mortar was recently developed to improve the sacrificial anode performance [29]. The non-rusted rebar condition is the most desirable initial condition when the sacrificial anode was applied on it in repaired concrete [30, 31]. Electrochemical incompatibility exists between repaired concrete and old concrete during the application of sacrificial anodes in repaired concrete [32]. Different measures to increase the service life of the repair system using sacrificial anodes are also suggested, such as incorporating a sacrificial embedded anode, timing of anode installation, combination with impressed current method, along with the use of surface coating or membrane as additional line defense [32]. The decision to apply for the sacrificial anode cathodic protection and the patch repair method to particular structures could be, in many cases, based on a preliminary investigation showing some high levels of chloride contamination, the possibility of steel bar corrosion, and damage appearance of the structures [33, 34].

The sacrificial anode cathodic protection system is suitable for small infrastructures and limited budget cost repair. Some countries in Southeast Asia including Indonesia have long coastlines which concrete structures located in a hot and humid climate zone tends to deteriorate from the corrosion of the reinforcement by airborne chloride [35-37]. Even the sacrificial anodes cathodic protection is considered as the most effective and widely used in controlling chloride-induced reinforcement corrosion [28, 38, 39]; however, the sacrificial anodes cathodic protection can lose its effectiveness with time due to the anode consumption during repair period [1]. The anode life is dependent on the average current output that affected by temperature, oxygen content, humidity and chloride content [40]. For the patch repaired concrete, it was reported that the sacrificial zinc anode had a profound effect on polarizing the potential of reinforcement in the substrate concrete and no significant protection in patch repair section [11]. However, this problem could be solved by adding the sacrificial anode in the patch repair section as reported by previous study that the patch repair containing zinc sacrificial anodes was rather effective in inhibiting the corrosion effect around the substrate-patch

interface [41]. In some cases, the application of sacrificial anodes was done several years, and the evaluation of anode performances resulted decreasing protection less than the protective criteria. Then, the additional anodes were installed to the concrete part surrounding the previous repair part in different time in the future. Therefore, it is necessary to assess the effect of time lag application of sacrificial anodes in patch and non-patch repair part by using real deteriorated structures that could not be found by the previous study. In this study, the time lag application of sacrificial anodes in the patch and non-patch repaired part were tested on severely damaged 44-year-old RC beam specimens exposed to natural marine environments. The result of this research will provide information for better understanding of sacrificial anodes cathodic protection performance in term of the application method in different time. This will improve the design practice of sacrificial anodes cathodic protection for specifying appropriate installation time of anodes to attain high efficiency of corrosion protection.

2. Experimental Programs

The flowchart of research methodology including preliminary investigation, repair design, repair stage 1, repair stage II, repair stage III, data analysis, and prediction of service life was presented in Figure 1.

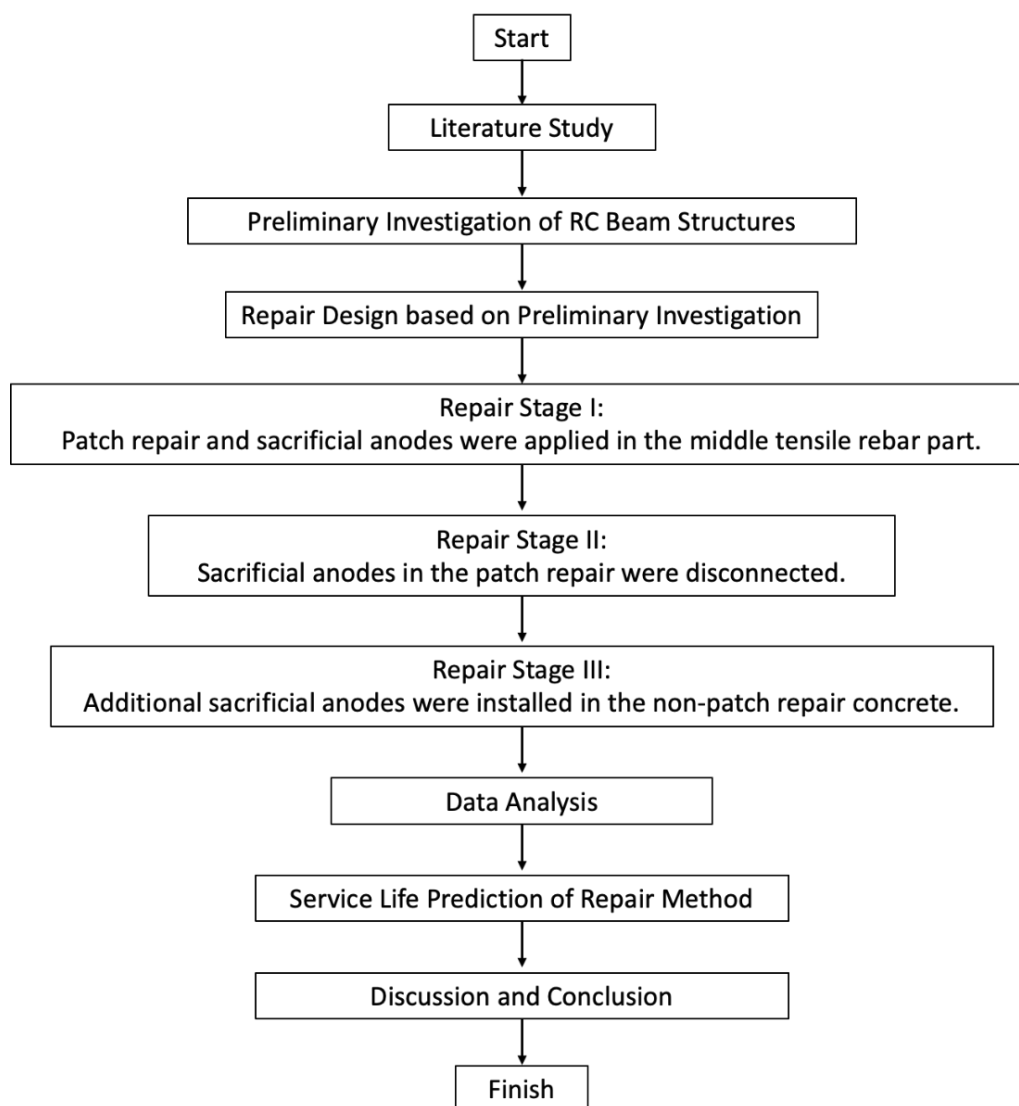


Figure 1. Flowchart of research methodology

2.1. RC Beam Specimen Detail

RC beam specimens with a span length of 2,400 mm, a cross-sectional area of 200×300 mm and 150×300 mm were used and denoted as RC-1 and RC-2, respectively. The concrete cover thickness is 50 mm for RC-1 and 30 mm for RC-2. Deformed steel bars with a diameter of 13 mm and 363 MPa in tensile strength were embedded as tensile rebars in the beam. Two round steel bars of 6 mm in diameter as compressive bars and stirrups with a spacing of 100 mm were embedded. The cross-section and reinforcing bar layout are depicted in Figure 2.

