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AN EXPERIMENTAL STUDY ON IMPROVEMENT OF CONCRETE AND REPAIR EFFECTS USING ELECTROCHEMICAL METHODS

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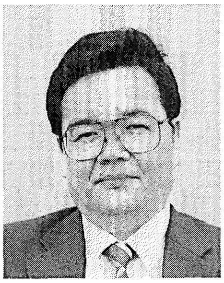

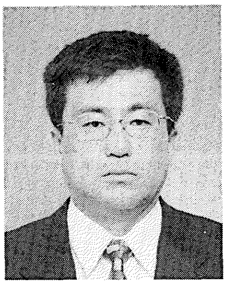

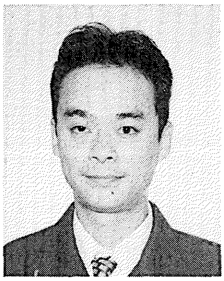

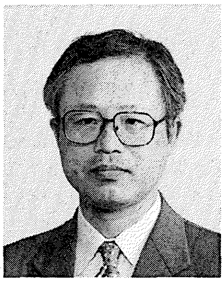

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An experimental study was carried out to find the threshold value of the chloride ion to hydroxy ion ratio ($[Cl^-]/[OH^-]$: mol ratio) for the onset of reinforcing steel corrosion in concrete. After application of an electrochemical concrete improvement method (desalination and realkalizaton) to specimens, a five-year follow-up survey was made to check the effectiveness of the rehabilitation method.
The results of tests showed that reinforcing steel did not corrode when pH value was 12 or higher and Cl^-/OH^- ratio was 1 or lower. Rehabilitation using the desalination and realkalization method increased pH value to between 12.6 and 13.2 and reduced water-soluble chloride content to 0.47 to 0.56 kg/m³ around reinforcing bars in concrete of existing structures.
Almost no rediffusion of chloride ions into concrete around reinforcing bars was detected in the five-year follow-up survey, and the presence of clear shadow spots behind the reinforcement was not noted.

Key Words: corrosion of reinforcing steel, threshold of $[Cl^-]/[OH^-]$, repair, desalination, realkalization

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1. INTRODUCTION

Concrete is known as a material of excellent durability. Not a few concrete structures have been in service for long periods and even now remain in satisfactory functional condition. However, it is also a fact that certain structures have deteriorated for various reasons, and concrete, which had been thought of as a maintenance-free material, was found actually not to be so.In fact, durability cannot be attained unless proper maintenance is carried out, and in a sense, what should have been obvious is only now beginning to be recognized.

Many of the changes seen in railway structures, of which rigid-frame viaducts are representative, are caused by corrosion of reinforcing steel. Where such deteriorated structures are located in normal environments, they are often treated by the " lining method", which entails giving a surface coating to the concrete. Results of previous investigations[1] [2] suggest that the causes of reinforcing bar corrosion are chlorides and carbonation due to inadequate chloride removal from fine aggregate, particularly the latter. The actual situation is that, in application of the lining method, there were many places where the chloride content of concrete had not been measured, and that when chloride content was high, repairs by lining and similar methods were not necessarily done after having removed the chloride. Recognizing that there are limits to the durability of lining materials themselves, a method that improves the quality of concrete itself through realkalization (after desalination) by an electrochemical method has been tried in recent years as a radical alternative to the lining method. In this case, it is necessary to clearly determine the chloride content and the alkaline environment after desalination and realkalizaton, which will ensure that subsequent reinforcing bar corrosion will not occur. However, it has not necessarily been made clear what influences chlorides and carbonation, respectively, have on reinforcing bar corrosion.

In this study, an attempt is made at the laboratory level to quantify the possibility of reinforcing bar corrosion in concrete with the chloride ion to hydroxy ion ratio ($[Cl^-]/[OH^-]$) as a parameter. The way in which the quality of concrete in an actual structure was improved through desalination and realkalizaton are also discussed. The results of a follow-up survey made over a period of five years are reported.

2. INFLUENCE OF CHLORIDE ION-HYDROXY ION RATIO ($[Cl^-]/[OH^-]$) ON REINFORCING STEEL CORROSION

The results of a previous survey[3] suggest that, with a structure in a normal environment, when carbonation of concrete has not progressed as far as reinforcing steel, no prominent corrosion of the steel is seen. This is in agreement with the thinking that, even though there may be chloride ions (Cl^-), the presence of hydroxy ions (OH^-) above certain concentration accelerates the formation of a passive film on the reinforcing bar surface, thus repairing any passive film, which may have been broken. In other words, whether or not reinforcing steel corrosion occurs should be greatly influenced by the $[Cl^-]/[OH^-]$ ratio of chloride ion concentration to hydroxy ion concentration in the steel's environment. Also threshold value should exist. The results of past studies are given in Table 1.

Table 1 Threshold $[Cl^-]/[OH^-]$ values

	pH	$[Cl^-/OH^-]$	Experimental conditions
Hausmann	11.6 12.4 13.2	0.5 0.61 0.83	Steel in alkaline solution
Gouda & Diamond	11.8 12.1 12.6 13.0 13.3	0.57 0.48 0.29 0.27 0.30	Steel in alkaline solution
Page	13.91	0.54	Steel in cement paste
Page & Havdahl		62	Steel in cement paste containing 30% silica fume
Yonezawa, V.Ashworth,R.P.M. Procter		5	Steel in cement mortar
S.E. Hussain et al.	Less than 13.3 More than 13.3	1.7~2.0 1.28~1.86	Steel in cement mortar

(1)Threshold Value for Reinforcing Steel Corrosion in Alkaline Solution

Hausmann's value of $[Cl^-]/[OH^-] \leqq 0.6$ [4] is well known as the threshold for reinforcing steel corrosion in an alkaline solution. Hausmann found that when pH values were 11.6, 12.4,and 13.2, corrosion would not occur if $[Cl^-]/[OH^-]$ values were 0.5, 0.61, and 0.83, respectively. The 0.83 value was considered as being of poor reliability because of difficulties met in measurement. Hausmann's conclusion was that although the threshold value of $[Cl^-]/[OH^-]$ with pH in the range of 11.6 to 12.4 shows a trend of increase, it is approximately 0.6.

Gouda studied alkaline solutions with pH values from 11.75 to more than 135 [5] . Diamond modified Gouda's results using the activity coefficient of NaOH to estimate hydroxy ion concentration from pH value. A check of the results reveals that the threshold $[Cl^-]/[OH^-]$ value decreases as pH value increases. In a range of pH 11.8 to 12.1, there is good agreement with Hausmann's value of 0.60. Further, Hausmann[6] holds that these values are influenced by the amount of oxygen supply. He reports that, consequently, the threshold value increases if the concrete is fully saturated with water.

(2) Threshold Value for Reinforcing Steel Corrosion in Cement Paste

Page et al. [7] [8] , in tests using reinforcing bars embedded in cement paste, found that when the pH value of pore water and $[Cl^-]/[OH^-]$ are 13.91 and 0.54, respectively, the steel is inactive. This value of 0.54 at a pH of 13.91 is more than double the $[Cl^-]/[OH^-] = 0.23$,the value estimated from Gouda's results. Therefore, in case of steel embedded in cement paste, $[Cl^-]/[OH^-]$ as the threshold value is thought to be a value far higher than that in an alkaline solution. Page et al.[8] found that steel embedded in hardened cement paste is inactive up to a $[Cl^-]/[OH^-]$ value of 62.

(3) Threshold Value for Reinforcing Steel Corrosion in Cement Mortar

In cement mortar, the conditions are still more lenient, and Yonezawa[9] et al. obtained a threshold of $[Cl^-]/[OH^-] \leqq$ approx. 5. They also reported on the characteristics of steel bar corrosion for two different states of adherence between steel bar and mortar, a) and b) as defined below.

a) Filter paper separating embedded steel bar and mortar
b) Steel bar embedded directly in mortar

Yonezawa found that the threshold value of $[Cl^-]/[OH^-]$ was far higher for steel bars embedded in cement mortar under both adherence conditions compared with that in an alkaline solution.

The report by S.E. Hussain et al.[10] concerns corrosion of steel in mortar with a water-cement ratio of 0.55. Although hydroxy ion concentrations were varied in the comparatively narrow range of pH 13.26 to 13.36, the threshold value of $[Cl^-]/[OH^-]$ decreased with increasing hydroxy ion concentration. For example, at hydroxy ion concentrations of pH 13.30 and under, the threshold values of $[Cl^-]/[OH^-]$ were in a range of 1.7 to 2.0, while at hydroxy ion concentrations above pH 13.30,the threshold values of $[Cl^-]/[OH^-]$ were in a range of 1.28 to 1.86.

