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A simplified method to determine the chloride migration coefficient of concrete by the electric current in steady state

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Abstract. This study presents a rapid method for determining the steady state migration coefficient of concrete by measuring the electric current. This study determines the steady state chloride migration coefficient using the accelerated chloride migration test (ACMT). There are two stages to obtain the chloride migration coefficient. The first stage, the steady-state condition was obtainedfrom the initial electric current at the beginning of ACMT. The second stage, the average electrical current in the steady state condition was used to determine the steadystate chloride migration coefficient. The chloride migration coefficient can be determined from the average steady state current to avoid sampling and analyzing chlorides during the ACMT.

Keywords: chloride migration coefficient; charge passed; electric current

1. Introduction

The resistance to chloride ion ingress into concrete is an important issue in the design of concrete structures, as it primarily determines the durability of concrete. Once the chloride ions have reached the reinforcement, they may initiate corrosion and induce the damage of concrete structure. Previous research uses the conventional diffusion test method and the 90-day salt ponding test (ASTM C1543 2002) to evaluate the chloride diffusion characteristics of cement-based materials. The disadvantages of these tests are that they are time-consuming, laborious, and expensive (Chiang and Yang 2007).

Researchers have developed many methods of rapid chloride ion diffusion (ASTM C1202 1994,Yang and Su 2002, NT Build 492 1999) based on the application of an external electrical field.The rapid chloride permeability test (RCPT), designated as ASTM C1202 (ASTM 1994), is a test for the rapid qualitative assessment of chloride permeability of concrete. The standards specify the rating of chloride permeability of concrete based on the charge passing through the specimen over a six hour (6h) testing period. The RCPT method has been used to investigate the mineral admixture effect on the resistance of chloride ion penetration (Jain and Neithalath 2010, Zhang *et*

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al. 2011), and the effect of crack width on chloride migration coefficient of concrere (Jang *et al.* 2011). McCarter *et al.* (2000) showed that the conductivity of the free pore fluid increases with increasing temperature, and many researchers (Feldman 1999, Detwiler and Fapohunda 1993, Andrade 1993) have indicated that the applied electrical potential of RCPT heats up the concrete specimen, which may affect the flow speed. Shi (1996) noted that the amount of charge passing through mixtures containing sodium alkali-activated slag mortars is related to the conductivity of the pore fluid within the mortar. For concrete containing mineral admixtures, RCPT can be to measure the concrete conductivity rather than the chloride permeability of concrete (Wee *et al.* 2000). However, Shane *et al.* (1999) pointed out that the RCPT procedure is not for use with concretes containing mineral additives such as silica fume, fly ash, and slag. The Accelerated Lithium Migration Technique (ALMT) has been used to investigate the effects of specimen length on ASR-related cation migration during the use of ALMT (Wang *et al.* 2012). The ACMT method has been used to investigate the chloride migration of concrete after carbonation (Zhang and Zhao 2012) and the chloride migration of interfacial transition zone (Yang and Weng 2013).

To simplify the migration test, Castellote *et al.* (2001) correlated the chloride concentration and conductivity in the anodic compartment of a migration test in the steady state. Tong and Gjorv (2001) correlated the initial electrical conductivity, the conductivity of NaCl saturated concrete, and the charge passed through the concrete.

In the previous works, the linear correction between the migration coefficient and charge passed obtained from the ACMT under steady-state condition was found (Yang 2004, Yang and Cho 2003). Yang and Cho (2004) indicated that the steady state diffusion coefficient and electrical current obtained from the steady state condition are linearly correlated. The disadvantages of these works are that they need to obtain the steady-state condition by measuring the chloride ions during the ACMT. In this study, the steady-state condition was obtained from the initial electric current at the beginning of ACMT, and the charge passed and the average electric current in the steady state was used to obtain the steady state chloride migration coefficient. This study develops a simplified testing procedure to obtain the steady state chloride migration coefficient of concrete without sampling and analyzing chlorides during the ACMT.

2. Experimental program

2.1 Materials

ASTM Type I Portland cement (specific gravity: 3.15), fly ash (specific gravity: 2.21), and slag (specific gravity: 2.83) were used as binders. River sand with a fineness modulus of 3.0 was used as fine aggregate, while crushed limestone with a nominal maximum aggregate size of 20 mm was used as coarse aggregate. Table 1 shows the details of the mix proportions for 16 different concrete mixes. The concrete samples were cured for two different ages (28 days and 91 days). Test specimens were marked in such a manner that the first letter(s) indicate(s) the type of binder: C, F, S and SF that stand for cement, fly ash, slag, and slag/fly ash respectively. The following number indicates the water/binder ratio. The amount of coarse and fine aggregates was kept constant at 946 and 806 kg/m^3, respectively. A superplasticiser (sulfonated naphthalene formaldehyde type) was also used for concrete mixtures with low w/b ratios, such as 0.35 and 0.45.

