

EFFECT OF THE DISTANCE BETWEEN ELECTRODES ON THE PERFORMANCE OF USING ELECTROCHEMICAL TECHNIQUE TO REPAIR THE CONCRETE DAMAGED BY AAR

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Abstract

This research is to study the effect of the distance between electrodes on the performance of the AAR damaged concrete repaired by using the rapid lithium migration technique (RLMT). Cylindrical concrete specimens made by reactive sandstone of $\varnothing 100\text{mm}$ with 50, 100, 150 and 200 mm high were prepared, and cured for 28 days in fog. RLMT was conducted using LiOH and $\text{Ca}(\text{OH})_2$ as electrolytes for anode and cathode, respectively. A current density of 9 A/m^2 was used to drive lithium into and to sodium out of the concrete. The results showed that the proportion of the sodium ion removed from the specimen and the rate of ions migration were decreased, and the time for lithium passing through the specimen was increased with increasing the distance between electrodes. The performance of RLMT to repair concrete damaged by AAR has accomplished the stage of non-steady-state migration of Li^+ . The transport of Li^+ in electrolyte in an electrical field as driving force does apparently not affect the non-steady-state diffusion coefficient of Li^+ .

KEYWORDS: alkali aggregate reaction, concrete, electrode, electrochemical technique, ion migration

1 INTRODUCTION

Lithium-containing compounds are frequently studied as the alkali-aggregate reaction (AAR) inhibiting admixtures. Proper application of lithium compounds is effective against expansion owing to AAR [1~16]. The surface charge of lithium ion is stronger than that of sodium and potassium ions, so the sodium and potassium ions in Na(K)-S-H gel are replaced by lithium ions, thus forming Li-S-H with properties of strong bonding and insolubility [2, 3, 5, 17].

To mix lithium salts in concrete to prevent AAR damage is convenient in construction. While the traditional methods of soaking or pounding for AAR damaged concrete are effective in preventing further expansion of existing structures, unfortunately they are time consuming [18].

The concept of using electrochemical technique to drive lithium ions into hardened concrete was first developed in 1992 by Page [19]. During the same period, Stark et al. proposed that lithium was required to deal with the electrochemical removal of chloride from concrete structures [9]. Subsequently, an electrochemical trial in Virginia showed that it is feasible to drive lithium into structures to remedy AAR [20, 21].

The authors proved the performance using an electrochemical technique with auxiliary anode and cathode that drove lithium ions into and removed sodium ions from mortar bars. The results demonstrated excellent effects of inhibiting the expansion due to AAR using LiOH or LiNO_3 as anolyte and $\text{Ca}(\text{OH})_2$ as catholyte, and moreover revealed that the rate of Li^+ ions being driven into and Na^+ ions being removed from the specimen increases with the alkali content of the cement [22].

This study proposes a rapid lithium migration technique (RLMT). The RLMT using electric characterization of anion moves toward anode and cation moves toward cathode to drive lithium ions of anolyte into the concrete specimen, and also to remove Na^+ ions from the concrete specimen into

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catholyte to increase the effect of inhibiting expansive AAR. Changes of the voltage, the time for lithium ion passing through the specimen, and the variation of sodium and lithium ion concentrations in catholyte were measured during the treatment. The distributions of sodium and lithium ions in the specimen were analyzed after the treatment. The expansive behavior of the concrete specimens cured at 38° and 100% RH for one year after the RLMT treatment was investigated to evaluate the feasibility of using RLMT to repair concrete damaged by AAR.

2 MATERIALS AND METHODS

2.1 Materials and mix proportions

Table 1 lists the chemical compositions of type I cement used in this work. The mix proportions of concrete specimens are determined in accordance with ACI 318 Code with an alkali content of 1.25wt% $\text{Na}_2\text{O}_{\text{eq}}$ adjusted by the addition of NaOH, as shown in Table 2.

The sandstone aggregate used in this study was obtained from the eastern area of Taiwan. The aggregate was tested using the ASTM methods, C289, C1260, C227 and C1293, and all of the results reveal that the aggregate is highly reactive. The aggregate was crushed, washed and graded as required by the ASTM C1293 test. The NaOH and $\text{LiOH}\cdot\text{H}_2\text{O}$ used in this study were both reagent grade.

