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FACTORS AFFECTING THRESHOLD CHLORIDE FOR REINFORCEMENT CORROSION IN CONCRETE

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ABSTRACT

Three cements with variable C_3A contents were mixed with different levels of chloride, alkali and sulfate contents to study the effect of these parameters on pore solution composition. Effect of exposure temperature was also studied by curing the chloride-treated specimens at 20° and 70°C. Pore solution was extracted using a high pressure pore solution extrusion device and analysed for chloride and hydroxyl ion concentrations. Threshold chloride for onset of reinforcement corrosion was computed using threshold $[Cl^-/OH^-]$ ratio of 0.3. The results showed that C_3A content and exposure temperature have very strong influence on threshold chloride content. Alkali content of cement has marginal effect whereas presence of sulfates along with chlorides has moderate effect on the threshold chloride content.

Introduction

Premature deterioration of concrete structures currently constitutes a major global concern for the construction industry throughout the world. Bridge decks, parking garages, marine structures and structures located in the aggressive environments such as in the Arabian Gulf region are among the structures undergoing deterioration due to chloride-induced corrosion of reinforcing steel. Reinforcement corrosion results in cracking and spalling of concrete, causing a serious loss of serviceability and structural integrity of the structure. Chlorides may be introduced into concrete through accelerating admixtures, chloride contaminated aggregates or brackish mixing water. Chlorides may also enter into concrete subsequently, by de-icing salts in bridge decks and parking structures, from sea water in marine structures, or from saline soil and ground water in structures in the Gulf region.

The process of corrosion of reinforcing steel comprises two phases (1,2), the corrosion initiation phase and corrosion propagation phase. The corrosion propagation leads to cracking and spalling of concrete. Once corrosion is initiated, cracking and spalling of concrete follows very shortly. Also, very little, with the exception of cathodic protection, can be done to stop the corrosion process once it is initiated.

Corrosion of reinforcement is initiated when $[Cl^-/OH^-]$ ratio of the pore solution at the steel concrete interface exceeds the threshold value. The importance of corrosion initiation time is well recognized and many investigators have attempted to find the threshold value of $[Cl^-/OH^-]$ ratio required for corrosion initiation. For instance, Hausmann (3) and Gouda (4) conducted studies on steel immersed in alkaline solutions similar to concrete pore solutions. Hausmann found the threshold [Cl^-/OH^-] ratio depended upon the alkalinity of the solution. Based on Gouda's results, Diamond (5) proposed a threshold [Cl^-/OH^-] ratio value of 0.3 for pH normally encountered in hardened concrete pore solutions. Lately, some investigators (6-8) carried out studies on steel embedded in concrete to find the threshold [Cl^-/OH^-] ratio. These investigators gave different values. However, the threshold [Cl^-/OH^-] ratio found using concrete, in general, was higher than that found using alkaline solutions. For example, Lambert et al (8) found a threshold [Cl^-/OH^-] ratio value of 3.0 compared to the value of 0.6 found by Hausmann (3) using alkaline solution. Due to difficulties for the measurement of [Cl^-/OH^-] ratio and dependency of corrosion initiation on numerous factors, there is no single value of threshold [Cl^-/OH^-] ratio which is accepted universally.

Some other investigators (9,10) have reported threshold chloride ion content (expressed as a proportion of cement) using performance data of actual concrete structures in the field. All these laboratory and field studies were conducted at normal exposure conditions and when concrete is contaminated with chlorides alone (admixed chlorides in the laboratory or de-icing salts used in the case of bridges).

Since it is the amount of free chlorides, rather than total chloride, present in the concrete pore solution which takes part in the corrosion reactions, corrosion initiation times are dependent upon factors which affect chloride binding capacity of cement. For instance, it has been shown (11) that corrosion initiation time of steel in different C_3A cements is a strong function C_3A content of the cement. For an increase in C_3A content from 2% to 14% and total chloride ion content of 1.2%, the chloride binding capacity and reinforcement corrosion initiation time were increased by 2.43 and 2.45 folds respectively. Other factors which affect chloride binding capacity of cement are its alkali content, level of sulfate ion contamination, exposure temperature, degree of carbonation and others. All these factors in turn affect threshold chloride content.

Apart from free chlorides present in concrete pore solution, threshold chloride content also depends upon OH^- concentration of the pore solution. Therefore, any factors which affect pore solution OH^- concentration also affect the threshold chloride. Some of these major factors are alkali content of the cement, level of sulfates, degree of carbonation and exposure temperature.