Table 1 Mix proportions and mix designation of concrete (kg/m^3)

Mix	w/b	Cement	Water	Fly ash	Slag	Fine aggregate	Coarse aggregate	SP
C35	0.35	509	178	0	0	947	806	0
C45	0.45	443	199	0	0	946	806	0
C55	0.55	392	216	0	0	946	806	0
C65	0.65	351	228	0	0	946	806	0
F35	0.35	390	166	98	0	947	806	5
F45	0.45	341	190	85	0	946	806	2
F55	0.55	303	208	76	0	946	806	0
F65	0.65	273	221	68	0	946	806	0
S35	0.35	298	174	0	199	946	806	0
S45	0.45	260	195	0	174	946	806	0
S55	0.55	231	212	0	154	946	806	0
S65	0.65	207	225	0	138	946	806	0
SF35	0.35	345	167	44	104	947	806	6
SF45	0.45	301	191	39	90	946	806	2
SF55	0.55	267	210	34	80	946	806	0
SF65	0.65	240	223	31	72	946	806	0

2.2 Specimen preparation

Three cylinders measuring 100 in diameter and 200 mm in height were cast in molds for each of the 16 concrete mixes. After demolding, the specimens were cured in water (23℃) for 28 days and 91 days. Three specimens measuring 30 mm thick were then cut from the mid-portion of the cylinders and vacuum saturated according to ASTM C1202 (ASTM 1994). The lateral surface of each specimen was coated with epoxy and the specimen was then placed in a vacuum desiccator at a pressure of less than 1 mm Hg for 3 hours. De-aerated water was added to immerse the specimen and the vacuum level was maintained for an additional hour. Specimens were soaked in the added water for 18 hours after turning off the pump, and then removed from the water and placed in the ACMT setup.

2.3 Experimental procedure

The electrochemical technique was used to accelerate the migration of chloride ions in concrete to estimate the chloride migration coefficient. Fig. 1 presents a schematic diagram of the test set-up (Yang and Su 2002, Yang and Weng 2003). The specimen was placed between two acrylic cells. One of the cells was filled with 4.5 liters of 0.3N NaOH solution, and the other with 4.5 liters of 3% NaCl solution. Brass meshes (φ 10 mm, # 20 mesh) were placed on the side of the specimen to serve as electrodes. The cells were connected to a 24V DC power source. The NaOH electrode acted as the anode, and the NaCl electrode as the cathode. The electrical current was measured and recorded at 5 minute intervals using a data logger. The quantity of chloride ions passing through the specimen in the anode cell was measured and determined periodically using potentiometric titration with 0.01N silver nitrate ($AgNO_3$) solution.

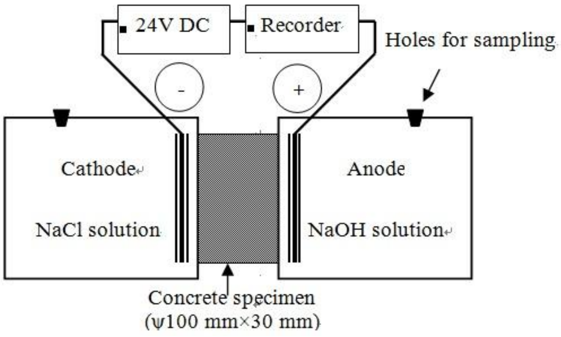

Fig. 1 Schematic diagram of the accelerated chloride migration test

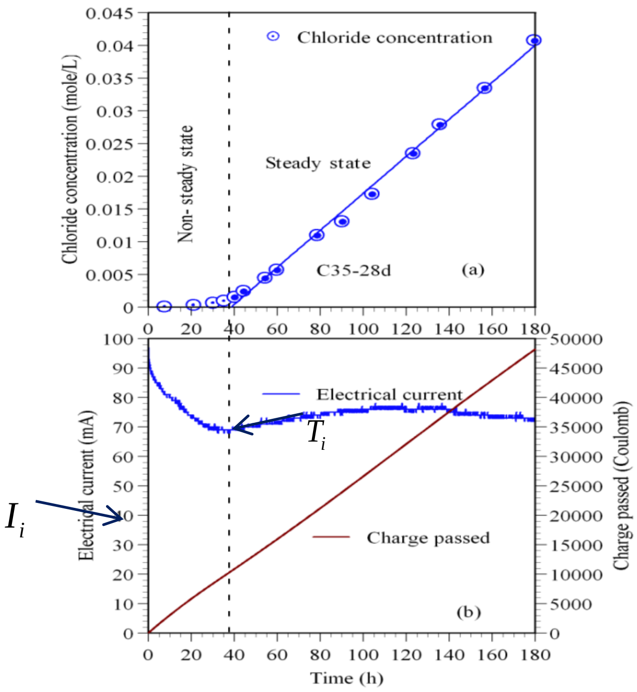

Fig. 2 Chloride concentration during the ACMT (C35-28d) (a), charge passed and electrical current during the ACMT (b)

3. Results and discussion

3.1 Chloride migration in ACMT

This study determines the chloride concentration from the solution in the anode cell. The chloride concentration results for each concrete mix represents the average of three test samples.