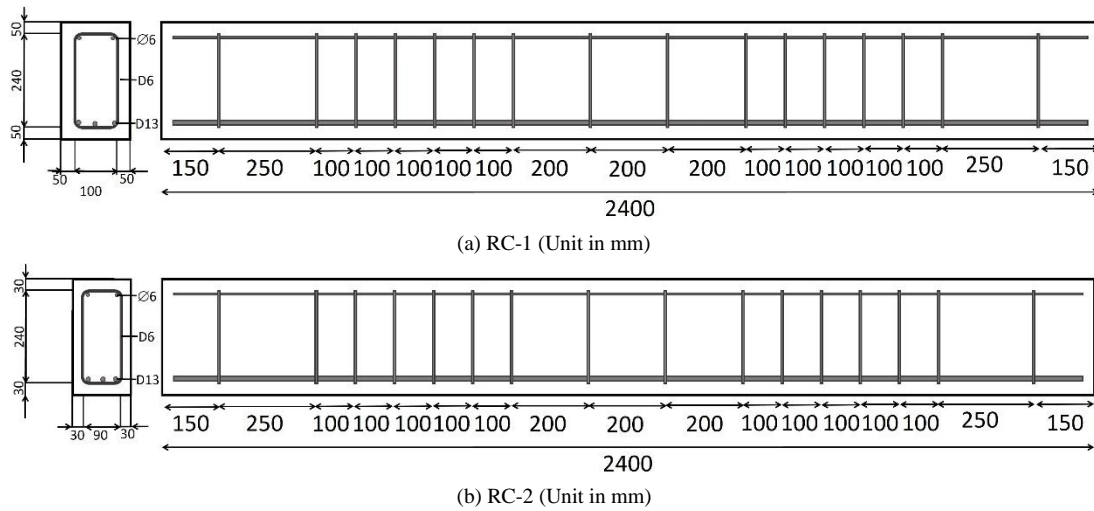


Figure 2. Cross-section and reinforcing bar layout of RC beams (a) RC-1 and (b) RC-2

Ordinary Portland cement (OPC) was used in the original old concrete. The specific gravity and fineness modulus of aggregates are summarized in Table 1. The concrete mix proportion is presented in Table 2. The specimens were moisture-cured for a day and demoulded before being air-cured in a laboratory. The specimens were exposed to natural marine environments, a tidal zone for 20 years from 1975 to 1995 at Sakata Port in Yamagata Prefecture, Japan. In 1995-2010, the specimens were kept and sheltered from the rain at the Port and Airport Research Institute (PARI) laboratory, Yokosuka, Japan [42, 43], then moved to the Kyushu University outside exposure site, Fukuoka, Japan. The exposure conditions until 41 years are displayed in Figure 3.

Table 1. Specific gravity and fineness modulus of aggregates

Aggregate Type	Specific gravity	Fineness modulus
Fine river sand	2.25	2.84
Coarse crushed stone	2.75	6.63

Table 2. Mix proportion of existing concrete

MSA (mm)	Slump (mm)	Air (%)	w/c (%)	s/a (%)	Unit weight (kg/m ³)				
					W	C	S	G	Adm.
20	12±2	4±1	68	47	204	300	793	964	1.2

MSA: maximum size of coarse aggregate; W: water; C: cement; S: sand; G: gravel; Adm.: admixture.



Figure 3. Exposure conditions

2.2. Preliminary Investigation

The defective appearance of the 41-years beams with cracks and rust stains is demonstrated in Figure 4. According to the previous research [44], the compressive strength test using core drilled specimens (50 mm in diameter and 100 mm in length) revealed that the compressive strength and elastic modulus after 40 years were 30.0 MPa and 27.0 GPa, respectively. These values after 40 years are almost the same as the 30 MPa and 22 GPa of compressive strength and elastic modulus, tested from cylinder specimens of 100 mm in diameter and 200 mm in length at the age of 28 days. Figure 5 illustrates the half-cell potential of rebar and crack patterns on the concrete surface at the age of 41-years. In both beam specimens, several longitudinal and transversal cracks were observed in the tensile area.