In this paper, an attempt has been made to quantify the relative effect of important factors such as C_3A and alkali content of cement, level of sulfate contamination and exposure temperature on chloride binding capacity and threshold [Cl^-/OH^-] ratio and chloride content of cements. Earlier work by the authors (11-14) and by Holden et al (15) discuss in detail the effect of these factors on pore solution composition and chloride binding capacity of cement. These data are used to deduce threshold chloride contents required for corrosion initiation. For the purpose of quantifying the relative effects of these factors, the most conservative threshold [Cl^-/OH^-] ratio value of 0.3 proposed by Diamond (5) has been used in this study.

The above discussion is pertinent to plain cements concrete and with similar physical characteristics. It has been shown that threshold [Cl^-/OH^-] ratio is not a unique value but rather depends on the physical characteristics of concrete (6). Therefore, the data presented in this paper should be used with caution as it may be applicable only to plain cement concrete of similar composition as the cement paste mixes used in this investigation.

Experimental Program

Three plain cements with variable C_3A contents of 2.43, 7.59 and 14% were used. The composition of the cements are given in Table 1. Four series of cement paste mixes were prepared. Series A with chloride addition only, Series B with chloride and alkali additions, Series C with chloride and sulfate additions and Series D relates to chloride-treated pastes cured at different temperatures. Details of mixes are given in Table 2.

TABLE 1
Composition of Cements (% by weight)

Cement No.	1	2	3
CaO	64.20	65.03	64.70
SiO_2	21.90	20.90	19.92
Al_2O_3	3.98	5.26	6.54
Fe_2O_3	4.80	3.75	2.09
SO_3	1.71	2.54	2.61
Na_2O	-	-	0.28
K_2O	-	-	0.56
Equivalent Na_2O	0.58	0.60	0.65
C_3S	54.30	55.83	54.50
C_2S	21.80	17.80	16.00
C_3A	2.43	7.59	14.00
C_4AF	14.61	11.41	6.50

TABLE 2
Details of Mixes

Series	Variable Parameter	Cement No.	Cl Addition* (% by weight of cement)	Levels of Variable Parameter
A	C_3A Content	1	0.3, 0.6, 1.2, 2.4	2.43% C_3A
		2	0.3, 0.6, 1.2, 2.4	7.59% C_3A
		3	0.3, 0.6, 1.2, 2.4	14% C_3A
B	Alkali Content	3	0.3, 0.6, 1.2	0.65% and 1.2% Na_2O Equivalent
C	Sulfate**	1	0.6, 1.2	0, 4, 8% SO_3#
		2	0.6, 1.2	0, 4, 8% SO_3#
		3	0.6, 1.2	0, 4, 8% SO_3#
D	Exposure Temperature	1	0.3, 0.6, 1.2	20 °C, 70 °C
		2	0.3, 0.6, 1.2	20 °C, 70 °C
		3	0.3, 0.6, 1.2	20 °C, 70 °C

* Added through NaCl
**SO_3 added through Na_2SO_4
#SO_3 was added to make the total SO_3 Content of Cements equal to 4 and 8%.

Cement paste specimens with a water-cement ratio of 0.60 were mixed and cured in sealed containers until equilibrium chloride concentrations in pore solution are achieved. No loss of water was allowed from the sealed containers during curing. After completion of curing, pore solutions were extracted from the specimens using a high pressure pore solution extrusion device. The pore solutions were analyzed for chloride and OH^- concentrations. Details of pore solution extraction and analysis are given elsewhere (11).

Results

Pore solution composition of series A specimens, with chloride additions of 0.3, 0.6 and 1.2%, are given in Table 3. Cl^-/OH^- ratios are plotted against chloride additions in Fig. 1. Values of threshold free chlorides (equivalent to water-soluble chlorides) and total chlorides (equivalent to acid-soluble chlorides) are scaled from the plot for a threshold Cl^-/OH^- value of 0.3. These values are given in Table 4.