(4) Threshold Value for Reinforcing Steel Corrosion in Concrete

With regard to the above reports, Glass and Buenfeld[11] studied the proposed $[Cl^-]/[OH^-]$ threshold value and chloride threshold value based on published data. The objects of examination were reinforced concrete structures; concrete specimens exposed to an outdoor environment; concrete, mortar, and cement paste specimens in a laboratory environment; and simulated pore water. The study also included parts of (1) to (3) above.

A summary of the examination results follows.
- Regarding the method of defining threshold value as the ratio of chloride content or water-soluble chloride content to hydroxy concentration, the effectiveness of such a definition cannot be verified by the data available.
- Initial oxidation at the anode increases local activity of chlorides to accelerate the release of bound chlorides.
- Even though chlorides are bound, there exists a risk of corrosion occurring.
- The most important corrosion-prevention property of cement is that of restricting pH value from declining locally to below 12.6.
- Regarding the threshold value for steel corrosion in concrete, it is best to indicate the total chloride content by weight of cement.

The maximum and minimum threshold values for concrete and mortar are compiled in Table 2. Threshold values are given as total chloride content by weight of cement.

Table 2 Threshold values for corrosion of steel in concrete and mortar

	Threshold values of total chloride ions for steel corrosion (Cl^-wt%Cem)
Concrete structure	0.17～ 2.2 (4)
Concrete specimen (outdoor exposure)	0.32～ 1.9 (4)
Concrete specimen (laboratory)	0.5～ 2.5 (4)
Mortar specimen (laboratory)	0.25～ 1.6 (2)

Figures in ( ) indicate numbers of references

3. EXPERIMENTS ON THRESHOLD $[Cl^-]/[OH^-]$ VALUES FOR REINFORCING STEEL CORROSION IN CEMENT MORTAR

Experiments were conducted on threshold [Cl]/[OH] values for reinforcing steel in cement mortar.

(1) Experimental Method

Mortar specimens were made with varying pH and [Cl]/[OH] values, placed in a desiccator, and whether or not corrosion of reinforcing bars in the specimens had occurred was investigated.

a) Materials

Reinforcing steel: steel bars for reinforcement (JIS G 3112) were shaved to form polished bars (ϕ 13 mm x 100mm). The chemical composition and results of tension tests of the original bars are as follows:

C = 0.14%, Si = 0.18%, Mn = 0.49%, P = 0.017%, S = 0.011%, yield point = 330 N/mm^2, tensile strength = 472 N/mm^2, elongation = 29%

Sand: standard sand ("standard sand for testing strength of cement" as specified in JIS R 5201), specific gravity: 2.64

Cement: ordinary portland cement, specific gravity: 3.16

Alkali: sodium hydroxide

Chloride: commercial table salt, NaCl: 99%

b) Specimens

Cylindrical mortar specimens of diameter 5 cm, height 10cm, with reinforcing bars embedded at centers were made with thickness of cover 19 mm. The bars were bonded to the bottom surfaces of molds using epoxy resin before casting the mortar. Cover at the ends of specimens was zero. The appearance and dimensions of a specimen are shown in Fig. 1.

Specimens were of the kinds indicated in Table 3, with 4 levels of pH and 10 levels of $[Cl^-]/[OH^-]$, two of each kind being made.

Table 3 Varieties of specimens

pH	12.0 12.5 13.0 13.5	4 levels
$[Cl^-/OH^-]$	0 0.1 0.5 1.0 1.5 2.0 3.0 5.0 7.5 10	10 levels

Mortar was JIS standard mortar with a water-cement ratio of0.50.The mixture proportions per cubic meter are given in Table 4.

Table 4 Mixture proportions of mortar

Ordinary portland cement	502kg/m^3
Standard sand	1507kg/m^3
Water	251kg/m^3

The pH values were set up by adjusting the amount of NaOH in mixing water. However, a pH of 12.0 was considered to mean a saturated calcium solution, and no adjustment with NaOH was node.

For example, for pH = 13.0 and $[Cl^-]/[OH^-]$ = 1.0, since $[OH^-]$ = 0.1 mol/l and $[Cl^-]/[OH^-]$ = 1.0 from pH = 13.0, $[Cl^-]$ =0.1mol/l = 3.55g/l (∵ molecular weight of chlorine), and since the water content of a cubic meter of mortar is 251 kg, the quantity of chloride ions used in the experiments can be determined by the following equation:

$$3.55\ (g/l) \times 251\ (l/m^3) = 891\ (g/m^3)$$

The amounts of chloride ions used in the experiments are given in Table 5.

Table 5 [Cl-]/[OH-] when placing mortar, total chlorides in mortar [g/m3], and percentage of total chlorides by weight ofcement [wt%]

		pH			
		13.5	13.0	12.5	12.0
Cl^- / OH	0	0 (0)	0 (0)	0 (0)	0 (0)
	0.1	285.1 (0.057)	89.1 (0.018)	26.7 (0.005)	8.9 (0.002)
	0.5	1425.7 (0.284)	445.5 (0.089)	133.7 (0.027)	44.5 (0.009)
	1	2851.4 (0.568)	891.0 (0.177)	267.3 (0.053)	89.0 (0.018)
	1.5	4277.1 (0.852)	1336.5 (0.266)	401.0 (0.080)	133.5 (0.027)
	2	5702.8 (1.136)	1782.0 (0.355)	534.6 (0.106)	178.0 (0.035)
	3	8554.2 (1.704)	2673.0 (0.532)	801.9 (0.160)	267.0 (0.053)
	5	14257.0 (2.840)	4455.0 (0.887)	1336.5 (0.266)	445.0 (0.089)
	7.5	21385.5 (4.260)	6682.5 (1.331)	2004.8 (0.399)	667.5 (0.133)
	10	28514.0 (5.680)	8910.0 (1.775)	2873.0 (0.572)	890.0 (0.177)

Figures in ( ) indicate percentages of total chlorides by weight of cement (wt%).
Shaded portions indicate corroded parts.

c) Environmental Conditions

The experimental apparatus for accelerating corrosion of steel is shown in Figs. 2.1 to 2.3.

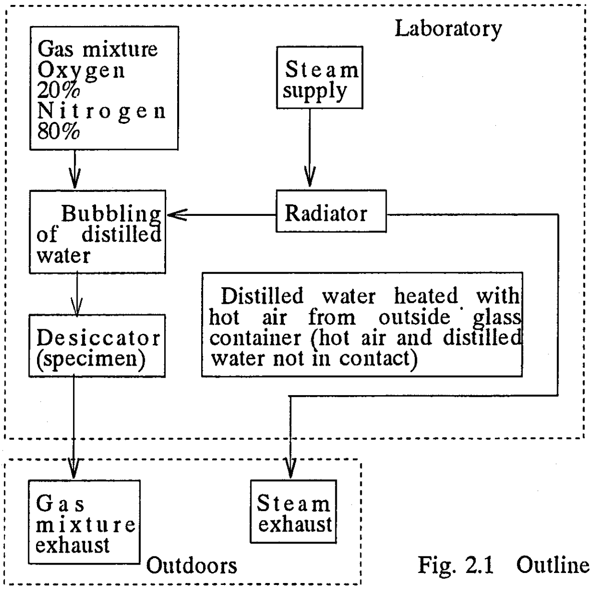

Fig. 2.1 Outline of experimental apparatus

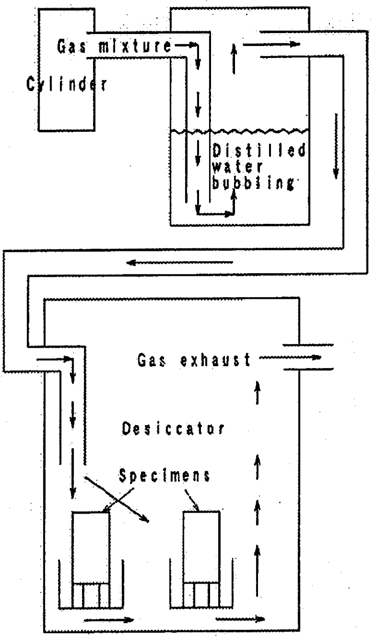

Fig. 2. 2 Schematic of experimental apparatus

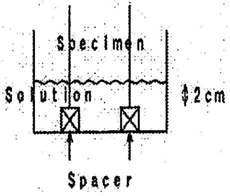

Fig. 2. 3Method of setting specimen

In order to prevent progress of carbonation, a gas mixture of 20% oxygen and 80% nitrogen (free of carbon dioxide) was passed through distilled water and conducted into the desiccator in which specimens were resting.