2.2 Rapid lithium migration technique (RLMT)

Figure 1 shows the RLMT test arrangement used in this study. The test uses a thick concrete disc between two electrodes (#20 mesh, 304 stainless steel) in an arrangement similar to the ASTM C1202 test (rapid chloride permeability test - RCPT). Both the RLMT and RCPT methods invoke a similar one-dimension electrical field.

2.3 Test procedures

Concrete cylinders $\text{Ø}100\times 200$ mm were demoulded after 24 hours and cured in a fog room at 23° for 28 days, and then the specimens were cut with the height of 5, 10, 15 and 20 cm, for proceeding the RLMT test. An electrical field of constant current density of 9 A/m² was applied between the electrodes, and the test was terminated for 30 days. Additionally, concentrations of sodium and lithium ions in the catholyte were measured during treatment, the distributions of the related ions in the concrete specimens were also measured after the RLMT treatment. The expansive behavior of the concrete specimens, cured at 38°C and 100% RH for one year following the RLMT treatment, was investigated to evaluate the future performance in inhibiting the AAR expansion.

3 RESULTS AND DISCUSSION

3.1 Applied voltage for maintaining constant current density

For concrete specimens with the identical mix proportion, Table 3 reveals that the applied initial voltage required to maintain a constant current density for different distances between electrodes in the RLMT test, is directly proportional to the distance between electrodes involved. For any distance between electrodes, the applied voltage to maintain constant current density always increases quite substantially with the time of the RLMT treatment. Figure 2 shows that white crystals appeared on the surface of 5 cm height specimen in the cathode after the RLMT test. However, the white crystals did not appear on the surface of the taller specimens after tests. The result of XRD had proved the white crystals were CaCO_3 . Infer the increasement of applied voltage during the RLMT test from the pores of specimen being blocked by CaCO_3 in chief.

3.2 Removal of Na^+ from specimen

Table 4 reveals that the remained amount of $\text{Na}_2\text{O}_{\text{eq}}$ for 50, 100, 150, and 200 mm high specimens after the RLMT treatment was 0.16wt% $\text{Na}_2\text{O}_{\text{eq}}$, 0.30% $\text{Na}_2\text{O}_{\text{eq}}$, 0.53wt% $\text{Na}_2\text{O}_{\text{eq}}$, and 0.70wt% $\text{Na}_2\text{O}_{\text{eq}}$ (ie. 0.62 kg/m³, 1.20 kg/m³, 2.10 kg/m³, and 2.79 kg/m³, respectively), all lower than the limit of 3 kg/m³ $\text{Na}_2\text{O}_{\text{eq}}$ defined in CSA A23.1. Figure 3 shows that the amount of removal of Na^+ from specimen increased with the time of treatment. If the retained amount of 3 kg/m³ $\text{Na}_2\text{O}_{\text{eq}}$

is the aim of treatment, the amount of Na⁺ removed from 50, 100, 150, and 200 mm high specimens is 0.420 g, 0.840 g, 1.260 g, and 1.680 g, respectively, for treatment extended over 6 days, 12 days, 18 days, and 24 days, respectively.

For the control specimen, 63% of total Na⁺ in the pore solution of the untreated specimen was measured using the water-soluble method, that is, 37.0% of total Na⁺ was fixed in the hydration product of the control specimen. Table 4 shows that the amount of Na⁺ fixed in the hydration product of the 50, 100, 150, and 200 mm high specimens after the RLMT treatment was reduced to 4.7%, 11.2%, 26.3%, and 34.8%, respectively. This finding shows that the Na⁺ is partly removed from the hydration product during the RLMT treatment, simultaneously. Furthermore, for specimens are treated with the same time, the proportion of Na⁺ removed from the hydration product decreases with the height of specimen. Figure 4 shows that Li⁺ may have replaced the Na⁺ of partly removed from the hydration product during the RLMT treatment.

For longer specimens, Table 3 reveals that maximizing the number of Na⁺ ions removed from the specimen by increasing the treated time was not easy because of the resistance of specimen increases too much. In practical applications, this problem limits the distance between electrodes for using the RLMT treatment to repair concrete damaged by AAR.