Fig. 2(a) plots the typical result of chloride concentration as a function of time. Fig. 2(a) shows that two stages exist, non-steady state, and steady state with respect to the change of the chloride concentration. In the non-steady state, the chloride ions are migrating through saturated pores in the concrete and have not yet reached the anode cell. In the steady state stage, the flux of chloride ions passing through the concrete specimen becomes constant. The steady state stage was obtained from the linear regression, and the best correlation coefficient ($R^2 \approx 1$) of the linear line was found using trial and error method. In this study, the time duration in steady state is the duration from the chloride ion reached the anode cell to the end of test. Table 2 lists the steady state durations for all mixes. This duration ranges between 55 h and 315 h for all mixes, and decreases with the increase w/b ratio (except for mixes F55 and SF45).

To calculate the chloride migration rate (K) for chloride ion penetration, this study applies linear regression to the steady state portion of the curve:

$$C = Kt + a, \tag{1}$$

where C is the chloride concentration in the anode cell in mole/L, t is the elapsed time in h, and K is the chloride migration rate in mole/L/h. The chloride migration rate (K) for each concrete mix was obtained from the steady state portion of the chloride-time curve using linear regression analysis. Table 3 lists the chloride migration rate and the correlation coefficient (R^2)for all mixes. The correlation coefficient (R^2)of linear regression analysis in the steady state for all mixes exceeds 0.99 (Table 3). This indicates that the slope of chloride-time curve in steady state is constant.

Table 2 Time duration in steady state of ACMT

Mix	Time duration in the steady state (h)	
	28 days	91 days
C35	199	190
C45	121	131
C55	74	78
C65	55	73
F35	196	304
F45	152	251
F55	109	299
F65	70	167
S35	272	269
S45	242	212
S55	126	163
S65	97	159
SF35	216	277
SF45	242	295
SF55	126	163
SF65	83	130

Table 3 Chloride migration rate and chloride migration coefficient in ACMT

Mix	K ,($\times 10^{-8}$ mole/L/s)				M_s , ($\times 10^{-12}$ m^2/s)	
	28 days age	R^2	91 days age	R^2	28 days age	91 days age
C35	8.03	0.997	5.81	0.999	2.80	2.03
C45	10.56	0.999	9.32	0.999	3.65	3.22
C55	12.58	0.994	14.94	0.999	4.79	5.68
C65	20.03	0.997	20.47	0.999	6.87	7.02
F35	5.72	0.997	3.65	0.998	2.06	1.31
F45	8.75	0.998	4.65	0.999	3.18	1.69
F55	10.37	0.994	4.76	0.999	3.88	1.78
F65	16.05	0.990	5.86	0.999	5.82	2.13
S35	3.77	0.995	3.48	0.999	1.33	1.22
S45	4.86	0.994	4.02	0.999	1.79	1.48
S55	6.37	0.997	5.38	0.999	2.21	1.87
S65	7.55	0.995	6.09	0.998	2.71	2.19
SF35	4.09	0.994	3.30	0.997	1.54	1.24
SF45	5.52	0.987	3.96	0.999	2.13	1.53
SF55	11.06	0.998	5.29	0.999	3.88	1.86
SF65	12.15	0.987	6.09	0.999	3.94	1.97

3.2 The electrical current and the charge passed

The electrical current across the cells was recorded with a data logger during the experiment. In an electrolyte solution of several ionic species, the total electrical current, I , is written as

$$I = \sum_j I_j , \tag{2}$$

where I_j is the electrical current resulting from the migration of an ionic species j . The charge passed, Q , can be determined by integrating the current-time curve and calculated by

$$Q = \int I(t) dt , \tag{3}$$

where Q is charge passed in coulombs. $I(t)$ is the time-dependent total electrical current in A and t is the elapsed time in s.

Fig. 2(b) plots the typical result of electrical current and charge passed from ACMT as a function of time. Fig. 3 plots the charge passed for mixes C, F, S, and FS at a curing age of 28 d as a function of time. This figure shows that charge passed is a linear function of time.