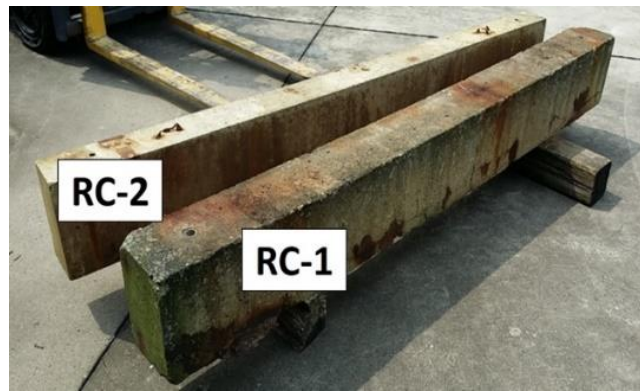


Figure 4. Defective appearance of RS beams

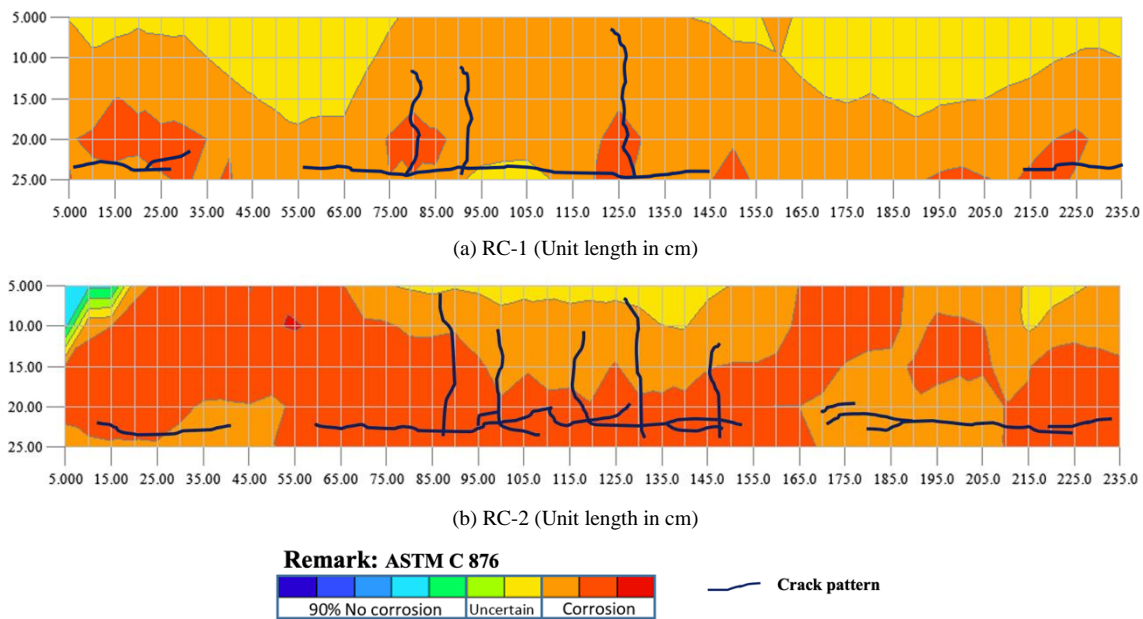
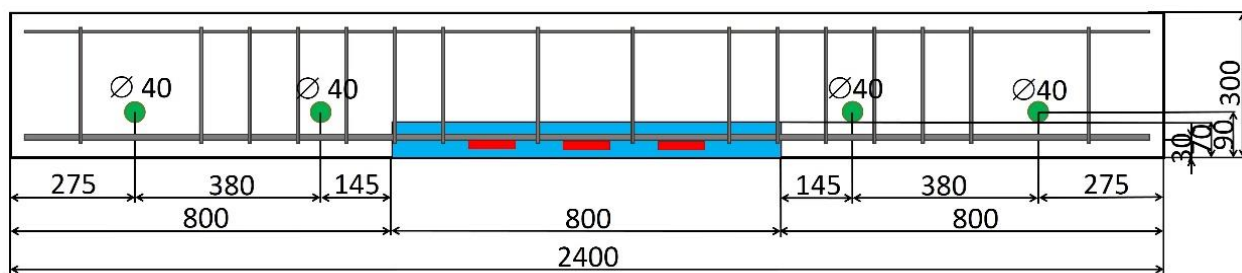


Figure 5. Half-cell potential and crack patterns, (a) RC-1 and (b) RC-2, before repair

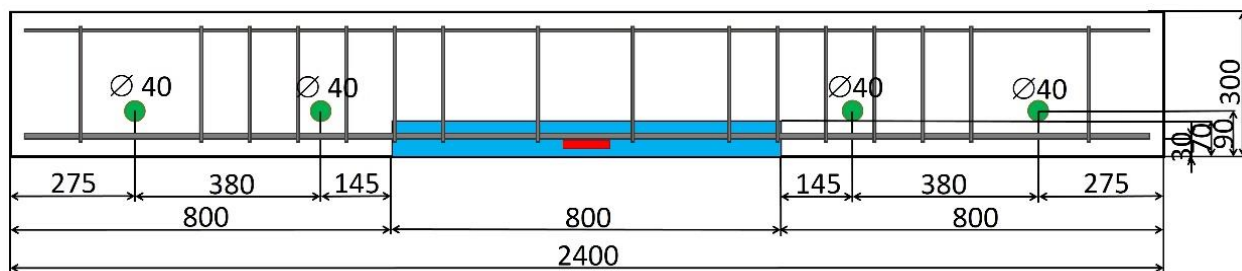
The longitudinal cracks coincide with the position of the tensile rebar with a maximum width of 1.9 mm and 2.2 mm for RC-1 and RC-2 respectively. No concrete spalling was observed in both specimens (RC-1, RC-2). In RC-1, 77% of area was categorized as “corrosion region” and 23% of area as “uncertainty region” according to the ASTM C 876, whereas 94% area of RC-2 was in “corrosion condition” and 6% was in “uncertainty condition”. The average total chloride ion concentration in the surrounding rebar was 4.65 kg/m³ in RC-1 and 4.75 kg/m³ in RC-2 respectively. These values are higher than the threshold chloride ion content of 1.2 kg/m³ for initiating corrosion (JSCE standard specification for concrete structures - 2007 “Design”). Based on JSCE standard specifications for concrete structures - 2018 “Maintenance”, these specimens are categorized as Grade II-1 (former Acceleration stage), where corrosion caused by cracking and rust appearance is observed. The intervention methods such as surface coating, patching, cathodic protection, or electrochemical desalination are expected to extend the service life of these deteriorated structures based on the JSCE standard specifications.

2.3. Repair Design

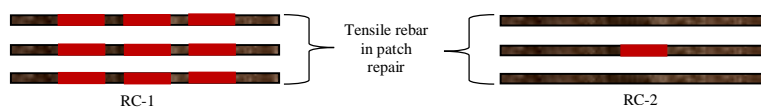
During preliminary investigation, several cracks were found in the middle tensile part. The corrosion probability of embedded steel bars was categorized as “high risk” with corrosion product (rust) accumulation on the steel bar surface. As a result, the application of the patching method with sacrificial anode cathodic protection was selected as the repair method for these deteriorated specimens. The nine (RC-1) and one (RC-2) sacrificial anodes were installed in the steel bar in the patch repair part. Furthermore, sacrificial anodes were inserted into a pre-drilled hole in the chloride contaminated old concrete area to stop corrosion progress. The location of the sacrificial anodes embedded in the patch repair area and the non-patch old concrete area is exhibited in Figure 6. The effectiveness expected for each stage, I, II and III in the repair process is summarized in Table 3. As presented in this table, in the first stage (Stage I), the patch repair part was protected by sacrificial anodes. In the second stage (Stage II), the protective current flow was interrupted and kept for one year to stabilize the steel surface. In the third stage (Stage III), the newly inserted sacrificial anodes protected the non-patch area (old concrete area).



(a) RC-1 (Unit in mm)



(b) RC-2 (Unit in mm)



(c) Anode setting position in the patch repair from the bottom view

Remark:

- Anode type A
- Anode type B
- Patch repair area

Figure 6. Repair design, (a) RC-1, (b) RC-2, and (c) anode setting position in the patch repair

Table 3. The details of the repair process [45]

Stage	Method	Duration	Expected effectiveness
I	Patch repair and sacrificial anodes were applied in the middle tensile rebar part.	200 days	Protected condition in the patch repair section by replacing “chloride contaminated concrete” into “chloride free mortar” and current flow generated from sacrificial anode to rebar.
II	Sacrificial anodes in the patch repair were disconnected.	One year	Protected condition by the patch repair material.
III	Additional sacrificial anodes were installed in the non-patch repair concrete.	One year	In order to protect all of the specimens, additional sacrificial anodes were embedded in the non-patch repair part.