3.3 Migration of Li⁺ into specimen

Although all specimens have similar properties of pore and ionic concentrations in the pore solution, based on the percolation ideas [24], the pore connectivity of the longer specimen is less than those of the shorter specimen. Therefore, the specimen height is directly proportional to the time required for the Li⁺ ions to pass through the specimen, for example, the required time for the specimen of 20 cm height is four times that for the specimen height of 5 cm shown in Table 5, but the increase rate of Li⁺ ion migration into the catholyte is significantly reduced for the longer specimen.

Table 5 illustrates that increase in specimen height impedes the process of driving Li⁺ ions into and remove Na⁺ ions from hardened concrete. Therefore, driving of lithium ions into hardened concrete for the longer specimens was less than that for the shorter specimens during the same treatment time. The table also reveals that the average Li/Na molar ratio decreases with the specimen height. Furthermore, for the specimens with height of 20 cm after 30 days of treatment, the average Li/Na molar ratio is increased to 2.09 exceeding value of effective inhibition of 0.7, and average alkali content is reduced to 0.70% Na₂O_{eq}.

Figure 5 shows a very good linear relationship between the average Li/Na molar ratio and the distance between electrodes, with a regression coefficient R² of 0.9743.

3.5 Relationship of distance between electrodes and migration rate of Li⁺

Figure 3 reveals that the flux of Li⁺ has not reached steady-state yet. The results of section 3.4 confirm that the performance of using RLMT to repair concrete damaged by AAR has accomplished on the stage of non-steady-state migration of Li⁺. Therefore, the non-steady-state diffusion coefficient of Li⁺ plays an important role for evaluating the required time of RLMT treatment.

The analytical solution of non-steady-state diffusion coefficient

From natural diffusion tests, Li⁺ diffusion coefficient can be calculated according to the second Fick's law as [24,25]:

$$\frac{dC}{dt} = D \left(\frac{d^2C}{dx^2} - \frac{dC}{dx} \right) \quad (1)$$

where C is concentration of ions (in mol/l) as a function of distance x, at time t, and with D as diffusion coefficient.

The diffusion coefficient D_n which applies to RLMT in non-steady-state can be calculated from the modified second Fick's law [24,25]:

$$\frac{dC}{dt} = D_n \left(\frac{d^2C}{dx^2} - \frac{|z|FE}{RT} \frac{dC}{dx} \right) \quad (2)$$

where D_n is the non-steady-state diffusion coefficient (dimension: m^2/s); z the electrical charge of Li^+ ; F the Faraday constant (96500 C/mol); E the strength of the electric field between the anode and cathode (V/m); R the universal gas constant (8.3144 J/mol/K); and T the absolute temperature (K). For RLMT, the following conditions are used to obtain the solution of Eq. (2), with initial conditions: $C = 0$, $x > 0$, $t = 0$; boundary condition: $C = 0$, $x \rightarrow \infty$, and t is very large (ie. long duration). The analytical solution for Eq. (2) is

$$C(x,t) = \frac{C_0}{2} \left[e^{\alpha x} \operatorname{erfc}\left(\frac{x + aD_n t}{2\sqrt{D_n t}}\right) + \operatorname{erfc}\left(\frac{x - aD_n t}{2\sqrt{D_n t}}\right) \right] \quad (3)$$

where $\beta = \frac{|z|FE}{RT}$, C_0 is Li^+ concentration in the source cell (anode cell), and erfc is the complementary error function.

From Eq. (3), the non-steady-state diffusion coefficient can be presented as:

$$D_n = \frac{1}{\beta} \left[\frac{x - \alpha\sqrt{x}}{t} \right] \quad (4)$$

where $\alpha = 2\sqrt{\frac{1}{\beta} \operatorname{erf}^{-1}\left(1 - \frac{2C}{C_0}\right)}$, and erf^{-1} is the inverse error function.

The experimental result of non-steady-state diffusion coefficient

In non-steady-state condition, the Li ions have not yet reached the cathode cell. The non-steady-state diffusion coefficient was determined based on measurements of time-span (T_s) for Li^+ penetration through specimen until the Li^+ concentration significant increase in the cathode cell. Since the Li^+ concentration was not continually monitored, a value of $C/C_0 = 0.005$ was used in all cases [24~26]. The non-steady-state diffusion coefficient (D_n) obtained from RLMT is calculated from Eq. (4), and listed in Table 7.