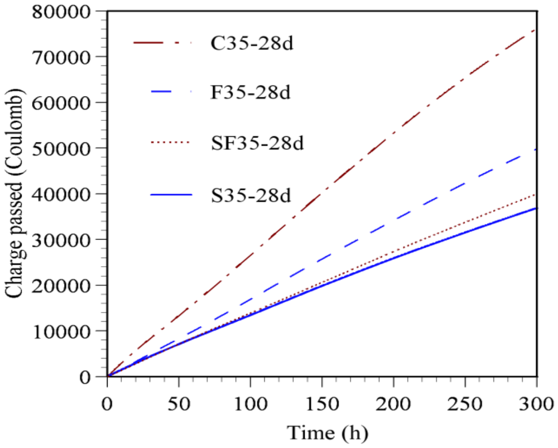

Fig. 3 Variations of charge passed for mixes C, F, S, and FS at a curing age of 28 d

3.3 The measuring time in the steady state

Fig. 2(a) and Table 3 show that the flux of chloride ions passing through the concrete specimen is constant in the steady state. Since the chloride concentration in the steady state portion of the chloride-time curve is a linear function of time, the chloride migration rate can be determined by any two chloride concentrations during the steady state. The difficulty is determining the right time to take the chloride samples.

Previous researcher (Yang and Chiang 2008) measures the initial electrical current (I_i) at the beginning of ACMT to obtain the chloride measuring times during the steady state. The measuring times during the steady state were obtained from the maximum of start time and the minimum of end time in steady state period for all test specimens. The initial electrical currents measured from various mixes were classified into four categories to obtain the measuring times during the steady state: greater than 80 mA (rank 1), 51-80 mA (rank 2), 21-50 mA (rank 3), and less than 20 mA (rank 4). Table 4 shows the initial measuring time T_i and the terminal measuring time T_f during the steady state in ACMT for each category.

Table 5 lists the initial electrical current (I_i) measured at the beginning of test for all mixes. The initial measuring time (T_i) of steady state period in Table 5 was obtained by measuring the initial electrical current according to Table 4. The initial electrical current (I_i) and the initial measuring time (T_i) of steady state period for C35-28d are marked in Fig. 2. Figs. 4 (a), (b) plot the typical results of chloride concentration and charge passed as a function of time, respectively. The time scale is between 35 h and 55 h to emphasize the initial measuring time (T_i).

These figures also show the steady state chloride concentration difference ($C_s = C_{i+9} - C_i$) and the steady state charge passed ($Q_s = Q_{i+9} - Q_i$)in the steady state. The steady state charge passed (Q_s) and the steady state chloride concentration difference (C_s) are defined as the 9 h charge passed and chloride concentration difference, respectively, from the initial measuring time T_i. This 9 h basis is 3/2 times longer than that of the RCPT (ASTM C1202 1994) method, because the

Table 4 The sampling time region and the ranks obtained from the initial current

Classification		Rank 1	Rank 2	Rank 3	Rank 4
	Initial current, mA	> 80	51~80	21~50	< 20
Sampling time	Initial sampling time (T_i), h	40	79	201	333
	Terminal sampling time (T_f), h	80	128	245	415

Table 5 The initial current, the initial measuring time, the chloride concentration difference (C_s), and the steady state charge passed (Q_s)

Mix	I_i (mA)		T_i (h)		C_s ($\times 10^{-3}$ mol/L)		Q_s (Coulomb)	
	28 days age	91 days age	28 days age	91 days age	28 days age	91 days age	28 days age	91 days age
C35	98	50	40	201	2.60	1.88	2260	1511
C45	130	67	40	79	3.42	3.02	2871	2583
C55	125	94	40	40	4.08	4.84	3552	3879
C65	181	114	40	40	6.49	6.63	5152	4921
F35	48	15	201	333	1.85	1.18	1494	811
F45	61	18	79	333	2.84	1.51	2104	1102
F55	79	21	79	201	3.36	1.54	2772	1181
F65	110	23	40	201	5.20	1.90	4118	1420
S35	48	24	201	201	1.22	1.13	1041	869
S45	49	22	201	201	1.57	1.30	1326	969
S55	59	34	79	201	2.06	1.74	1720	1317
S65	66	34	79	201	2.45	1.97	1784	1451
SF35	44	15	201	333	1.33	1.07	1188	743
SF45	50	19	201	333	1.79	1.28	1423	870
SF55	80	31	79	201	3.58	1.71	2600	1563
SF65	92	28	40	201	3.94	1.97	3101	1446

applied voltage in the ACMT is 24/60 times that of the RCPT method and the thickness of the specimen is 3/5 times that of the RCPT specimen. Table 5 lists the steady state charge passed (Q_s) and the steady state chloride concentration difference (C_s) in the steady state for all mixes.