Polymer-modified mortar with compressive strength of 40.5 MPa and bending strength of 8.6 MPa at 28 days (Figure 7-a) was utilized as the patch repair material after applying EVA (Ethylene/Vinyl Acetate), copolymer emulsion as an adhesive material coating agent on the boundary surface (Figure 7-b). Two types of sacrificial anodes, Type A and Type B (Figures 7-c and 7-d), were employed to protect the rebars in non-patch and patch repair parts.

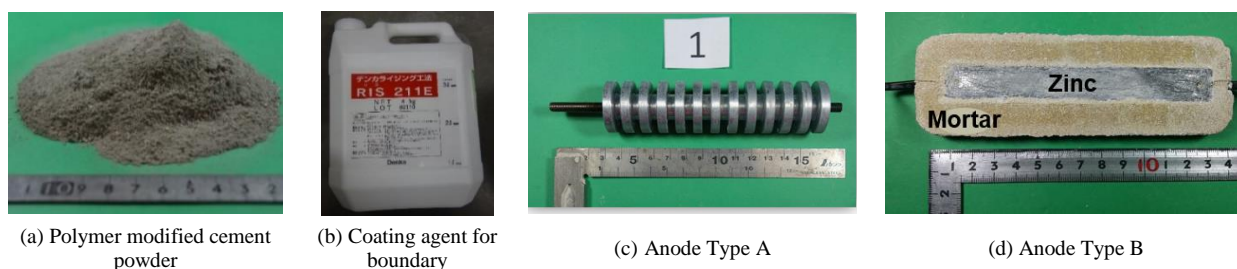


Figure 7. Materials for repair process

The anode type selection in this research was based on the available space where it was embedded. Type B with thickness, width and length of 13 mm, 45 mm, and 140 mm was applied to the patch repair part, while Type A, cylindrical

ribbed anode with a diameter of 30 mm and length of 130 mm, was applied to the existing concrete. These anodes were made of zinc covered with porous mortar, including lithium mono-hydrated solution, to maintain the zinc corrosion active. The lithium-based solution keeps the surroundings of the zinc humid and the efficiency of the anode during its service life [46].

3. Results and Discussion

3.1. Performance of Sacrificial Anodes in the Patch Repair

In the first stage of the repair process, nine and one sacrificial anodes were installed in polymer-modified mortar in the middle tensile part of RC-1 and RC-2, respectively. This repair method has been reported previously [45]. The potential change of steel bars until two-month is presented in Figures 8 and 9. Instant-off and resting potential of the tensile rebar in RC-1 and RC-2 unveiled the trend that the polarization by sacrificial anodes is highly limited. The polarized area was wider for RC1 than for RC2 due to the difference in the number of the sacrificial anodes, nine anodes for RC1 and only one anode for RC2.

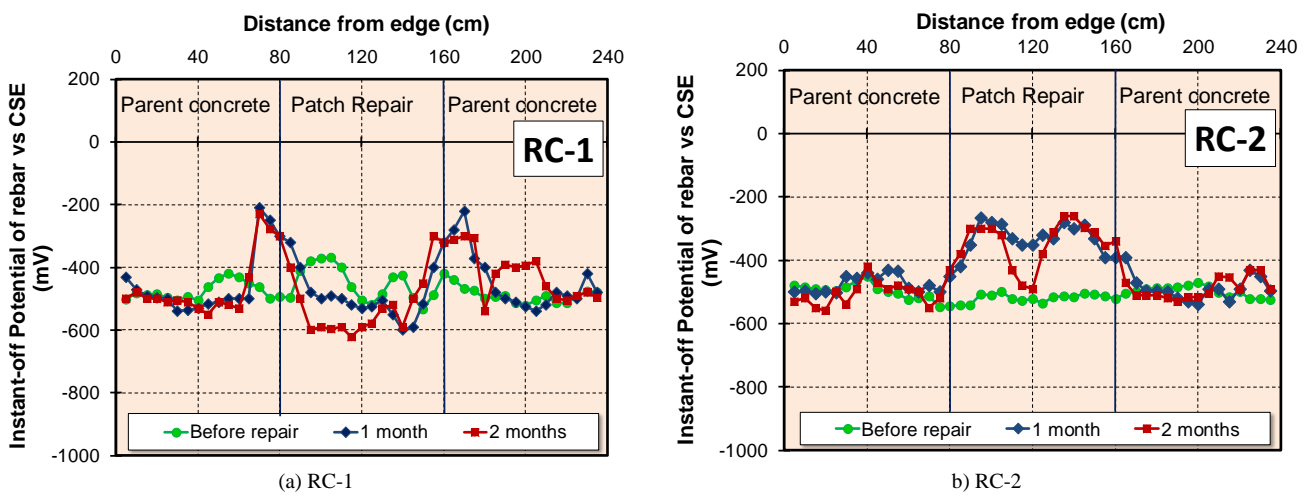


Figure 8. Instant-off potential of the rebar (The sacrificial anodes were in the patch repair, Stage I)

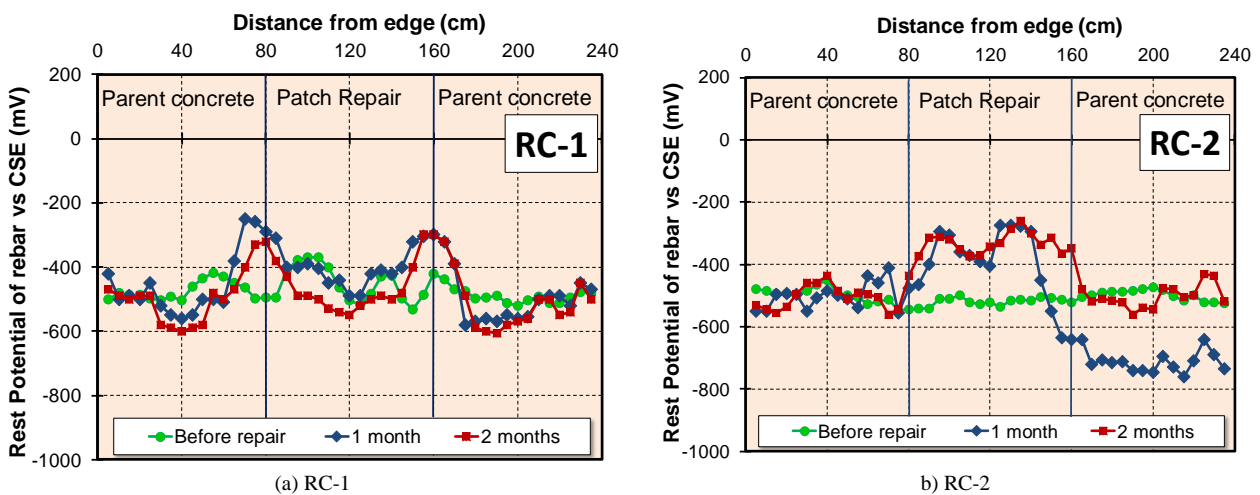


Figure 9. Resting potential of the rebar (The sacrificial anodes were in the patch repair, Stage I)

As depicted in Figure 10, even in RC-1, the polarization was also limited to the patch repair section; the protective current did not reach the existing concrete due to the electrochemical incompatibility between the repairing mortar and old concrete. In RC-1, some patch repair areas were protected by sacrificial anodes, while none of the areas around the anode was protected in RC-2.