Table 7 shows that the non-steady-state diffusion coefficient D_n of Li^+ is relate to the properties of specimens and alter the distance between electrodes do not affect apparently the D_n of Li^+ , with a range of D_n $4.69 \times 10^{-12} m^2 s^{-1} \sim 9.91 \times 10^{-12} m^2 s^{-1}$.

4 DISCUSSION

According to the previous discussion, the RLMT using electric characterization to drive Li^+ into and remove Na^+ from concrete to increase the effect of inhibiting expansive AAR. The expansive behavior of the concrete specimens cured at $38^\circ C$ and 100% RH for one year after the RLMT treatment shows that the RLMT treatment is feasible for repairing the concrete damaged by AAR.

The height of specimen, that is, the distance between the electrodes, affects the migration results of the related ions in the concrete for the RLMT treatment, and the performance of the treatment then is also influenced by the migration results. From the above discussion, the performance of the RLMT treatment increases with reducing average alkali content and increasing Li/Na molar ratio in the treated concrete.

In the initial stage of RLMT treatment, the concentration of $Na^+(K^+)$ in the pore solution of specimen is so high that inhibiting the existence of Ca^{2+} , and then $Na^+(K^+)$ decreases and Ca^{2+}

increases in the pore solution with the time of RLMT treatment, and CO_2 in the air is dissolved into catholyte formed CO_3^{2-} , simultaneously. Because of Ca^{2+} move to the negative electrode and CO_3^{2-} to the positive one the CaCO_3 crystals block pores of specimen gradually. Therefore, the application of the RLMT treatment must notice that CaCO_3 crystals would block the pores of specimen with time so that the applied voltage for maintaining constant current density increases gradually. In addition, how to avoid CO_2 dissolving into the catholyte during the process of the RLMT treatment is worth further research.

The longer electrodes distance increases the resistance, the applied voltage for maintaining constant current density and the time required for the Li^+ ions to pass through the specimen both are directly proportional to the distance between electrodes, therefore, using the RLMT treatment limits the maximum distance between electrodes. Although both longer and shorter specimens have similar properties of pore and ionic concentrations, based on the percolation ideas, the pore connectivity of the longer specimens is less than those of the shorter specimens, but transport of Li^+ in electrolytes due to the action of an electrical field as driving force do not affect apparently the D_n of Li^+ .

The results of this paper confirm that the performance of using RLMT to repair concrete damaged by AAR has accomplished on the stage of non-steady-state migration of Li^+ . Therefore, the non-steady-state diffusion coefficient of Li^+ plays an important role for evaluating the required time of RLMT treatment. Furthermore, the result shows that the non-steady-state diffusion coefficient D_n of Li^+ is not affect apparently by the electrode distance. Hence, before conducting the extensive RLMT treatment in the field, it is useful to use the RLMT model proposed in this study to obtain the D_n of Li^+ for several heights of specimens, Li^+ in every portion of the specimen at any time can derive from Eq. (3), and then we can obtain the related parameter for designing the RLMT treatment.

5 CONCLUSIONS

- (1) The expansive behavior of the concrete specimens cured at 38°C and 100% RH for one year after the RLMT treatment shows that the RLMT treatment can repair the concrete damaged by AAR.
- (2) For any electrode distance, because of CaCO_3 blocks pores the applied voltage required to maintain constant current always increases with the time of the RLMT treatment.
- (3) For different electrode distance in the RLMT test, based on the percolation ideas, the applied initial voltage to maintain constant current is directly proportion to the electrode distance. Therefore, the maximum distance of the electrodes is limited using the RLMT treatment.
- (4) The distance between electrodes is directly proportion to the time required for Li^+ ions to pass through the specimen, but the rate of increase of Li^+ ions migrated into the catholyte for longer electrode distance is significantly reduced.
- (5) The performance of using RLMT to repair concrete damaged by AAR has accomplished on the stage of non-steady-state migration of Li^+ .
- (6) The transport of Li^+ in electrolytes due to the action of an electrical field as driving force is not affect apparently the D_n of Li^+ .
- (7) Before conducting the extensive RLMT treatment in the field, it is useful to use the RLMT model proposed in this study to obtain the D_n of Li^+ and the related parameter for designing the RLMT treatment.