Fig. 5 illustrates the relationship between the steady state charge passed (Q_s) and the steady state chloride concentration difference (C_s) in the steady state. The empirical relationship that relates the chloride concentration difference (C_s) to the steady state charge passed (Q_s) can be derived as

$$C_s = 1.262 \times 10^{-6} Q_s, \tag{4}$$

where C_s is in mol/L and Q_s is in coulombs. The correlation coefficient of linear regression R^2 is 0.986. The linear relationship between C_s and Q_s is fairly good.

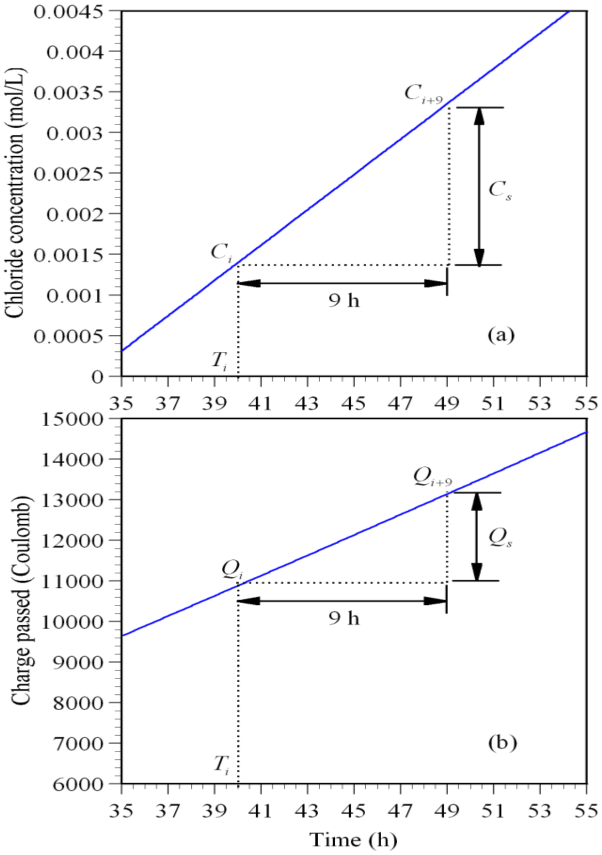

Fig. 4 The steady state chloride concentration difference (a), and the steady state charge passed (b)

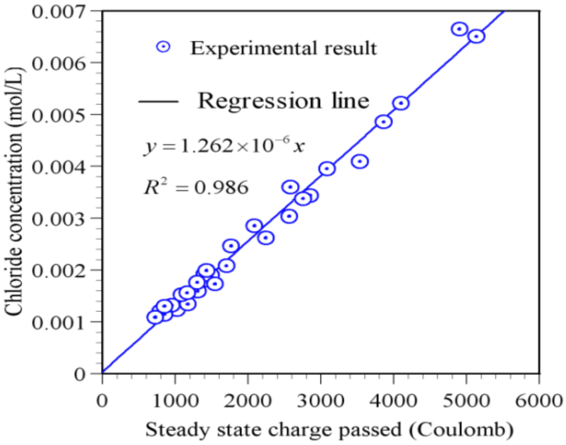

Fig. 5 The relationship between the steady state charge passed (Q_s) and the chloride concentration difference (C_s) in the steady state

3.4 Steady state migration coefficient in ACMT

This study determines the steady state chloride migration coefficient (M_s) based on the chloride migration rate (K)and Nernst-Planck's equation as (Andrade 1993)

$$M_s = \frac{RT}{|z|C_oFE}\frac{V_a}{A}K, \tag{5}$$

where R is the universal gas constant (8.3 Joule/mole/K), T is absolute temperature (K), z is the electrical charge of chloride, C_o is chloride concentration in the source cell (mole/L), F is the Faraday constant (96500 coulomb /mole), E is the strength of the electric field between the anode and cathode (V/m), V_a is the volume of solution in anode cell (m^3), and A is the specimen surface exposed to chloride ions (m^2). Table 3 lists the chloride migration coefficient (M_s) for all mixes. Figs. 6 and 7 show the correlation between w/b ratio and the steady state chloride migration coefficient of concrete (M_s) obtained from the ACMT of all mixes tested at the curing age of 28 days and 91 days, respectively. These figures show that the steady state chloride migration coefficient (M_s) increases as the w/b ratio increases because more pores and diffusing paths may form as the w/b ratios increase. The steady state chloride migration coefficient decreased significantly for concrete containing mineral admixtures cured for 91 days. For a given w/b ratio, the steady state chloride migration coefficient of mixes F, S, and SF is lower than the mixes C.