The specimens in the desiccator were in a moist but not dripping condition. Temperature and humidity were measured using an automatic temperature-humidity recorder. Testing was performed up to the age of 115 days. However, because of the possibility that corrosion of steel would not occur even over this length of time, parts of the experimental environment were changed. To elaborate, the conditions between 4days and 33days of age were temperatures from 20 to 30℃ and relative humidities from 80 to 90%. Then, later, the conditions were changed to temperatures from 20 to 30 ℃ and relative humidities from 80 to 90% up to 90days of age, with the bottom 2 cm of mortar specimens immersed in an aqueous solution. Also, it was decided to warm the distilled water used for bubbling. Thereafter, the ambient temperature was made 60℃ up to 115days of age. The rates of absorption in mortar portions of specimens were measured after completion of the experiments.

Absorption was measured by following JIS A 1110 and the procedure below was used.

Absorption by mortar specimens was allowed to proceed for 24 hours in water at a temperature of 20 ± 2 °C . Upon removal from the water, surface water was wiped off with an absorbent cloth to obtain a saturated surface-dry condition, after which the mass (m_s) was immediately measured.

Upon determination of mass (m_s), specimens were dried for 24 hours at 100-110°C and, after cooling to room temperature, the mass (m_D) was measured.

The absorption of specimens was calculated by the following equation:

$$Q = m_s - m_D \ / \ m_D \times 100$$

where, Q: absorption (%)

m_s: mass of sample in saturated surface-dry state (g)

m_D: mass of sample after drying (g)

(2) Experimental Results

The temperatures and humidities in the dessicator satisfied the conditions set up. Reinforcing bars were taken out at the age of 115days and the degrees of corrosion were investigated. The results are shown in Photo. 1.

The results of determining whether or not corrosion had occurred are given in Table 5. The shaded portions of the table indicate specimens, which had corroded. Since cover at ends of specimens was zero, there were some specimens in which corrosion appeared to have progressed from the ends. Here, the effects of cracking and inadequate cover frequently seen in actual structures were taken into consideration, and these specimens were included among cases of corrosion to be on the conservative side.

The absorption of mortar portions of specimens was indicated as 7.6 wt% as a result of measurements. The unit mass of mortar was 2050 kg/m^3 . Portions of cement paste which had been in direct contact with reinforcing bar surfaces, where specimens had been split and bars removed, were sampled and X-ray diffraction was performed. Distinct peaks of Friedel's salt were not seen in the results.

Given the results of the experiments, and since it was expected that the $[Cl^-]/[OH^-]$ conditions initially set up would change during the course of the experiments, pH values of mortar in the specimens were measured for $[Cl^-]/[OH^-]$ values of 0.1, 1.5, and 2.0.

The method of measurement was as follows:

① Dry mortar for 7 days in a dryer at 60 °C .

② Coarsely grind the dried mortar followed by fine pulverization with a mortar and pestle to 88 μm and under.

③ Add 40 g of distilled water to 100 g of the finely pulverized mortar and store in a desiccaator (filled with N_2 gas) at 20 °C for 7 days.

④ Using the suction filtering method, separate moisture from the finely-divided mortar to which distilled water had been added.

⑤ Measure the separated moisture by pH electrode.

⑥ $[OH^-]/40\ g \times 1000\ g = [OH^-]$ mol

⑦ $[OH^-]$ mol × moisture content of mortar

The results are given in Table 6. It may be considered that pH of the solution had changed because temperature conditions had been altered during the testing period. For example, the solution of pH 12.0 was considered a saturated calcium solution and adjustment with NaOH was not

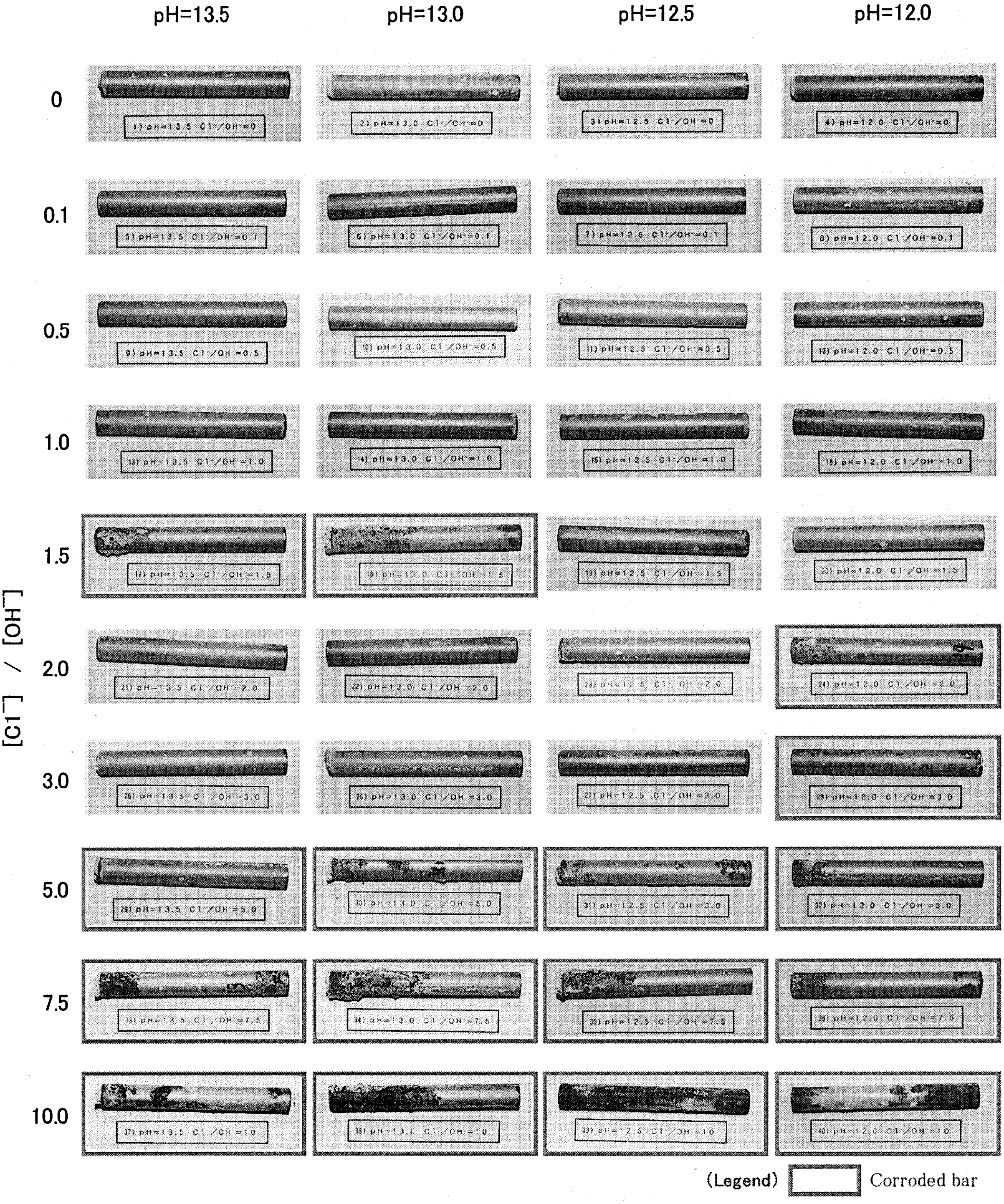

Photo 1 Corroded conditions of reinforcing bars

made. Since the pH at 20 °C was 12.6, and at 60 °C it was 11.4, pH during the experiments varied in a range of 11.4 to 12.6. However, the differences between pH values at completion of the experiments and the originally set pH value were 0.07 to 0.1, close to the value initially targeted.

Table 6 Values of pH after completion of tests

		pH			
		13.5	13.0	12.5	12.0
$[Cl^-]/[OH^-]$	1.0	13.07 (13.58)	12.46 (12.97)	12.07 (12.58)	11.56 (12.07)
	1.5	12.56 (13.07)	12.27 (12.78)	11.96 (12.47)	11.57 (12.08)
	2.0	13.10 (13.61)	12.41 (12.92)	12.18 (12.69)	11.59 (12.10)

Upper tier: measured
Lower tier: calculated

Even when NaOH had been added, it may be considered that the pH of the solution varied depending on temperature conditions. The differences between pH values at completion of experiments and initial pH values ranged from 0.03 to 0.43, indicating considerable scatter.

(3) Discussion

The testing environment ensured that specimens were in a saturated state and it may be assumed that all pores (= voids) were close to full with pore solution.