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TABLE 1: Chemical composition of the cement

Oxide	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	Na ₂ O	K ₂ O	LOI	Na ₂ O _{eq}
wt%	20.86	4.80	3.71	63.50	3.82	1.92	0.18	0.50	0.63	0.51

TABLE 2: The mix proportions of concrete

ingredient	weight (kg/m ³)
cement	396.55
water	238.15
coarse aggregate	786.51
fine aggregate	844.23
Na ₂ O _{eq}	4.96

TABLE 3: Applied voltage for maintaining constant current density for different distances between electrodes in the RLMT test (V)

distance between electrode (cm)	time (day)						
	0	6	9	12	18	24	30
5	8.6	10.0	10.3	11.0	13.7	55.5	93.1
10	17.1	17.7	18.8	20.4	21.1	51.7	77.2
15	25.1	27.7	31.3	30.6	36.4	81.8	93.0
20	34.5	35.4	39.0	42.7	40.7	65.8	81.5

TABLE 4: Amount of the Na⁺ removed from specimens

specimen height (cm)	amount of Na ⁺ ions in the specimen before treatment (g)	amount of Na ⁺ ions removed from the specimen after treatment (g)[percentage, %]	amount of Na ⁺ ions in pore solution of the specimen after treatment* (g)[percentage, %]	amount of Na ⁺ ions was fixed in the hydration product of the specimen after treatment (g)[percentage, %]	average in the specimen after treatment (% Na ₂ O _{eq}) [kg/m ³]
5	1.062	0.929 [87.5]	0.083 [7.8]	0.050 [4.7]	0.16 [0.62]
10	2.125	1.610 [75.8]	0.276 [13.0]	0.238 [11.2]	0.30 [1.20]
15	3.187	1.840 [57.7]	0.507 [16.0]	0.839 [26.3]	0.53 [2.10]
20	4.250	1.863 [43.8]	0.908 [21.4]	1.477 [34.8]	0.70 [2.79]

* The 63.0% of the Na⁺ ions in the pore solution of the untreated specimen were measured using the water-soluble method.

TABLE 5: Migration results of Li⁺ ions for different height of specimen

specimen height (cm)	required time for Li ⁺ ions pass through the specimen (day)	contents of the related ions in the concrete specimens (kg/m ³)		average Li/Na molar ratio
		Li ⁺	Na ⁺	
5	6	0.957	0.211	10.16
10	12	0.603	0.351	6.30
15	18	0.374	0.431	4.56
20	24	0.381	0.578	2.09

Table 6: The vertical expansions of the specimens cured at 38°C and 100% RH for one year following the RLMT treatment (%)

Specimen	distance between electrodes (cm)	5	10	15	20
	RLMT treatment		-0.252	-0.090	0.004
control		0.289	-	-	0.836

Table 7: Non-steady-state diffusion coefficient of Li⁺

distance between electrodes (cm)	T _s (day)	average voltage (V)	$\beta = \frac{ z FE}{RT}$	$\alpha = 2\sqrt{\frac{1}{\beta}} \operatorname{erf}^{-1} \left(1 - \frac{2C}{C_0}\right)$	diffusion coefficient D _n (×10 ⁻¹² m ² s ⁻¹)
5	13.0	9.96	7771.81	0.04084	4.69
10	19.0	19.27	7518.21	0.04152	7.04
15	19.5	31.06	8078.73	0.04005	9.91
20	30.0	48.52	9465.07	0.03700	7.48

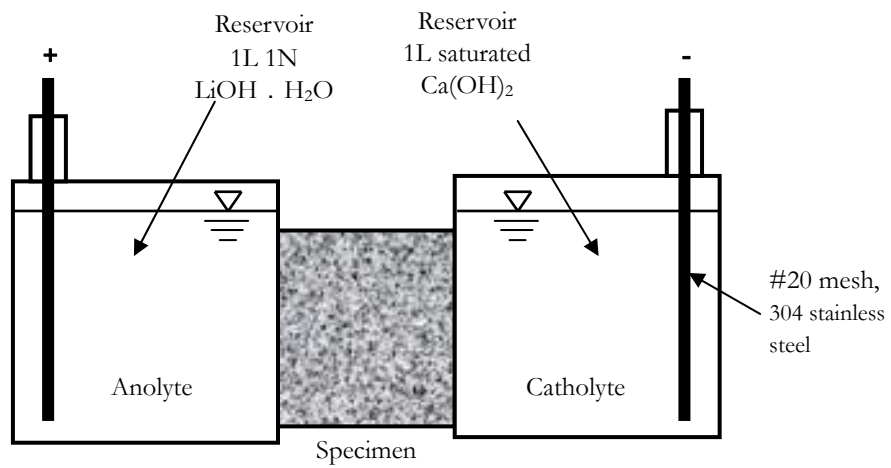


Figure 1: Migration cell of the RLMT test



Figure 2: White crystals of CaCO_3 appear on the surface of 5cm height specimen in the cathode after the RLMT test