Fig. 8 shows the relationship between the steady state charge passed (Q_s) and the steady state chloride migration coefficient (M_s) for all mixes, and the corresponding regression results. Using linear regression, the empirical relationship between M_s and Q_s is statistically derived as

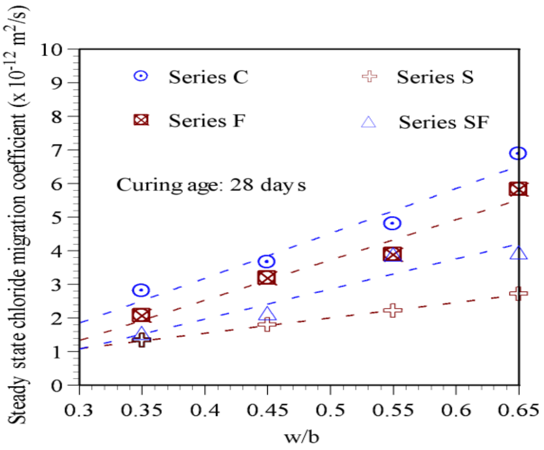

Fig. 6 The correlation between w/b ratios and the migration coefficient of concrete for all mixes at a curing age of 28 days

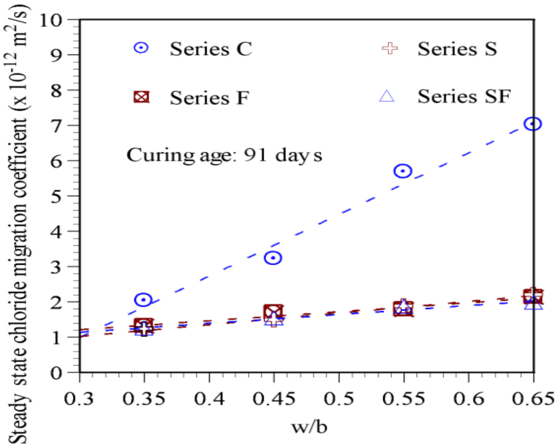

Fig. 7 The correlation between w/b ratios and the migration coefficient of concrete for all mixes at a curing age of 91 days

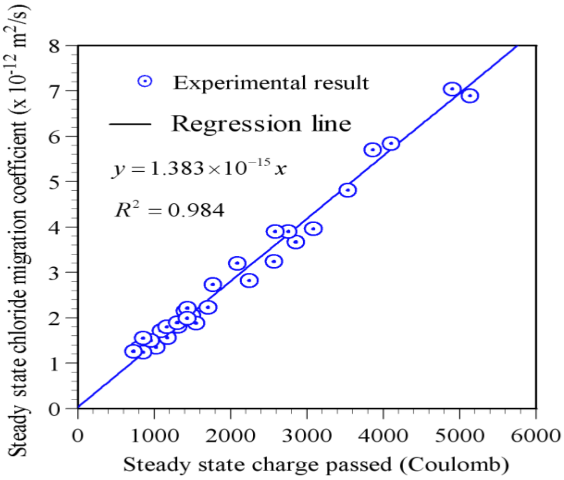

Fig. 8 The relationship between the steady state charge passed (Q_s) and the steady state chloride migration coefficient (M_s), and the corresponding regression results

$$M_s = 1.383\times10^{-15} Q_s, \tag{6}$$

where Q_s is in coulombs, and M_s is in m^2/s. Linear regression shows that the correlation coefficient R^2 for the model [Eq. (6)] is 0.984. This implies that Q_s correlates linearly with M_s regardless of concrete mixes.The steady state chloride migration coefficient (M_s) is based on the chloride migration rate (K), and the chloride concentration in the source cell, the strength of the electric field, and the specimen surface exposed to chloride ions are considered in Eq. (5) to calculate the migration coefficient. The chloride concentration in the source cell, the strength of the electric field, and the specimen surface exposed to chloride ions have insignificant influence on the empirical coefficient in Eq. (6) theoretically. Spiesz and Brouwers (2012) study the influence of the applied voltage (electrical field) on the value of the chloride migration coefficient, as determined with the Rapid Chloride Migration (RCM) test, and pointed out that for the applied voltage of 35 V and 47.5 V the calculated average values of the migration coefficient are almost identical and for the voltage of 60 V they are slightly increased (about 10%). The strength of the electric field may slightly influence on the empirical coefficient in Eq. (6).