Though pore solution is consumed by the hydration reaction, since the testing environment ensured an adequate moisture supply, it was assumed that Cl^- and OH^- in the pore solution would not become concentrated. Also, although pH varied because of changing temperature, and pH values at completion showed considerable scatter, it is assumed here that the $[Cl^-]/[OH^-]$ ratio did not vary and that the initially set pH value remained unchanged.

As already noted the shaded portions of table 5 indicate specimens in which corrosion was recognized. Although comparatively on the conservative side, these results indicate that if pH is 12.0 or higher and [Cl]/[OH] is 1.0 or lower, steel does not corrode. Further, although judged as corroded in case pH was 13.5 or higher and $[Cl^-]/[OH^-]$ was 1.5, and in case pH was 13.0 and $[Cl^-]/[OH]$ was 1.5, it is possible from the conditions of adjacent specimens that the results had been of special nature, and it is considered that further studies are still required to be made. This result is considerably different from the threshold value in an alkaline solution. That is, the threshold value in mortar, at pH of 12.0 to 13.0, is higher than in an alkaline solution, while at pH of 13.5 they are approximately equal. The value is small compared with the results obtained by Yonezawa et al, and this is thought to have been because of the lack of cover at the specimens ends and the considerable influence of differences in oxygen supply conditions.

So far as can be judged from Table 5, corrosion of steel is influenced by pH, and it appears that what Glass and Buenfeld have said that it is best to express the threshold value for corrosion of steel in concrete in terms of total chloride content by weight of cement is not necessarily true.

Further, the limit value of [Cl$^-$]/[OH$^-$] was studied using polished reinforcing bars. It is known that limit values of [Cl]/[OH] differ for polished bars and bars with mill scale on their surface.The experiments in this case were carried out strictly with the aim of gaining basic knowledge, and the influence of mill scale is a matter for future study.

The absorption at the mortar portion of a specimen after testing was measured and was found to be 7.6 wt%. The unit mass of mortar was measured and it was 2,050 kg/m^3. Therefore, the absorption in mortar was

$$2{,}050\ \mathrm{kg/m^3} \times 7.6\ \mathrm{wt\%} = 155.8\ \mathrm{kg/m^3}$$

and the pores involved in absorption were approximately 15.6 vol%.

On the other hand, absorption of concrete cores sampled from actual concrete structures (number of samples:38) indicated 7 wt% (= 16.1vol%, provided that the unit mass of concrete assumed to be 2,300 kg/m^3), close to the absorption of the mortar specimens used in these experiments. Therefore, it is considered that the results obtained here can be used in evaluation of actual structures.

In measuring of moisture contents of actual structures, since it is said pore structure is altered by carbonation, sampling was done from portions not carbonated.

4. IMPROVEMENT OF CONCRETE IN ACTUAL STRUCTURES BY ELECTRO-CHEMICAL TECHNIQUES

Improvement of way concrete in an actual structure by desalination and realkalization will be described, along with the results of a follow-up survey carried out over a period of five years after treatment. Treatment was also done on adjacent structures using realkalization only. The effects of this treatment and the results of a one-year follow-up survey are described.

(1) Structures Treated

The structures tested were single-line, single-column beam-and-slab rigid-frame viaducts, as shown in Fig.3.

These viaducts were constructed at a time when regulations concerning chlorides in concrete were not clear-cut, and the chloride content was high due to the use of marine sand. As a result, carbonation had progressed.

The mixture proportions of the concrete at the time of construction were as given in Table 7. Whether admixtures had been used is not known.

Table 7 Estimated specified mix of concrete

Design strength (kgf/cm^2)	Max. size coarse agg, (mm)	Slump (cm)	Air (%)	W / C (%)	Unit content (kg/m^3)			
					Water	Cement	Fine agg.	Coarse agg.
240	25	12 ± 2	4.5 ± 1	53	157	297	699	1158

Admixture: AE water-reducing agent

Test treatments were applied to columns, beams, and cantilevered slabs. Prior to treatment, the structures were examined, detail and methods of investigation, and the results, are described below.

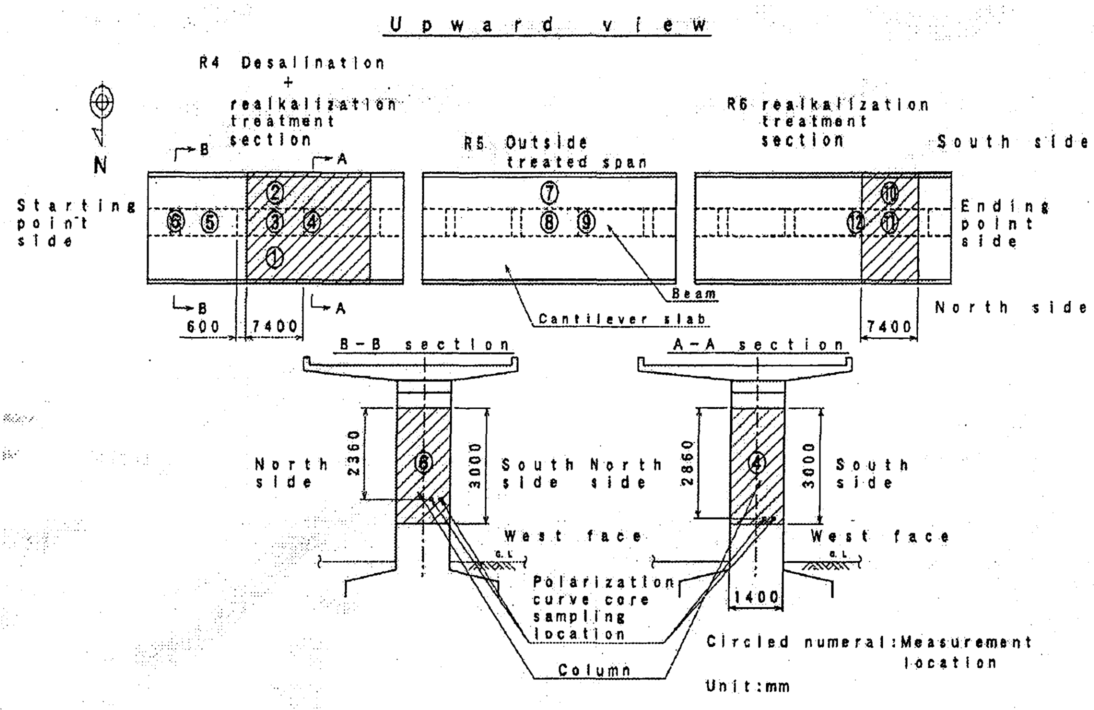

Fig. 3. Structure tested

a)Visual Inspection of Appearance

Visual inspections of overall appearance ware carried out, but no prominent cracking was recognized. Cores sampled from columns and beams were split in half longitudinally,and the fractured surfaces were visually inspected, but no cracks or leaching of gel-like substances were noted from either columns or beams. Not did coarse aggregate particles show signs cracking or reaction rings.

b) Measurement of Reinforcing Bar Corrosion, Cover, and Depth of Carbonation

Concrete cover was removed by chipping and the corroded state of reinforcing bars judged according to the grading given in Table 8. At the same time, measurements were made of cover and carbonation depth.

Table 8 Corrosion degrees

Corrosion degree	Visually observed state
0	Condition at time of construction maintained, with no subsequent corrosion noticeable
I	Corrosion in parts seen Light corrosion
II	Greater part of surface corroded Cross section broken off in parts
III	Cross section broken off over entire circumference of reinforcing bar
IV	Cross section of reinforcing bar about 1/6 to 1/3 missing

The method of measuring carbonation depth is as described below.

After chipping, any powdered concrete on the surface was removed by means of a blower or similar apparatus, and a 1% phenolphthalein solution was sprayed on the chipped surface. The distance from the concrete surface to the portion turned red was measured using calipers. Measurements were made at roughly 20 equidistant points around the circumference of the chipped portion and the average was taken as the carbonation depth. The results are given in Table 9.

Table 9 Reinforcement corrosion degree, cover, and carbonation depth investigation results

Part	Reinforcement type	Bar diameter	Corrosion degree	Cover(mm)	Carbonation depth (mm)
Column (west face)④	Main reinforcement	D32	0	62	33.6
	Hoop reinforcement	ϕ 9	II	52	
Beam (around center)③	Main reinforcement	D 32	0	44	22.1
	Stirrup	ϕ 13	II	19	

The results of carbonation depth tests carried out on sampled cores are given in Table 10.