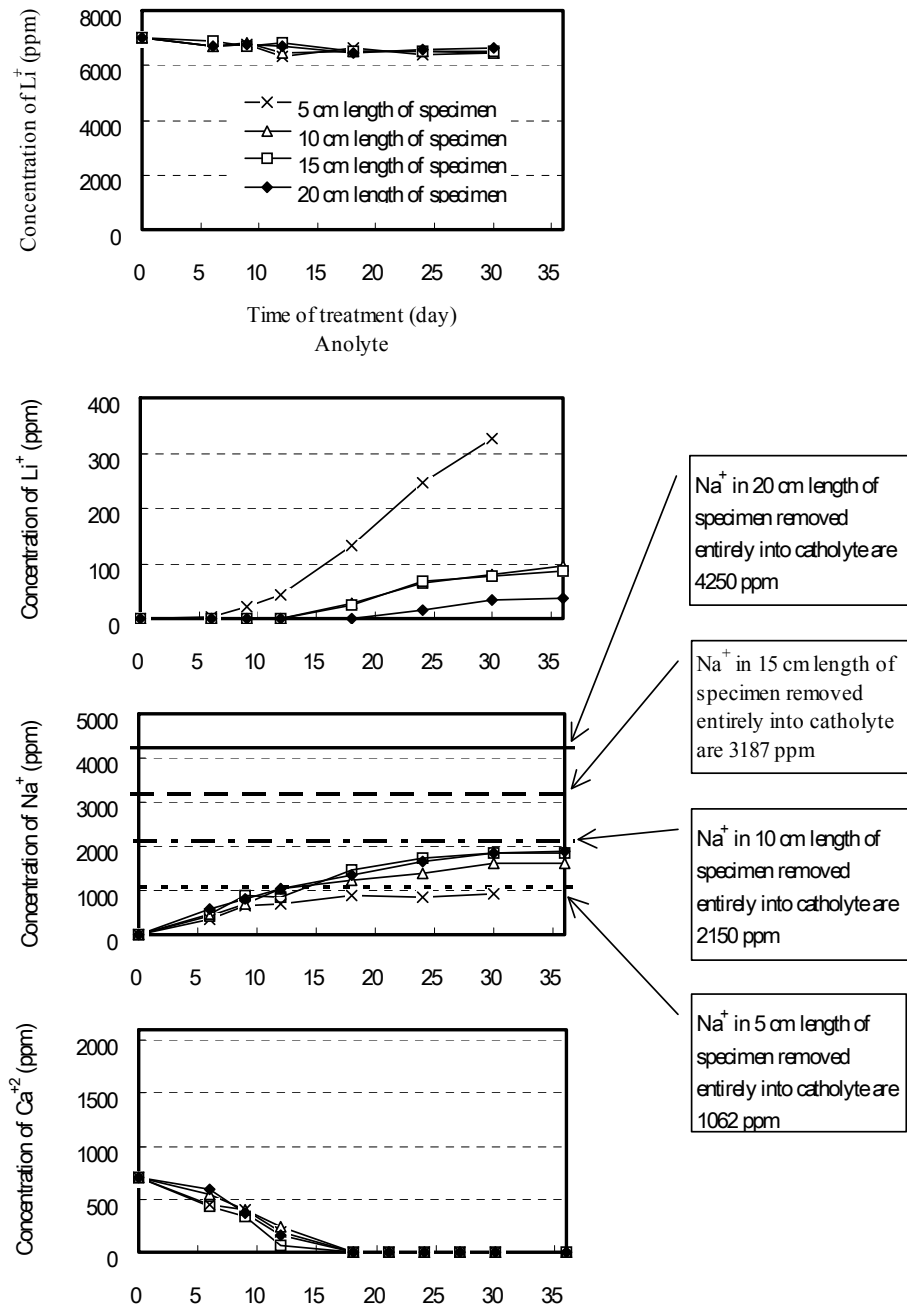


Figure 3: Concentrations of the related anions in electrolytes during the RLMT treatment