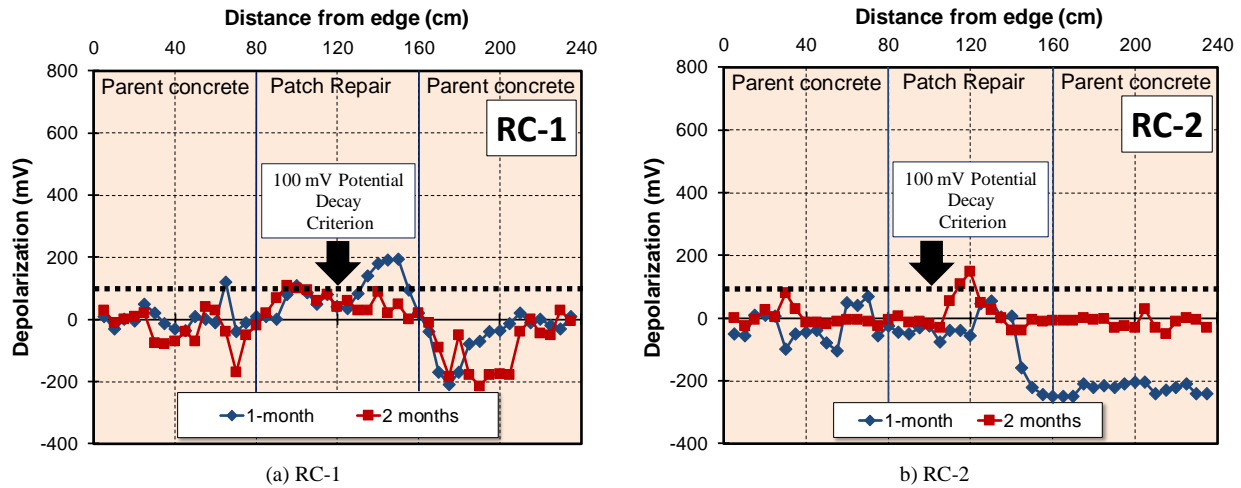


Figure 10. Depolarization value of the rebar (The sacrificial anodes were in the patch repair, Stage I)

3.2. Current Interruption of Sacrificial Anodes in the Patch Repair

After 200 days of connection (Stage I), the sacrificial anodes in the patch repair were disconnected for a year as the second stage of the repair process. In this stage, Stage II, RC beam specimens were exposed to the dry condition in the laboratory. The resting potential of the tensile rebar before and after the one-year period of current interruption is presented in Figure 11. This demonstrates that the resting potential of tensile steel bars in the patch repair shifted farther more than that in the non-patch repair area. In RC-1, the potential shift to noble was particularly remarkable in the patch repair area. This was due to the effectiveness of the current flow generated by the sacrificial anode. Both in RC-1 and RC-2, the resting potential of the steel bar in the patch repair section was in the “90% no corrosion” region according to the ASTM criteria. This was due to the effectiveness of polymer-modified mortar as the replacement material without chloride contamination.

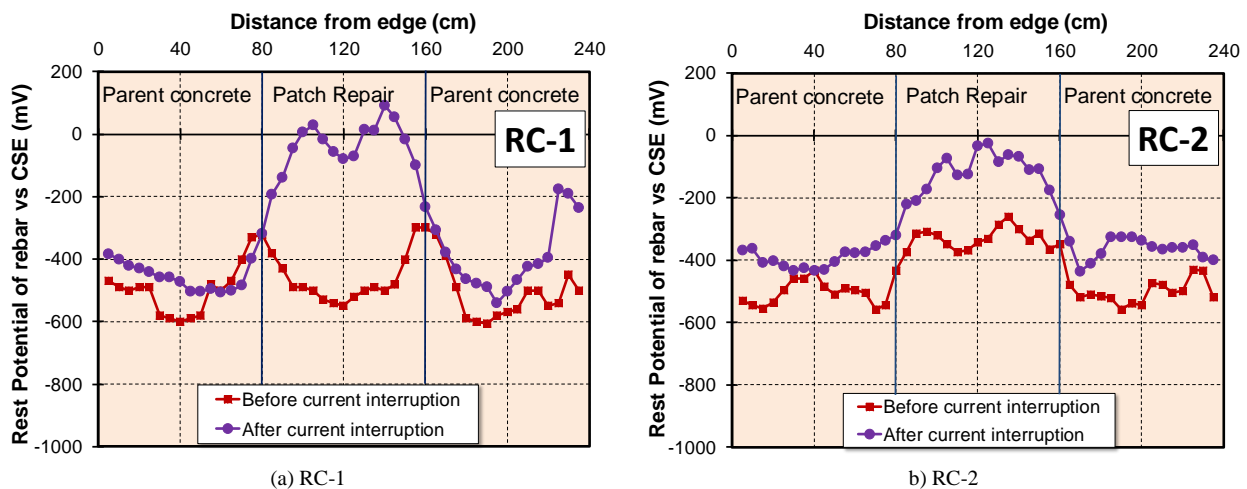


Figure 11. Resting potential of the tensile steel bar before and after the one-year current interruption, Stage II

3.3. Effect of Sacrificial Anodes in the Non-Patch Repair

In the third stage of the repair process, four additional sacrificial anodes were installed in the existing old concrete to protect the steel bars. After two months of the anodes connection, sacrificial anodes in the patch repair were also re-connected. The protective current density of the Type B anode in RC-1 and RC-2 was almost the same as 10 - 20 mA/m², which is the minimum design limit of cathodic protection according to the BS EN ISO 12696 [47].

Instant-off potential, resting potential, and depolarization value of the tensile rebar after one year of installing the Type A anode in old concrete are displayed in Figures 12 to 14. The 100 mV potential decay and the common criteria are exhibited in Figure 13. As illustrated in these figures, a fall in potential was observed for all specimens after applying sacrificial anodes. This indicates that these anodes polarized the potential of the rebar to more negative than -750 mV vs. CSE. Here, the potential shift was almost limited to approximately 200 mm away from the anode. This implies that the optimum distance of one sacrificial anode in the non-patch repair was about 400 mm.