TABLE 3

Effect of C_3A Content of Cement on Pore Solution Composition

Cement No.	C_3A Content of Cement (% by wt.)	Cl^- Addition (% by weight of cement)	Pore Solution Composition Cl^- (mM/L)	OH^- (mM/L)	pH	Cl^-/OH^-
1	2.43	0.3	69.7	258	13.41	0.2702
1	2.43	0.6	209.9	265	13.42	0.7922
1	2.43	1.2	529.9	254	13.40	2.0862
1	2.43	2.4	1368.0	231	13.36	5.9221
2	7.59	0.3	35.0	385	13.59	0.091
2	7.59	0.6	109.0	391	13.59	0.279
2	7.59	1.2	342.0	413	13.62	0.828
2	7.59	2.4	987.0	268	13.43	3.683
3	14.00	0.3	14.8	524	13.72	0.028
3	14.00	0.6	51.0	503	13.70	0.101
3	14.00	1.2	216.0	534	13.73	0.405
3	14.00	2.4	904.0	518	13.71	1.745

The threshold values of free chlorides for C_3A contents of 2.43, 7.59 and 14% C_3A cements are respectively 0.134, 0.165 and 0.215% by weight of cement. The threshold values in terms of total chlorides are 0.35, 0.62 and 1.0% by weight of cement for 2.43, 7.59 and 14% C_3A cements respectively. These data show a strong relationship between the total threshold chlorides and the C_3A content of the cement with the tolerable total chlorides for the 14% C_3A Type I cement being about 3 times the tolerable chlorides for the 2.43% C_3A Type V cement.

Table 5 shows the pore solution composition for series B cement paste mixes. In this series only 14% C_3A Type I cement was used. The original Na_2O equivalent alkali content for this cement is 0.65%. Another cement paste was prepared by adding NaOH to obtain Na_2O equivalent alkalies of 1.2%. Cl^-/OH^- ratios are plotted in Fig. 2 for the two cements with Na_2O equivalent alkali contents of 0.65% and 1.2%. It can be seen from the data of Table 5 that whereas an increase in the alkali content of the cement from 0.65% to 1.2% increases OH^- concentration in the pore solution increase in cement alkalies also concomitantly reduces chloride binding capacity ofthe cement hydrates. The net effect is a small increase in the Cl^-/OH^- ratio of the pore solution. Also, with an increase in the alkalies from 0.65% to 1.2% the threshold total chloride value is marginally lowered from 1.0% to 0.9%.

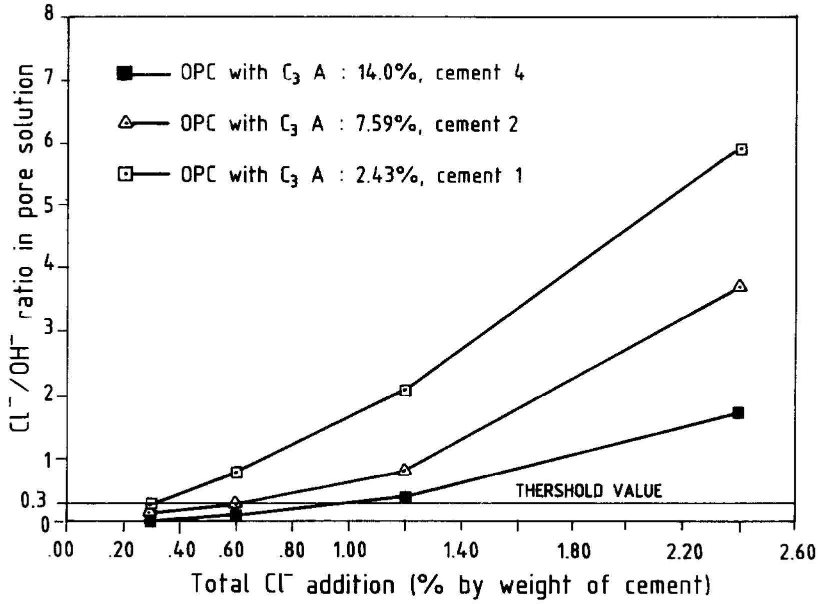

FIG. 1. Cl^-/OH^- Ratios in Pore Solutions of Different Cements for Various Levels of Chloride Addition.

TABLE 4

Effect of C_3A Content of Cement on Threshold Chloride Values

Cement Number	C_3A Content of Cement (% by weight of cement)	Threshold Chloride (% by weight of cement)	
		Free Cl^-	Total Cl^-
1	2.43	0.135	0.35
2	7.59	0.165	0.62
3	14.00	0.215	1.00

Fig. 3 is drawn from data developed by Page and Vennesland (16) and Diamond (5) which show Cl^-/OH^- ratio for cements with close C_3A contents of 7.37 and 9.1% respectively. Alkali contents of these cements were substantially different with values of 1.19% and 0.55% respectively. It can be seen from this presentation that for a given level of chlorides the Cl^-/OH^- ratio for the low alkali cement is higher compared to the corresponding value for the high alkali cement. Also, the threshold chloride value for the low alkali cement is 0.40% whereas it is 0.58% for high alkali cement. It is clear that in this case the effect of an increase in alkali content is