Table 10 Core carbonation tests

Part	Carbonation depth (mm)					Av. carbonation depth (mm)
	①	②	③	④	⑤	
Column (east side/west face)	29	23	28	26	28	26.8
Beam (around center)	22	19	20	23	25	21.8

c) Chloride Content Analysis

The results of analyses of total chloride contents of the cores sampled are given in Table 11. The analysis results were determined on the hypothesis that the mass per cubic meter of concrete was 2,300 kg. Chloride analysis was performed following JCI-SC4, "Method of Analysis for Chlorides Contained in Concrete."

Table 11 Chloride quantity in concrete (kg/m³)

Part	Depth of sampling from surface (mm)					
	0 ~ 15	15 ~ 30	30 ~ 45	45 ~ 60	60 ~ 75	75 ~ 90
Column	1.00	2.51	3.30	2.85	1.90	2.06
Beam	1.06	2.51	4.53	4.00	3.01	2.54

(2) Details of Test Treatment

The test treatment consisted firstly of carrying out a process of desalination and realkalization (= desalination + realkalization), four years after which realkalization only was performed on an adjacent viaduct.

The desalination + realkalization procedure consisted of applying electric current for 60 days for desalination, then carrying out realkalization for a further 15 days. The anode for the desalination + realkalization procedure consisted of pneumatically applying fibers of cellulose nature together with the specified electrolytic solution (saturated calcium hydroxide solution) to the concrete surface, fixing a titanium mesh over this, and then applying one more layer of fibers and electrolytic solution. Maintaining the anode in a moist condition, reinforcing steel embedded in the structure was made the cathode, and direct current was applied for 60 days to achieve desalination. The current density was 1 A per square meter of concrete surface.

Realkalization was by a method called electroendosmosis in which a lithium-based alkali metal salt (lithium carbonate) is introduced from the concrete surface to the vicinity of reinforcing steel aim of restoring the alkalinity of the carbonated concrete. More specifically, alkalization was achieved by maintaining the anode used in desalination in a moist condition with a solution containing a combination sodium carbonate plus lithium carbonate solution (Li/Na mol ratio = 0.1). After applying electric current for desalination, the electric current was applied for 15 days for realkalization.

In cases where realkalization only was carried out, current was similarly applied for 15 days; however, that in this case steel mesh was used as the anode.

(3) Confirmation of Effect

In the follow-up survey, investigations of desalination + realkalization locations were carried out a total of eight times after treatment, at 3, 6, 9, 12, 18, 24, 36, and 60 months.

Where realkalization only was implemented, follow-up surveys were carried out three times, at 3, 6, and 12 months. In both cases, comparisons were made against the state before application of electric current two days after completing treatment.

a) Chloride Analysis of Concrete

Concrete cores sampled before treatment and periodically after application of electric current were used to measure the chloride content of concrete and thereby ascertain the effects of desalination.

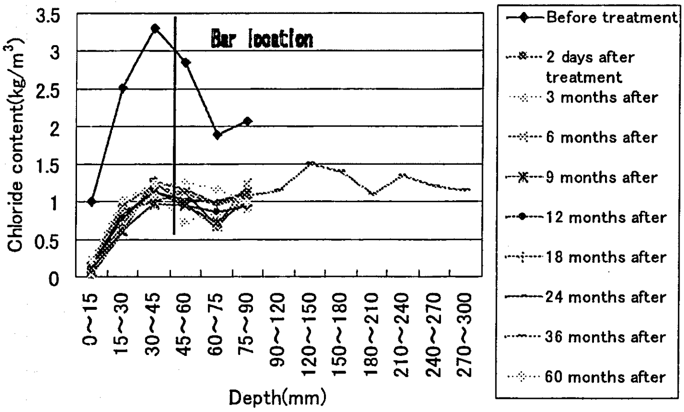

Fig. 4 Total chloride content (desalination + realkalization)column④

With regard to changes with time in chloride contents of column portions subjected to the desalination + realkalization procedure, total chloride contents are shown in Fig. 4 and soluble chloride contents in Fig. 5.

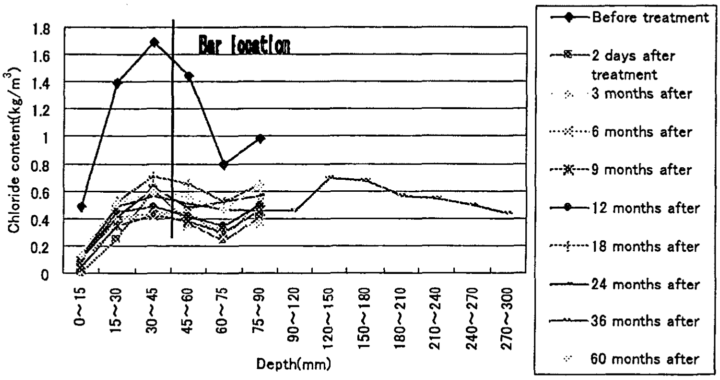

Fig. 5 Soluble chloride content (desalination + realkalization) column④

At 36 months, coring was done to column centers and measurements were made of total chloride and soluble chloride contents. Chloride content was measured according to "Method of Potential Difference Titration Using Chloride Ion Selective Electrode" in JCI-SC4, "Method of Analysis for Chloride Contained in Hardened Concrete." Significant reductions were seen in both total chloride and soluble chloride contents after treatment, while hardly any increase was seen in chloride content even at 60 months after treatment.

The broken lines in the figures indicate reinforcing bar locations where almost no rediffusion of chloride ions into the surrounding concrete took place, while it appears prominent shadow spots behind reinforcing bars were not recognizable.

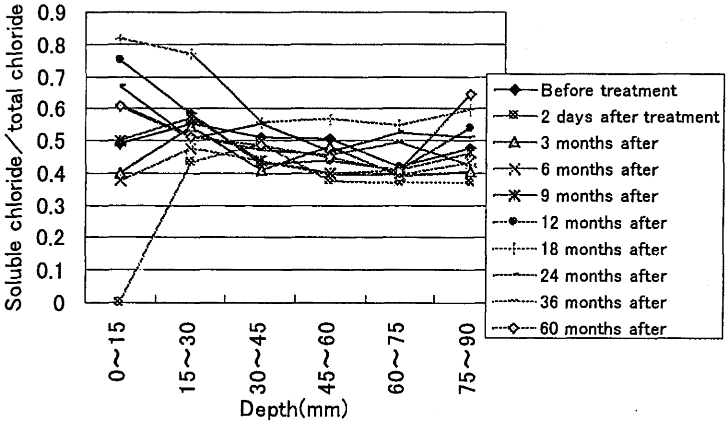

Fig. 6 Percentage of soluble chlorides (desalination + realkalization)

The ratios of soluble to total chloride contents are shown in Fig.6. Immediately after treatment, the ratio at a point 15 mm from the surface was zero, and this was due to both total and soluble chloride contents being practically zero. To around 30 mm from the surface, portions with high soluble chloride content ratios more or less overlapped with areas that had been carbonated. Apparently, chlorides existed in soluble form in portions that had been carbonated.

Below 30 mm, where carbonation had not occurred, the ratio was around 50% even after 60 months had elapsed, and no great variation was seen. The soluble chloride content was reduced by desalination treatment, but it is thought fixed chloride became soluble so the ratio of soluble chloride to total chloride did not vary to any great extent.

Regarding changes with time in chloride contents where realkalization only had been implemented, total chloride contents are shown in Fig. 7 and soluble chloride contents in Fig. 8. Compared with the situation before treatment, there was little change in either total chloride content or soluble chloride content.