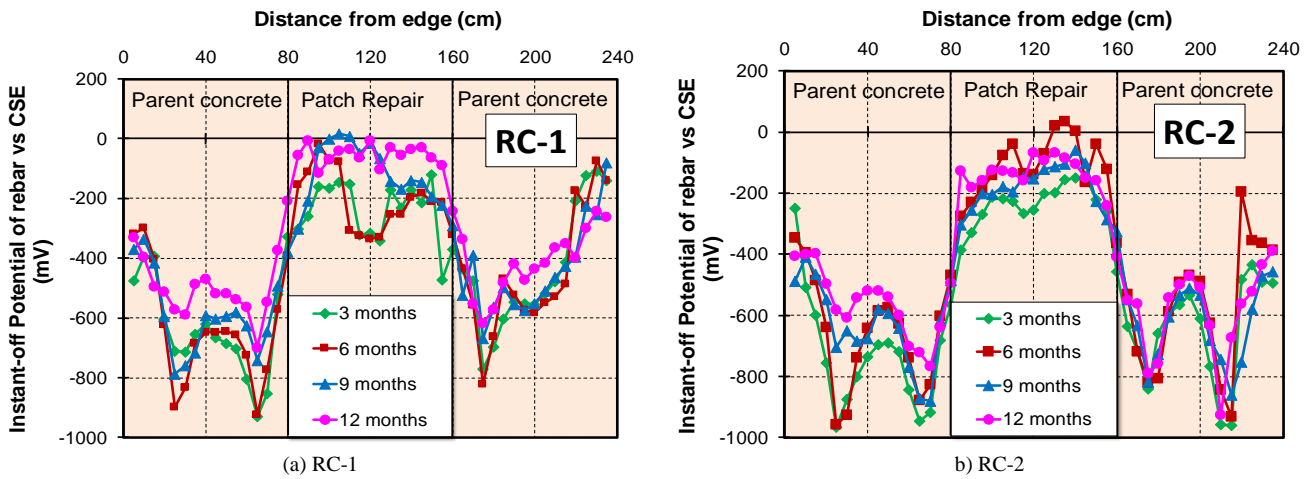


Figure 12. Instant-off potential of tensile rebar in RC-1 and RC-2, Stage III

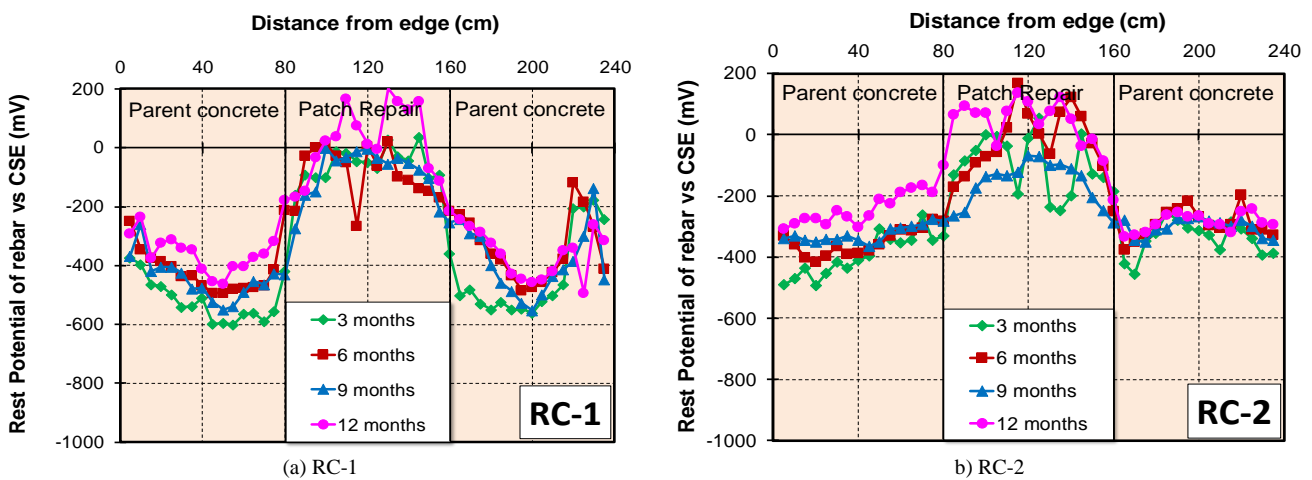


Figure 13. Resting potential of tensile rebar in RC-1 and RC-2, Stage III

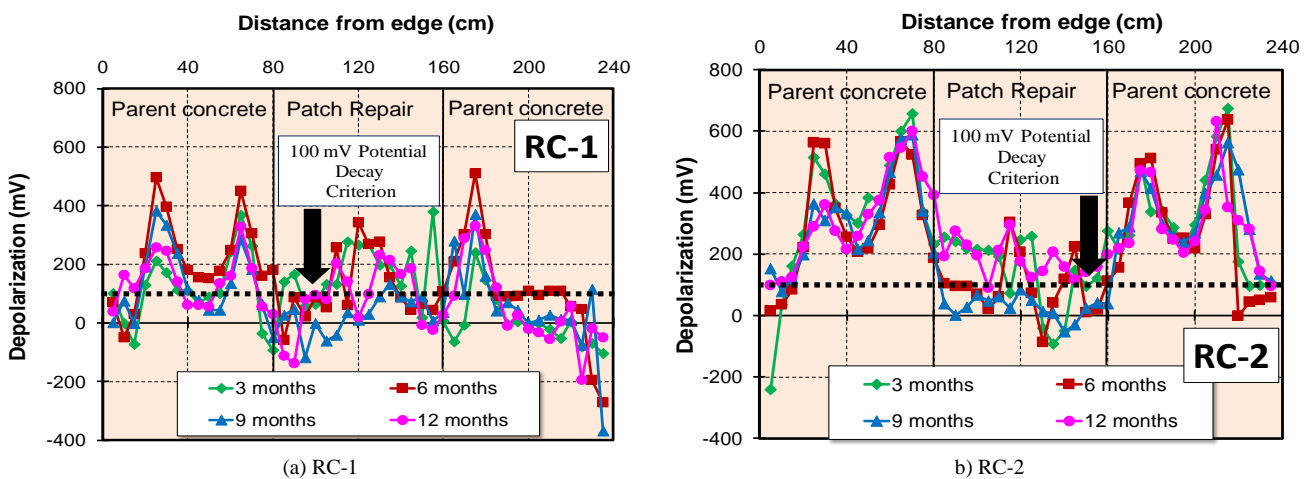


Figure 14. Resting potential of tensile rebar in RC-1 and RC-2, Stage III

Figure 15 presents the resting potential of the beam specimens after one year of the connection. This figure depicts that the whole area of the beam was in “90% no corrosion” or “uncertain”. This signifies that the steel bars surfaces were improved by sacrificial anodes connection. The potential in the patch repair part, as shown in blue, was rather positive, indicating that the steel bar was in a better conditions. Compared to the patch repair part, the potential in the non-patch repair, in the old concrete, demonstrated negative potential, in yellow or green color. However, the steel bars in old concrete were also in a protected condition.