TABLE 5

Effect of Alkali Content of Cement on Pore Solution Composition

C_3A Content of Cement (% by weight)	Equivalent Na_2O Content of Cement (% by weight)	Total Cl^- Addition (% by wt. of cement)	Pore Solution Composition: Cl^- (mM/L)	OH^- (mM/L)	pH	Cl^-/OH^-
14	0.65	0.0	-	348	13.54	-
14	1.20	0.0	-	735	13.87	-
14	0.65	0.3	14.8	524	13.72	0.0282
14	1.20	0.3	28.5	755	13.88	0.0377
14	0.65	0.6	50.9	503	13.70	0.1014
14	1.20	0.6	93.8	740	13.87	0.1268
14	0.65	1.2	216.0	534	13.73	0.4045
14	1.20	1.2	362.8	750	13.88	0.4837

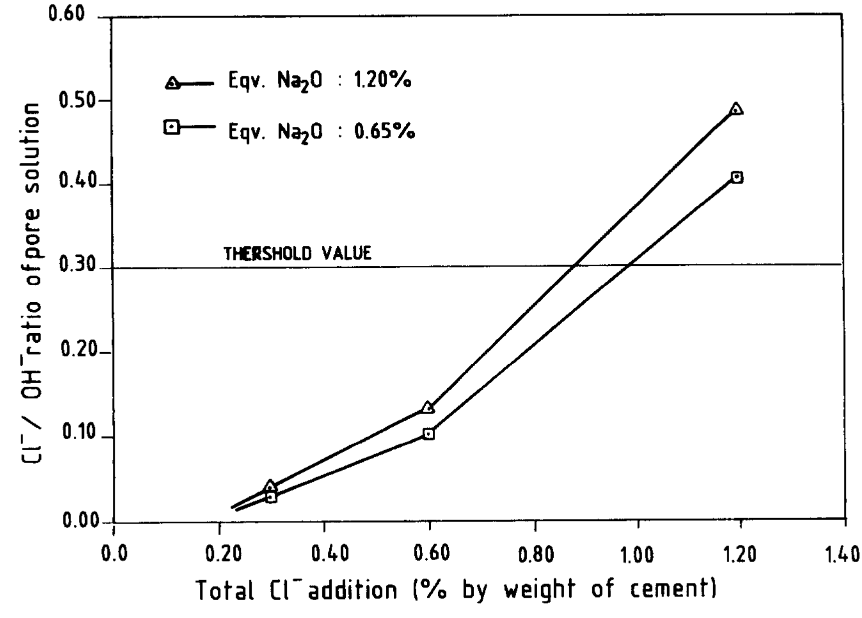

FIG. 2. Effect of Alkali Content of Cement on Cl^-/OH^- Ratio in the Pore Solution of Type 1 Cement C_3A: 14%.

opposite to that observed in the 14% C_3A cement used in this study. Therefore, the net result of an increase in cement alkalies will depend on the outcome of two opposing effects on pore solution chemistry: the beneficial increase in the OH ion concentration and the adverse reduction in chloride binding. It is seen that in the case of high 14% C_3A cement, increase in alkalies from 0.65% to 1.2% marginally increased pore solution aggressivity and slightly reduced chloride threshold. However, a similar alkali increase in medium C_3A cements significantly reduced pore solution aggressivity and increased chloride threshold.