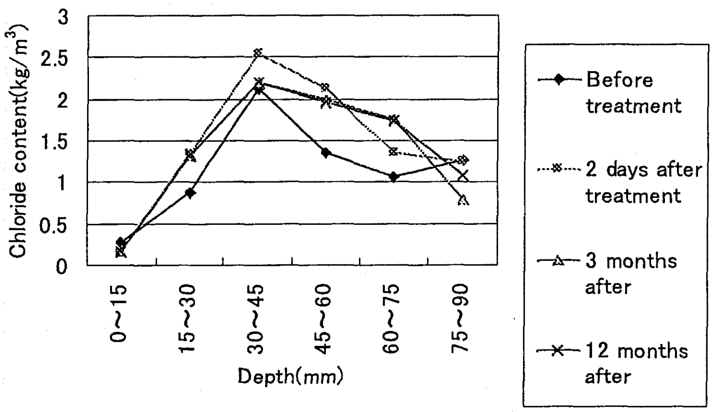

Fig. 7 Total chlorides (realkalization only)

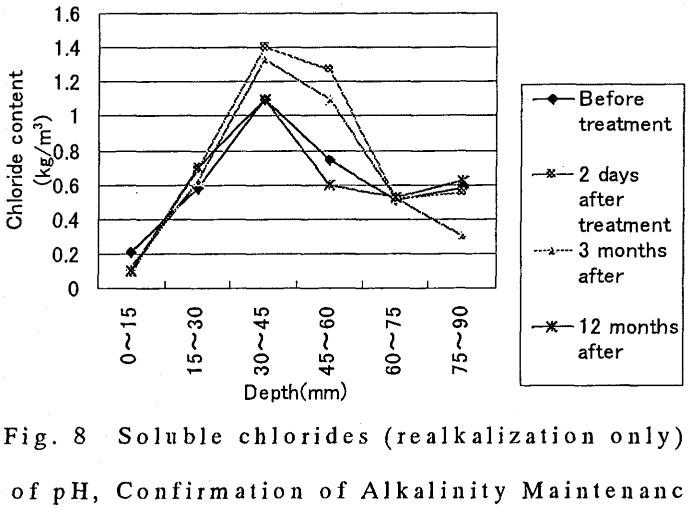

Fig. 8 Soluble chlorides (realkalization only)

b) Measurement of pH, Confirmation of Alkalinity Maintenance

Concrete cores were split, and the concrete surface sides and Reinforcement in concrete were soaked with alizarin yellow (pH test paper, pH 10.0-12.0) and alkali blue (pH test paper, pH 11.0-13.6) to judge how well concrete alkalinity was maintained after realkalizaton; note, however, that this method using pH test paper only allowed judgments in terms of ranges such as ±0.2 to 0.4. It must also be noted that even a single split surface exhibited scatter, such as for example pH = 12.8-13.6. Therefore, it was decided to express results as average pH values obtained by adding together maximum and minimum values and dividing by two. The average pH values of cores before treatment, two days after completion of treatment, and 6, 12, 24, 36, and 60 months after treatment, are shown in Fig. 9.

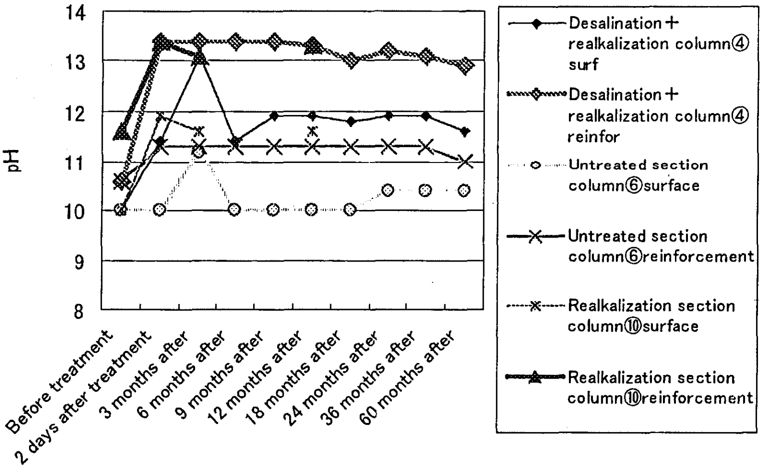

Fig. 9 Average pH of cores

At locations where desalination + realkalization was implemented, in the period between treatment and 60 months after treatment, pH values were 11.2-12.0 at concrete surfaces and 12.8-13.6 in vicinities of reinforcing bars, so it is clear that there were stable alkaline zones.

For parts that underwent realkalization only, the surroundings of reinforcement in treated parts had pH values of 13.0-13.6,12 months after treatment.

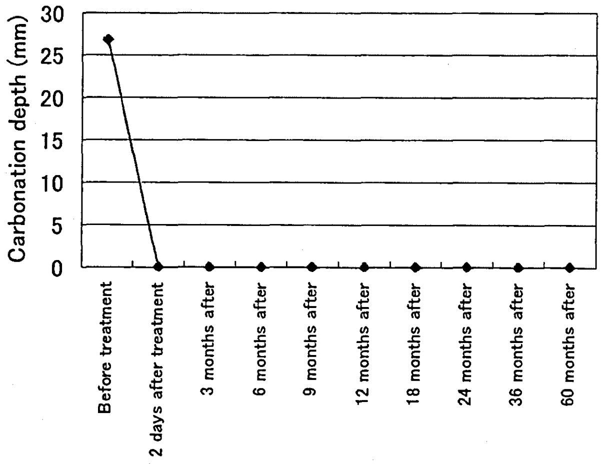

Fig.10 Results of carbonation depth measurements

Carbonation depths were measured spraying 1% phenolphthalein solution on concrete cores. The results of these measurements are shown in Fig. 10. Immediately after treatment, the carbonation depth became 0 mm, while even at 60 months after treatment, it remained 0 mm.

c) Measurement of Spontaneous Potential

Measurements of spontaneous potentials were made at the parts listed below. Copper sulfate electrodes were used as reference electrodes.

Sections treated with desalination + realkalization:
① cantilever slab, undersurface (north side)
② cantilever slab, undersurface (south side)
③ beam, undersurface
④ column, side surface
Untreated sections of the same viaduct ("untreated locations"):
⑤ beam, undersurface
⑥ column, side surface
Measured sections of adjacent viaduct ("outside treated span"):
⑦ slab, undersurface (south side)
⑧ beam, undersurface
⑨ column, side surface

Examples of spontaneous potential measurement results are shown for beam sections, in Fig. 11 for ③ , in Fig. 12 for ⑤ , and in Fig. 13 for⑧ .

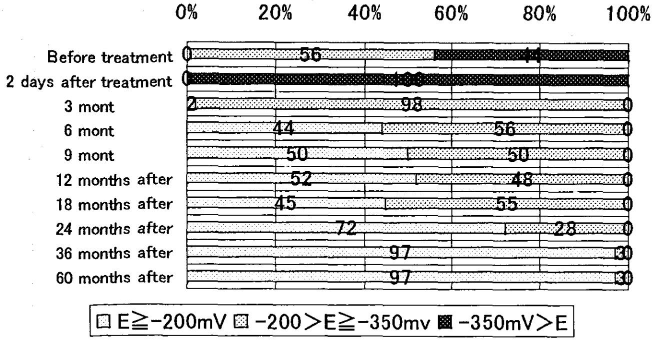

Fig. 11 Spontaneous potential (beam undersurface③)
measurement results/(desalination + realkalization)

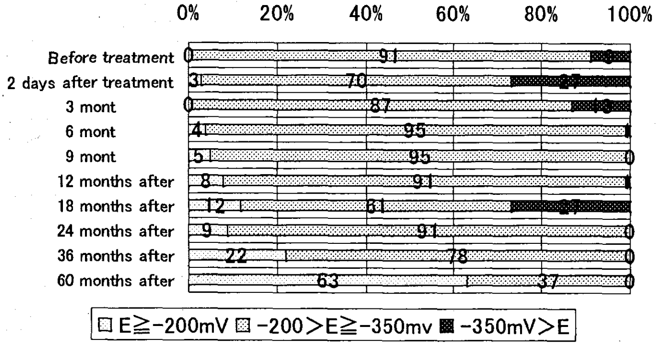

Fig. 12 Spontaneous potential (beam undersurface⑤)
measurement results/untreated part

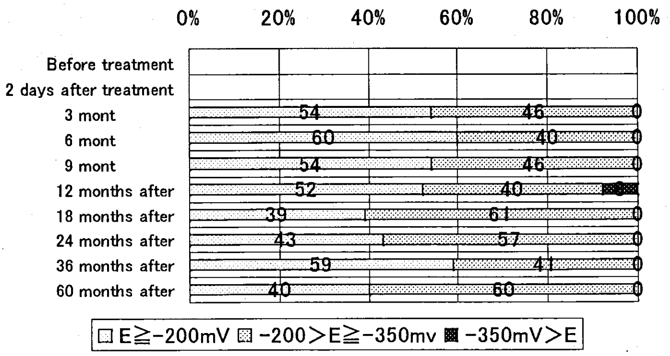

Fig. 13 Spontaneous potential (beam undersurface⑧) measurement results/outside treated span

For desalination + realkalization sections, the potential distribution 18 months after treatment was little different compared with 12 months after treatment, but the potential distributions at 24, 36, and 60 months had shifted in the noble direction.

Meanwhile, at untreated parts and outside treated spans, there was some shift in the noble direction, but as a whole, no large changes in potential were recognized.

Variations in potential at parts treated with realkalization only are shown in Fig. 14. The effect was that of realkalization having been done on parts, which were comparatively on the noble side. It can be seen that what went initially to the base side after treatment shifted later in the noble direction.