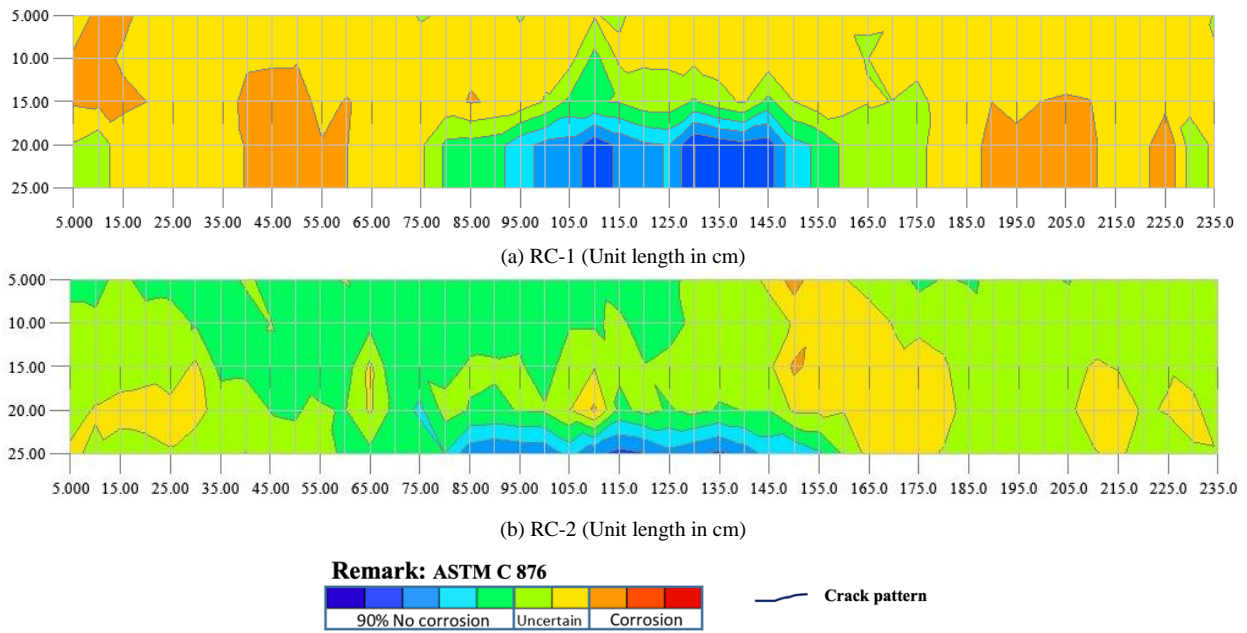


Figure 15. Resting potential at the end of the test (one year after the second anode application)

3.4. Service Life Estimation of Sacrificial Anodes

To assess the service life of applied sacrificial anodes, it is necessary to calculate the total mass loss of zinc material during its operational lifetime. Here, a theoretical prediction based on Faraday’s law was performed. Figure 16 presents the current flow in both specimens, RC-1 and RC-2. The mass loss of zinc was calculated by using the Equation 1:

$$Mass\ Loss\ (g) = \frac{Charge\ passed\ (C)}{Faraday's\ constant\ (\frac{C}{mol})} \times \frac{Molecular\ Mass\ of\ Zinc\ (\frac{g}{mol})}{Valency\ of\ Zinc} \tag{1}$$

where, Faraday’s constant = 96500 Coulomb/mol, Molecular mass of zinc = 65.382 g/mol, and Valency of zinc = 2.

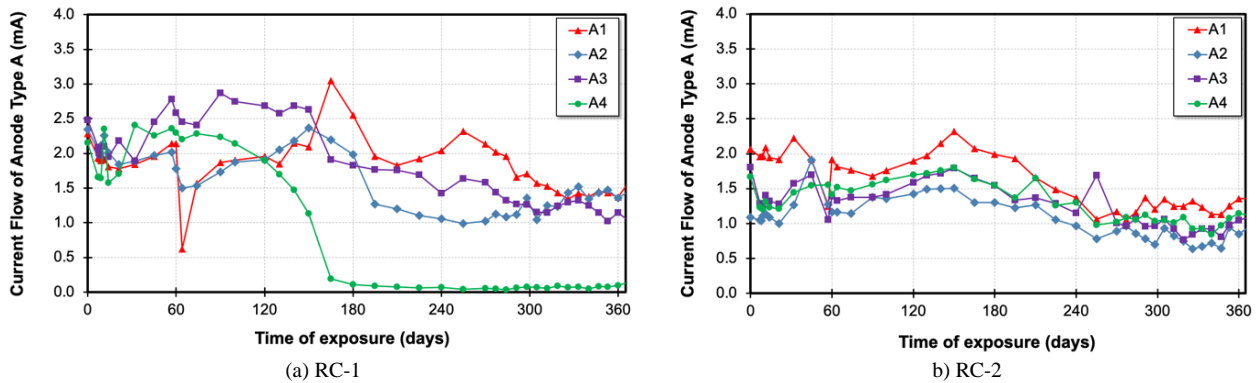


Figure 16. Current flow of each anode in (a) RC-1 and (b) RC-2

The cumulative charge for sacrificial anodes after 365 days in RC-1 and RC-2 was 48992 Coulomb and 31957 Coulomb, respectively. By substituting these values in Equation (1), theoretical mass loss 57.12 grams and 57.13 grams for three sacrificial anodes in RC-1 and four sacrificial anodes in RC-2 were calculated. An “A4” anode in RC-1 became inactive after 160 days, as illustrated in this figure. Therefore, the service life prediction for RC-1 was based on three active anodes. The initial weight of sacrificial anodes installed in the non-patch repair was 1,251 grams of three anodes in RC-1 and 1,667 grams of four anodes in RC-2. The remained service life of the anodes was calculated by Equation (2). Table 4 presents the calculated service life by assuming a constant consumption of anodes for the entire service life. It depicts a service life prediction of 21 years and 28 years for RC-1 and RC-2.

$$Service\ life\ (year) = \frac{Initial\ weight\ of\ anodes\ (g)}{Rate\ of\ consumption\ (\frac{g}{day}) \times 365} \tag{2}$$