3.5 Steady state electrical current in ACMT

The experiments in this study recorded the electrical current across the cells with a data logger. Feldman *et al*. (1999,1994) used the RCPT to obtain initial current at the beginning of the test and the charge passed in 6 h, and pointed that there is a good correlation between the initial current charge passed. Fig. 9(a) presents the relationship between the initial current at the beginning of the ACMT and the initial charge passed (Q_i) obtained from the first 9 h of the ACMT. Fig. 9(b) presents the relationship between the initial current and the steady state chloride migration coefficient (M_s) for all mixes. The initial current and initial charge passed from ACMT is linearly correlated, but the correlation of the initial current (I_i) and the steady state migration coefficient(M_s) is poor. This implies that the steady state migration coefficient cannot be determined using the initial current.

The steady state electrical current (I_s) was obtained from the electrical current during the 9 h period after the initial measuring time T_i in Table 5. Figs. 10 and 11 present the steady state electrical current for some mixes, while Table 6 lists the range of steady state electrical current for all mixes. Only small variations appeared inthe current during the 9 h experiment. Table 6 lists the average steady state electrical current (I_{sa}) for all mixes. The small current variations in this table illustrate that the effect of other ions in the pore solution on the current is insignificant in the steady state. Fig. 12 depicts the relationship between the average steady state electrical current (I_{sa}) and the steady state chloride migration coefficient (M_s) for all mixes, and the corresponding regression results. Using linear regression, the empirical relationship between M_s and I_{sa} is statistically derived as

$$M_s = 4.485 \times 10^{-14} I_{sa}, \tag{7}$$

where I_{sa} is in mA, and M_s is in m^2/s. Linear regression shows that the correlation coefficient R^2 for the model [Eq. (7)] is 0.984. This implies that I_{sa} correlates linearly with M_s regardless of concrete mixes.

The accelerated chloride migration test requires a great amount of chloride sampling and analysis to obtain the chloride-time curve, making this kind of test laborious and expensive. To avoid sampling and analyzing chlorides during the ACMT, the initial electrical current was

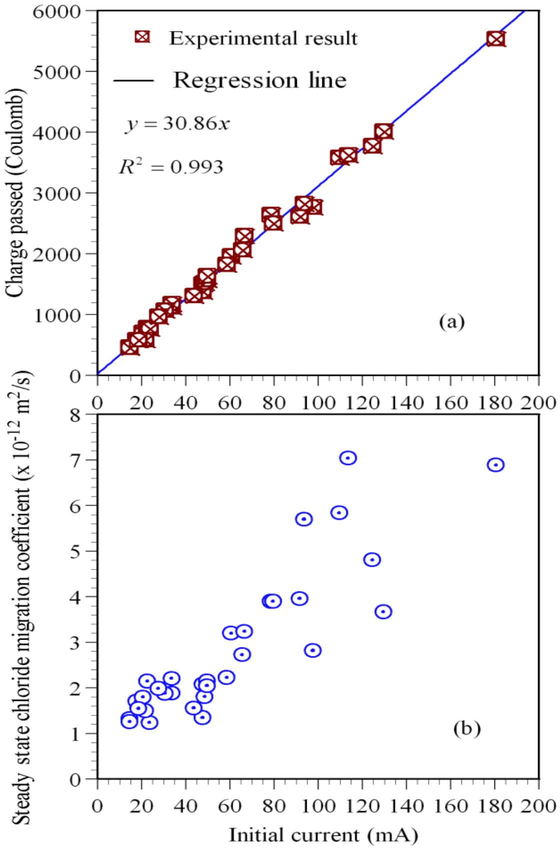

Fig. 9 The relationship between the initial current and the charge passed obtained from the first 9 h of the CMT (a), and the relationship between the initial current and the steady state chloride migration coefficient (b)

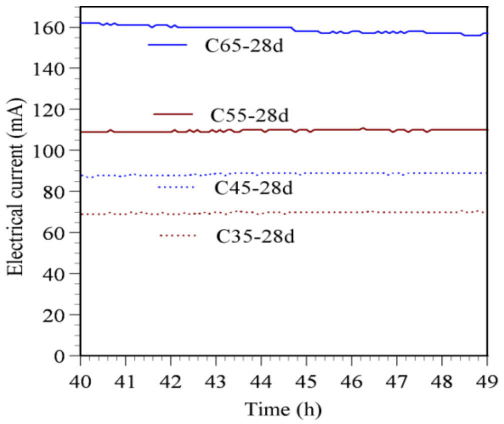

Fig. 10 The variation of steady state electrical current (I_s) obtained from the electrical current during the 9 h period after the initial sampling time T_i at a curing age of 28 days