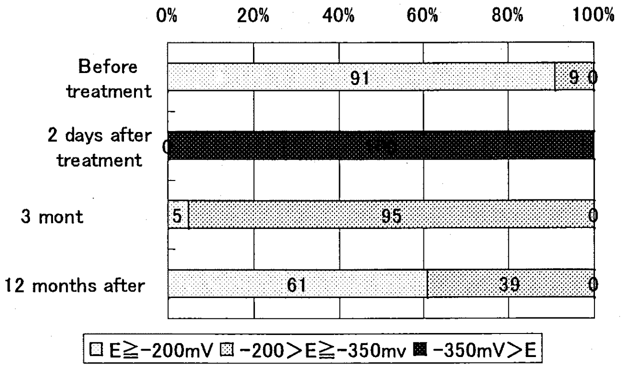

Fig. 14 Spontaneous potential (column member⑩) measurement results/realkalization only

Spontaneous potentials and corrosion probability assessments according to ASTM are given in Table 12.

Looking at Fig.13, on comparing the results of spontaneous potential measurements on the undersurfaces of beams before treatment and 60 months after desalination + realkalization treatment following ASTM C 876-87, what were 0% of Rank I in corrosion activity (E > -200 mV) or "90% or higher probability of no corrosion" were 97% 60 months after treatment. What were 56% of Rank II (-200 mV ≧ E > -350 mV), "indeterminate," were reduced to 3% 60 months after treatment.

Table 12 Spontaneous potential and corrosion probability

Corrosion activity rank	Spontaneous potential (mV) vs CSE	Corrosion probability
I	E>-200	90% or higher probability of no corrosion
II	-200 ≧ E>-350	Indeterminate
III	-350 ≧ E>-500	90% or higher probability of corrosion
IV	-500 ≧ E	Cracking in approximately one half of specimens

What were 44% of Ranks III and IV in corrosion activity (-350mV ≧ E), "90% or higher probability of corrosion," were 0% 60 months after treatment.

It can be seen that, as a whole, there was a shift to the noble side 60 months after treatment.

Similar results were obtained at other treated parts, "slab undersurface (north side), slab undersurface (south side), side surface of column," and it may be thought permissible to consider that a shift had been made to a stable spontaneous potential zone where a passive film was formed on reinforcing bars in concrete.

In contrast, with regard to undersurfaces of beams outside treated spans and side surfaces of columns, hardly any changes were seen in spontaneous potential measurement results.

d) Measurement of Anode Polarization Curve of Reinforcing Steel (Laboratory)

Anode polarization curves of reinforcing bars were measured using concrete cores with hoop reinforcement. The maximum current density Imax between +200 mV and +600 mV was determined from the spontaneous potential, and it was attempted to judge whether or not passive films are formed on reinforcing bars in concrete.

Core samples obtained from "column ④ " of the column portion of the desalination + realkalization section and "column ⑥ " of the column portion of the untreated section for comparison were used, and polarization curves of reinforcing bars were measured. The cores were collected in a form that ensured parts of hoop reinforcement would be included.

Values of Imax up to 60 months after treatment in the desalination + realkalization section and Imax up to 12 months in the realkalization only section are plotted in Fig. 15.

At 18 months after treatment the desalination + realkalization section indicated a slight increase to 17 μ A/cm^2, but by 60 months it was 5 μ A/cm^2. It is considered that a passive state had been reproduced.

Evaluation criteria for anode polarization curves are given in Table 13.

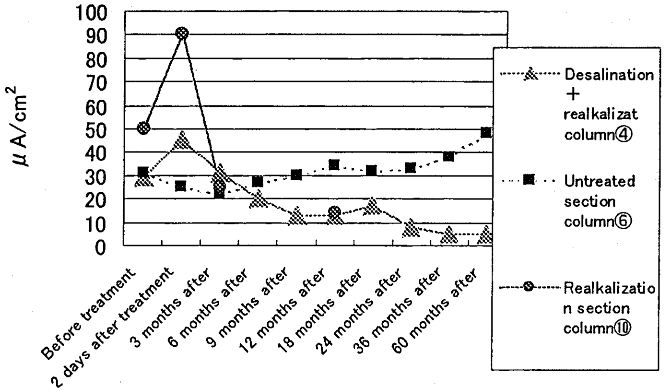

Fig. 15 Results of anode polarizatiom tests (Imax)

Imax where desalination + realkalization had been performed was 29 μA/cm^2 and in the zone of Grade 1, "Incomplete but some amount of passivity" before treatment. Contractedly, at 60 months after treatment, it was 5 μA/cm^2 and had shifted to a Grade 2 or 3 zone, in the direction of forming passive films on bar surfaces.

Table 13 Evaluation criteria for anode polarization curves〔13〕

Grade	Evaluation criterion
0	Current density exceeds 100 μA/cm^2 at least once (completely no passivity)
1	Current density 10～100 μA/cm^2 (though incomplete,some amount of passivity)
2	Current density exceeds 10 μA/cm^2 at least once, but not included in Grades 0, 1 or 3
3	Current density 1～10 μA/cm^2
4	Current density exceeds 1 μA/cm^2 at least once, but not included in Grades 0, 1, 2, or 3
5	Current density does not exceed 1 μA/cm^2 (very good passivity)

At sections of realkalization only, current density became 90 μA/cm^2, but 14 μA/cm^2 was indicated 12 months after treatment, a value roughly the same as for locations where desalination + realkalization had been performed.

The maximum current density in the anode polarization curve for untreated sections, which was 31 μA/cm^2 before treatment, was 38 μA/cm^2 60 months after treatment, indicating hardly any change. If remained in the zone of Grade 1, "Incomplete but some amounts of passivity."

Anode polarization curves of reinforcing bars were measured using concrete cores taken to include hoop reinforcement, and polarization resistance from the anode polarization curve gradient at a spontaneous potential plus 50 mV. From this, it was attempted to judge whether or not passive films would be formed on reinforcing steel in the concrete. The values of polarization resistance obtained from the polarization curve gradient before treatment and 60 months after treatment are shown in Fig.

16.

Criteria for corrosion evaluation from polarization resistance are given in Table 14.

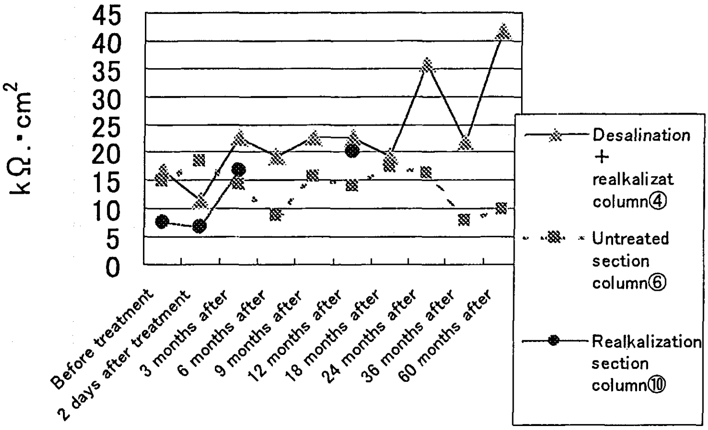

Fig. 16 Polarization resistance

Table 14 Criteria for corrosion evaluation by polarization resistance(14)

Rank	Polarization resistance	Corrosion evaluation
I	Less than4k Ω·cm^2	Zone of great reinforcement corrosion with occurrence of cracking
II	4～8k Ω·cm^2	Zone of medium reinforcement corrosion
III	8～12k Ω·cm^2	Zone of medium reinforcement corrosion
IV	More than12k Ω·cm^2	Zone of no reinforcement corrosion or zone of minute corrosion

The polarization resistance of a part where desalination +realkalization was performed, 16.7 k Ω · cm^2 before treatment, became 35.7 kΩ · cm^2 24 months after treatment but by 60 months it had declined slightly. However, it was of Rank IV, "Zone of no reinforcement corrosion or zone of minute corrosion" (12.k Ω · cm^2 or higher), and it may be considered that polarization resistance is increased by desalination + realkalization and there is a shift toward passive film formation.

At the realkalization only section, a value slightly lower than where desalination + realkalization had been carried out was indicated immediately after treatment, but 12 months after treatment it was 20 kΩ · cm^2, roughly the same as where desalination + realkalization had been done. Polarization resistance outside the treated span, which was 14.7 kΩ · cm^2 before treatment, became 7.7 k Ω · cm^2 60 months after treatment, Rank II, "Medium reinforcing steel corrosion zone" (4-8 kΩ · cm^2).