Table 4. Experimental rate of anodes consumption per day

Specimen	Initial weight of anodes (gram)	Mass loss until 365 days (gram)	Rate of consumption (gram/day)	Predicted service life (year)
RC-1	1,251.0	57.12	0.052	20.81
RC-2	1,666.4	57.13	0.040	28.23

Previous research [45] reported the deterioration progress and the performance degradation of typical 40-year-RC beam specimens with or without initial pre-cracks with the recorded data at ten years [42] and 20 years [43, 48]. Deterioration progress due to corrosion involves the initiation, propagation, acceleration, and deterioration stages. The deterioration assessment criteria based on Yokota et al. [43] are exhibited in Table 5. In this paper, two RC beam specimens without initial cracks were used to test the effectiveness of the repair method. Specimens RC-1 and RC-2 were almost at the same deterioration degree as the 41-year-old one with no pre-crack specimen tested by Dasar et al. [44]. Figure 17 presents the effectiveness of the repair method described in the deterioration progress diagram, which continued until 64 years for RC-1 and 72 years for RC-2, respectively. To assure effectiveness, it is recommended that regular monitoring of potential or current flow generated by the sacrificial anodes should be carried out until the expected service life of the repaired structures. With this monitoring, the service life of RC-1 and RC-2 could be extended to around 64 and 72 years, respectively.

Table 5. Assessment criteria of the deterioration stage [43, 44]

Evaluation Item	Deterioration Degree					
	0	1	2	3	4	5
Corrosion of reinforcing steel (X_1)	None	Rust spots found on concrete surface	Partial rust stains were found on the concrete surface	Significant rust staining	Significant floating rust	A dramatically increased amount of floating rust
Cracking (X_2)	None	Partial cracks found on concrete surface	Some cracks	Many cracks, including some of several millimeters or more in width	Many cracks of several millimeters in width	-
Spalling of covering concrete (X_3)	None	None	Partial floating of concrete found	Partial spalling found	Significant spalling	Drastic spalling

Total deterioration progress value = $X_1 + X_2 + X_3$

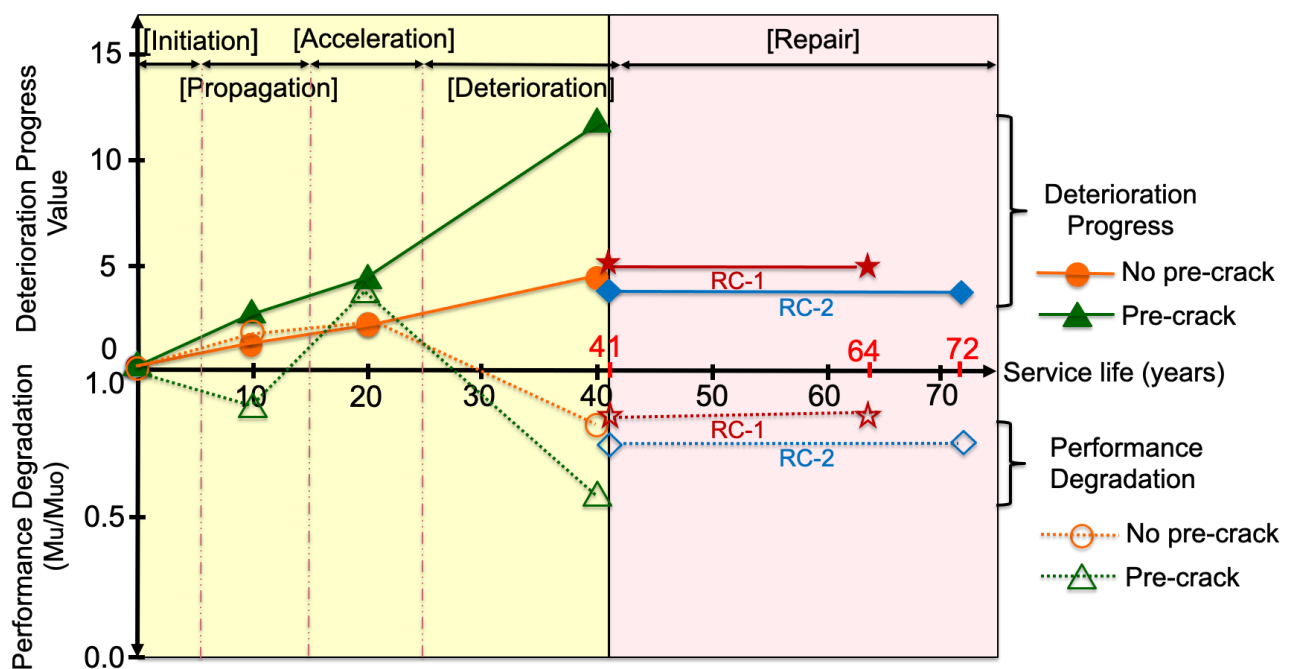


Figure 17. Concept of the deterioration progress and performance degradation of RC members

4. Conclusion

The time lag application of sacrificial anode cathodic protection as a repair strategy on patch and non-patch repairs was experimentally and successfully tested on deteriorated 41-year-old RC beams. This type of repair method is used on patch structures that have utilized sacrificial anodes in the previous stage, but these structures will require additional sacrificial anodes in the future. The installation of the new sacrificial anodes is conducted when the previous anodes performance is low and current interruption is applied. Based on the tests, the application of sacrificial anodes in the patch repair area enhanced the corrosion resistance of steel rebar, which was indicated by the resting potential shift to a noble value. After one year of interruption, the steel bars embedded in chloride-free polymer-modified mortar remained passive without any sign of corrosion. It indicates that polymer-modified mortar had a persistent protective effect against the steel corrosion inside. Sacrificial anodes embedded in both non-patch and patch repair concrete could control rebar corrosion with an optimum distance of 400 mm covered by one sacrificial anode. However, this repair method, which was necessary to obtain protective effects for both patch repair and non-patch repair areas. Based on the service life prediction of sacrificial abodes, RC-1 and RC-2 could be extended by 20 to 30 years.

5. Declarations

5.1. Author Contributions

Conceptualization, P.A., R.S.R., and H.H.; data collection, P.A. and V.A.; validation, P.A. and R.S.R.; data analysis and interpretation, P.A., D.Y., and H.H.; writing—original draft preparation, P.A.; writing—review and editing, H.H.; supervision, H.H.; project administration, D.Y.; funding acquisition, H.H. All authors have read and agreed to the published version of the manuscript.

5.2. Data Availability Statement

The data presented in this study are available in article.

5.3. Funding

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5.5. Conflicts of Interest

The authors declare no conflict of interest.

6. References

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