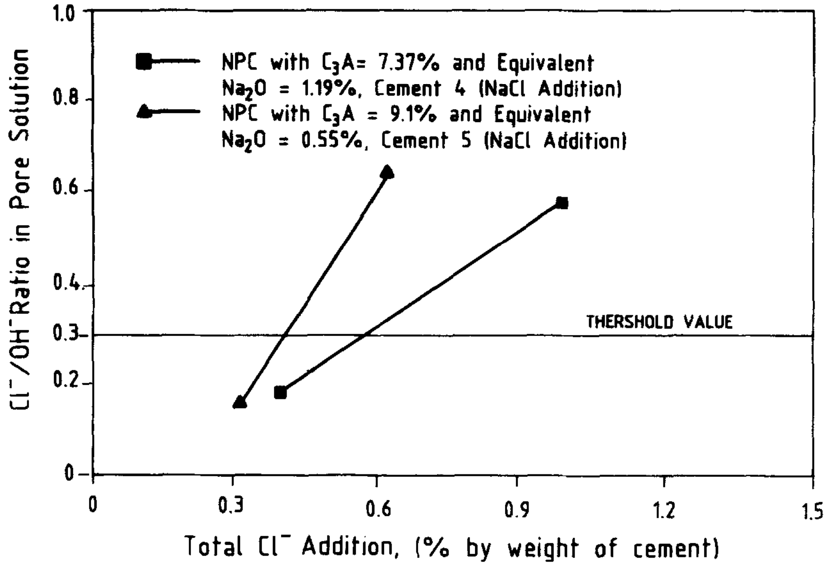

FIG. 3. Cl^-/OH^- Ratios for Various Levels of Chloride Addition in the Pore Solutions Expressed From Different Cement after the Establishment of Equilibirum Chloride Concentration.

Pore solution composition of series C specimens with both chloride and sulfate additions are given in Table 6. Two levels of chloride addition at 0.6% and 1.2% and two levels of sulfate addition corresponding to final SO_3 contents of 4 and 8% were used. Cl^-/OH^- ratios for different SO_3 contents are shown in Fig. 4. From Fig. 4, values of total threshold chlorides are scaled corresponding to (Cl^-/OH^-) ratio of 0.30 and are shown in Table 7. It can be seen that the effect of an increase in the sulfate content is not consistent in the three cements tested. The effect of sulfate addition on Cl^-/OH^- ratio and on threshold chloride content seems to depend upon C_3A and alkali contents of the cement. For 2.43% C_3A cement, addition of sulfate lowers the threshold chloride value. For 7.37% C_3A cement, 4% SO_3 addition results in an increase in the threshold chloride, but a further increase in SO_3 to 8% almost brings the threshold chloride back to the original value. In the 14% C_3A cement, the increase in SO_3 content results in a gradual reduction in the threshold chloride content. Therefore, caution has to be exercised when chloride limits are specified for situations where concrete is expected to get contaminated with chlorides and sulfates concomitantly. This happens in marine structures, in substructures exposed to salt bearing soils and ground water and in structures where unprepared aggregates introduce chlorides and sulfates to a concrete mix right at the time of making concrete. Approaching the problem conservatively, a 25% lower limit of allowable chloride content should be specified in such situations due to the fact that simultaneous sulfate presence may cause a reduction in the threshold chloride contents in medium and high C_3A cements as is evident from the data of Table 7.

Table 8 and Fig. 5 show the effect of temperature on Cl^-/OH^- ratio for the three cements tested. It can be seen that for all three cements, increase in exposure temperature from 20° to 70°C causes a sharp increase in the Cl^-/OH^- ratio. Threshold chloride contents scaled from Fig. 5 corresponding to threshold Cl^-/OH^- ratio of 0.3 are tabulated in Table 9. It can be seen that exposure temperature has a very strong effect on threshold chloride content. For all three cements, increase in temperature from 20° to 70°C causes at least fivefold reduction in the threshold chloride content. The performance of 2.43% C_3A Type V cement exposed to 20°C is even superior to that of 14% C_3A Type I cement exposed to 70°C.