(4) Summary

The following summarized the effects of test application of desalination + realkalization and realkalization only treatments on actual structures:

a) Application of Desalination + Realkalization

1) Evaluations of corrosion activity of reinforcing steel based on spontaneous potential, anode polarization curves, and polar-ization resistance, indicate that, for the period up to 60 months after treatment, passive films tended to be formed on reinforcing steel.

2) The alkalinity of the concrete after treatment, as measured using pH test paper and 1% phenolphthalein solution, was found to be stable even 60 months after treatment.

3) The chloride distribution in the concrete after treatment indicated that there was no rediffusion of chloride ions to the vicinity of reinforcement even 60 months after treatment.

4) The ratio of soluble chlorides to total chlorides remained except at parts extremely near the surface, around 50% even after 60 months, and no major changes were noted.

5) No clear shadow spots could be seen behind reinforcing bars.

b) Application of Realkalization Only

1) Anode polarization curves and polarization resistance 12 months after treatment were roughly equivalent to cases where desalination + realkalization was applied.

2) In vicinities of reinforcing bars in treated parts 12 months after treatment pH values of 13.0-13.6 were indicated.

5.COMPARISON OF LABORATORY TEST RESULTS AND MEASUREMENTS ON ACTUAL STRUCTURES

Desalination and realkalization were evaluated through a comparison of laboratory test results and measurements on actual structures.

Changes in pH and Cl^-/OH^- after application of desalination + realkalization are summarized in Table 15. Specific values of pH could not be identified because of problems with measurement techniques, but the application of desalination + realkalization resulted in pH values around reinforcement being improved to 12.6-13.2 60 months after treatment, while soluble chlorides were also reduced to 0.47-0.56kg/m^3. Considering pores in concrete as amounting to 16.1vol% of total volume, [Cl]/[OH] would be between 1.28 and 5.49.

Table 15 Changes in pH and $[Cl^-]/[OH^-]$ due to application of desalination + realkalization

	Depth (mm)	pH	Total chlorides (kg /m^3)	Soluble chlorides (kg /m^3)	[Cl-] / [OH-]
Before treatment	45 ~ 60	10.4 10.8	2.85 (0.96)	1.44 (0.48)	790.8 ~ 1988.0
	60 ~ 75	10.4 10.8	1.90 (0.64)	0.80 (0.27)	526.1 ~ 1322.7
60months after	45 ~ 60	12.6 13.2	1.25 (0.42)	0.56 (0.19)	1.38 ~ 5.49
	60 ~ 75	12.6 13.2	1.16 (0.39)	0.47 (0.16)	1.28 ~ 5.10

Note) pH values for reinforcement parts in treated portion ④
Calculation of $[Cl^-]/[OH^-]$ performed for total chlorides with quantity of pores in concrete taken as 16.1 vol% of total volume.
Figures in ( ) ratios (%) of chloride ion quantity to unit cement content

The conclusion drawn from laboratory tests carried out in tandem with treatment tests on structures based on the fact that moisture contents of mortar used in laboratory tests and concrete cores collected from actual structures were approximately equal was that steel does not corrode when pH is 12 or higher and $[Cl^-]/[OH^-]$ is 1.0 or under. If this condition were to be applied without modification, it would be satisfactory with regard to pH, but not with regard to $[Cl^-]/[OH^-]$.

Changes in pH and $[Cl^-]/[OH^-]$ after application of realkalization only are summarizedin Table 16. Through application of realkalization only, pH in the vicinity of reinforcement was improved to a range of 13.0-13.6, while $[Cl^-]/[OH^-]$ was between 0.76 and 3.41.

Table 16 Changes in pH and $[Cl^-]/[OH^-]$ due to application of realkalization only

	Depth (mm)	pH	Total chlorides (kg /m^3)	Soluble chlorides (kg /m^3)	[Cl] / [OH]
12 months after treatment	45 ~ 60	13.0 13.6	1.95 (0.66)	0.60 (0.20)	0.86 ~ 3.41
	60 ~ 75	13.0 13.6	1.74 (0.59)	0.53 (0.18)	0.76 ~ 3.04

Note) pH values for reinforcement parts in treated portion ⑩
Calculation of $[Cl^-]/[OH^-]$ performed for total chlorides with quantity of pores in concrete taken as 16.1 vol% of total volume.
Figures in ( ) ratios (%) of chloride ion quantity to unit cement content.

In this case, the conditions for preventing steel corrosion are satisfied with respect to pH, while with regard to $[Cl^-]/[OH^-]$ also, there is a possibility that it will be not more than 1.0. The reduction in chloride ions and the increase in pH (shift in strong alkali direction) both contribute to the fall in [Cl]/[OH], but since pH is logarithmic, the contribution due to rise in pH is claminant.

The relationship between pH and total chloride content for [Cl]/[OH] values of 1.0 and 1.3 and with pH in the range 12.6-13.6 as obtained in treatment is shown in Fig. 17. With the threshold value of reinforcing steel corrosion

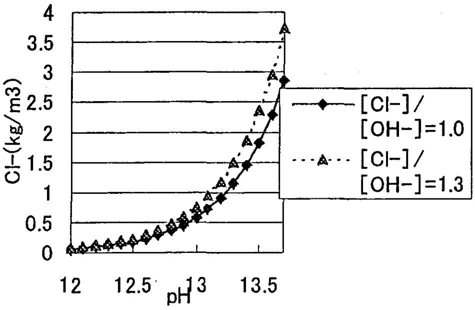

Fig. 17 Corrosion threshold value

$[\mathrm{Cl}^-]/[\mathrm{OH}^-] \leqq 1.0$, as obtained from the results of laboratory tests, the part below the solid line in the figure is the range which may be considered "not corroded."

Considering that an actual structure shift toward formation of a passive film after application of desalination + realkalization if up to $[\mathrm{Cl}^-]/[\mathrm{OH}^-] \leqq 1.3$ is allowed on the threshold from Table 14, the part below the broken line in the figure is the range, which may be considered "not corroded."

The past research summarized in Chapter 2 indicates that the threshold value of $[\mathrm{Cl}^-]/[\mathrm{OH}^-]$ for corrosion of reinforcing steel in cement mortar noted by S. E. Hussain et al. [10] at a pH of less than 13.30 is in a range of 1.7-2.0, while for pH greater than 13.30, the range is 1.28-1.86. In Fig. 18, based on these values, and with a pore volume of 16.1 vol%, the relationship with total chloride is shown, further superimposing Fig. 17.

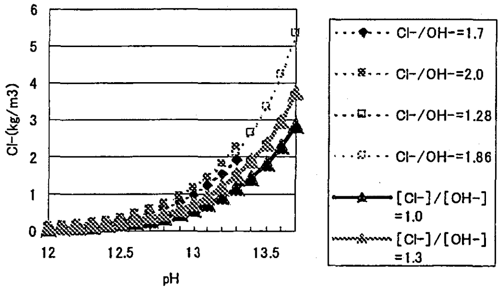

Fig. 18 Corrosion threshold values according to S.E. Hussain et al. and according to this study

It can be seen that even with $[\mathrm{Cl}^-]/[\mathrm{OH}^-] \leqq 1.3$, a relatively conservative value is obtained.

6. CONCLUSIONS

This investigation leads to the following conclusions:

(1) Experiments with mortar indicate that if pH is 12 or higher and $[\mathrm{Cl}^-]/[\mathrm{OH}^-]$ is 1.0 or less, steel reinforcement will not corrode. So far as judged by the results of these experiments, whether corrosion occurs or not

steel is greatly influenced by pH. From the results of experiments on actual structures, it is concluded that steel will not corrode if [Cl]/[OH] is 1.3 or under. This result is in conformity with results of past research.

(2) By carrying out desalination + realkalization, pH values around reinforcing steel in structures were improved to 12.6-13.2 60 months after treatment, while soluble chlorides were reduced to 0.47-0.56 kg/m^3 . Even 60 months after treatment, practically no changes were noted in the alkalinity and chloride content of the concrete, while there was hardly any rediffusion of chloride ions around reinforcing bars. No prominent shadow spots were seen behind reinforcing bars either.

(3) The ratio of soluble chlorides to total chlorides was estimated to be about 50% after the elapse of 60 months, except extremely close to the surface, and no large variations were seen.

(4) Where realkalization only was carried out as treatment, after treatment, pH values around reinforcement in structures had improved to 13.0-13.6,12 months later, while $[Cl^-]/[OH^-]$ values were between 0.76 and 3.41. Hence, there is a possibility that the threshold $[Cl^-]/[OH^-]$ value will be satisfied through application of realkalization only.

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