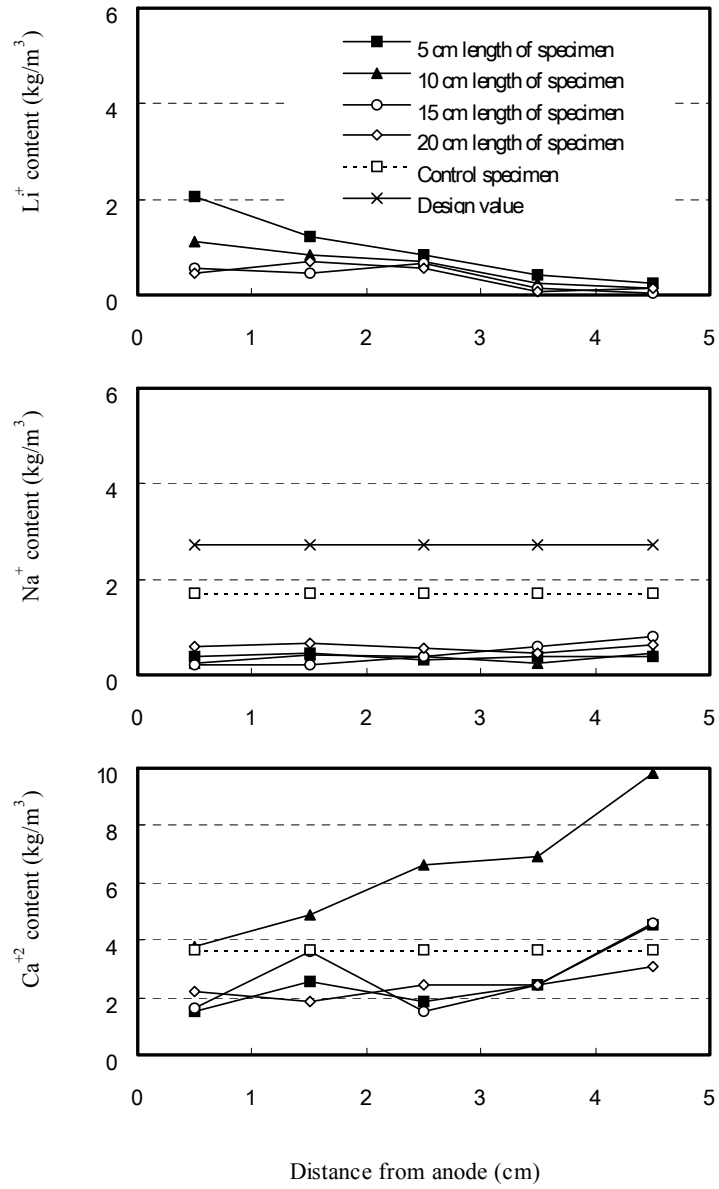


Figure 4: Distributions of the related ions in the samples after RLMT treatment

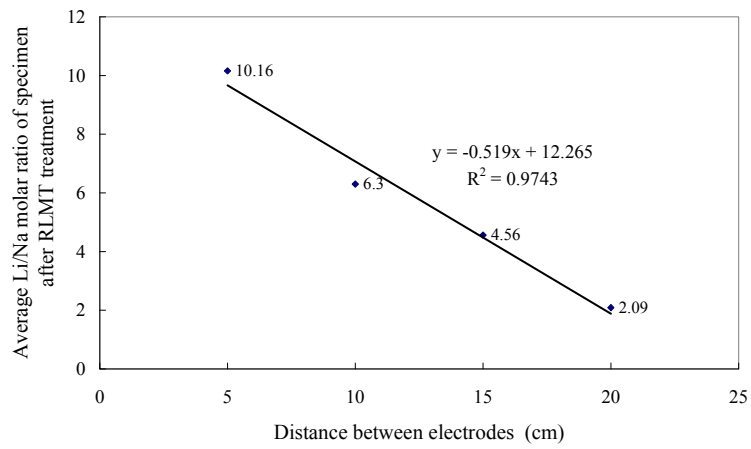


Figure 5: Relationship of the average Li/Na molar ratio and the distance between electrodes