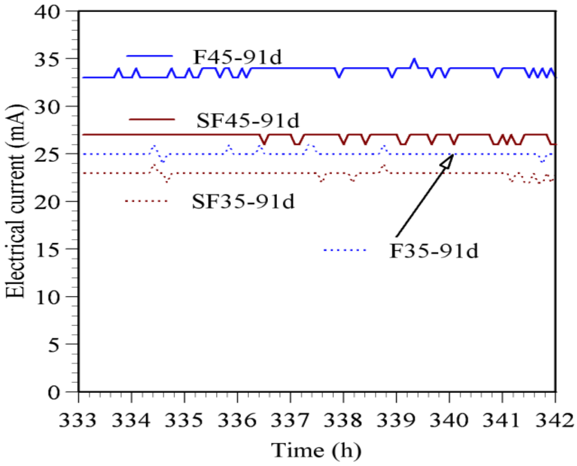

Fig. 11 The variation of steady state electrical current (I_s) obtained from the electrical current during the 9 h period after the initial sampling time T_i at a curing age of 91 days

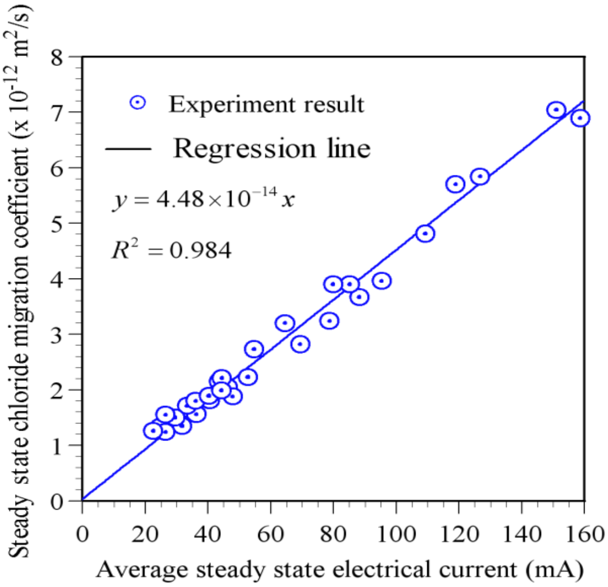

Fig. 12 The average steady state electrical current (I_{sa}) and the steady state migration coefficient

measured at the beginning of ACMT. The initial measuring time T_i can be obtained from Table 4 according to the initial current. The steady state charge passed (Q_s) and the average steady state electrical current (I_{sa}) were obtained from the charge passed and electrical current during the 9 h period after the initial measuring time. The steady state chloride migration coefficient (M_s) can be determined by Eq. (6) from the Q_s or by Eq. (7) from the I_{sa} .

Table 6 The range of steady state current and the average steady state current

Mix	Range of steady state current, (mA)		Average steady state current I_{sa}, (mA)	
	28 days age	91 days age	28 days age	91 days age
C35	69 ~ 71	46 ~ 47	69.76	46.63
C45	87 ~ 89	77 ~ 80	88.61	79.12
C55	109 ~ 110	118 ~ 120	109.63	119.29
C65	158 ~ 162	150 ~ 153	159.01	151.43
F35	46 ~ 47	24 ~ 26	46.13	25.04
F45	64 ~ 66	33 ~ 34	64.94	33.68
F55	85 ~ 86	36 ~ 37	85.55	36.45
F65	124 ~ 130	43 ~ 44	127.10	43.82
S35	31 ~ 33	26 ~ 27	32.13	26.83
S45	40 ~ 42	29 ~ 31	40.94	29.91
S55	53 ~ 54	40 ~ 42	53.10	40.66
S65	54 ~ 57	44 ~ 45	55.06	44.78
SF35	36 ~ 37	22 ~ 24	36.67	22.94
SF45	43 ~ 44	26 ~ 27	43.92	26.85
SF55	80 ~ 81	48 ~ 51	80.24	48.24
SF65	95 ~ 97	44 ~ 45	95.71	44.64

4. Conclusions

Concrete resistance to chloride ingress can be determined from the chloride migration coefficient obtained from the proposed ACMT. The following conclusions can be drawn from the results of the present experimental investigation.

(1)In the steady state condition of ACMT, the accumulated chloride concentration and the charge passed are both linear functions of time.

(2)The initial current (I_i) measured at the beginning of ACMT is linearly correlated with the initial charge passed (Q_i) obtained from the first 9 h of ACMT, but the correlation of the initial current and the steady state migration coefficient (M_s) is poor. The steady state migration coefficient cannot be determined using the initial current.

(3)The steady state electrical current (I_s) was measured from the electrical current over a 9 h period after the initial sampling time T_i. There are only small current variations in the steady state condition, indicating that the effect of other ions in the pore solution on the current is insignificant in the steady state.

(4)There is a good correlation between the steady state chloride migration coefficient and the steady state charge passed, and between the steady state chloride migration coefficient and the average steady state current. The chloride migration coefficient can be determined from the steady state charge passed or the average steady state current to avoid sampling and analyzing chlorides during the ACMT

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