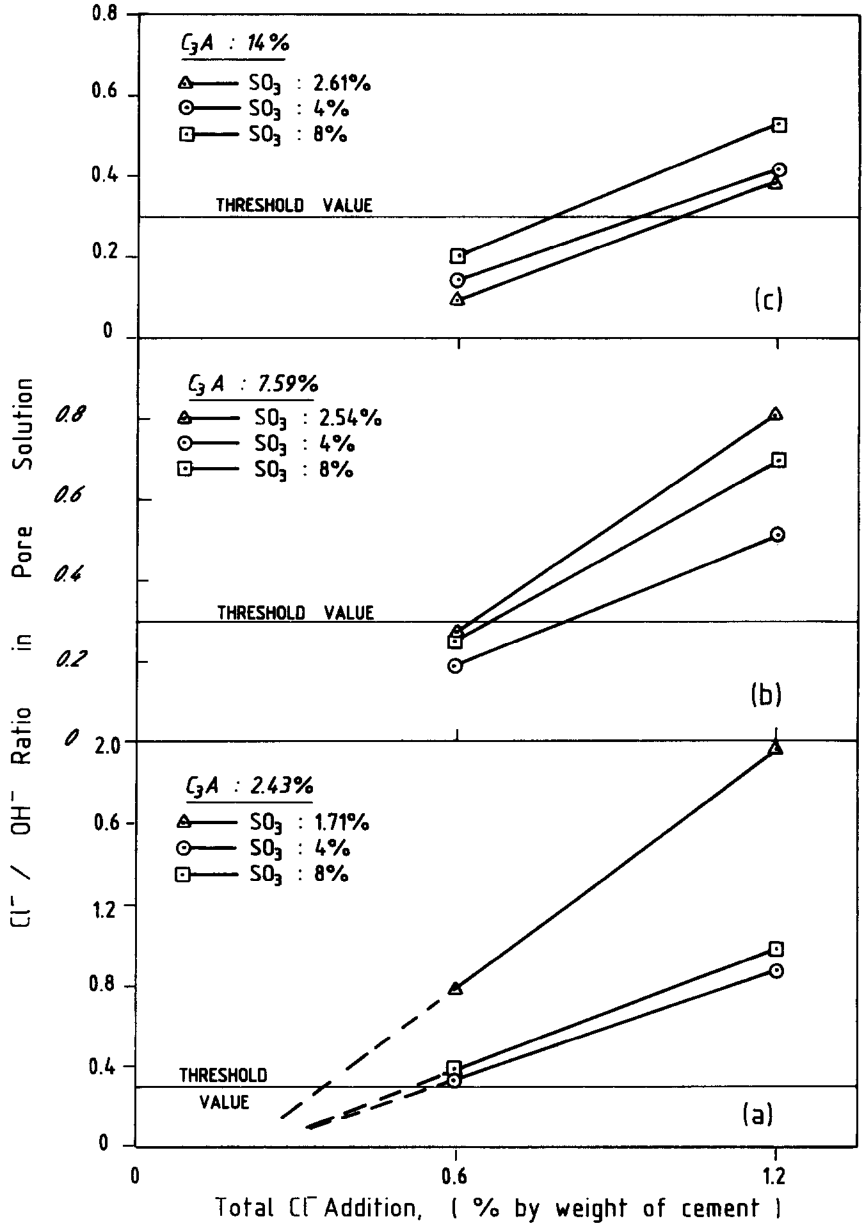

FIG. 4. Cl^-/OH^- Ratio for Cements Containing Chloride Sulfates.

Discussion

For the purpose of analyzing the effects of factors studied in this investigation, the threshold chloride contents for 2.43% C_3A Type V and 14% C_3A Type I cements are summarized in Table 10. It can be seen that increasing the alkali content of cement reduces the threshold chloride only very marginally. As mentioned earlier, other investigations (5,16) have observed slight increase in the threshold chloride due to increase in the alkali content of cement. Although increase in alkali content causes increase in OH^- concentration in the pore solution, it also inhibits chloride binding

TABLE 6

Pore Solution Composition with Different Levels of Chlorides and Sulfates

Cement No.	C_3A Content of Cement (% by wt.)	Total Cl^- Addition(% by wt. of cement)	Total SO_3 Content (% by weight of cement)	Pore Solution Composition Cl^- (mM/L)	OH^- (mM/L)	pH	Cl^-/OH^-
1	2.43	0.6	1.71	210	265	13.42	0.79
1	2.43	0.6	4.00	267	770	13.89	0.35
1	2.43	0.6	8.00	300	786	13.90	0.38
1	2.43	1.2	1.71	530	254	13.40	2.09
1	2.43	1.2	4.00	590	646	13.81	0.91
1	2.43	1.2	8.00	705	694	13.84	1.02
2	7.59	0.6	2.50	109	391	13.59	0.28
2	7.59	0.6	4.00	161	786	13.90	0.20
2	7.59	0.6	8.00	263	960	13.98	0.27
2	7.59	1.2	2.50	342	413	13.62	0.83
2	7.59	1.2	4.00	415	786	13.90	0.53
2	7.59	1.2	8.00	580	816	13.91	0.71
3	14.00	0.6	2.60	51	503	13.70	0.10
3	14.00	0.6	4.00	121	812	13.91	0.15
3	14.00	0.6	8.00	201	980	13.99	0.21
3	14.00	1.2	2.60	216	534	13.73	0.40
3	14.00	1.2	4.00	341	784	13.89	0.43
3	14.00	1.2	8.00	503	920	13.96	0.55

TABLE 7

Effect of Sulfates on Threshold Chloride Values

Cement No.	C_3A Content of Cement (% by weight of cement)	Total SO_3 Content (% by weight of cement)	Threshold Chloride Content (% by weight of cement)
1	2.43	1.71	0.35
1	2.43	4.00	0.53
1	2.43	8.00	0.54
2	7.59	2.50	0.62
2	7.59	4.00	0.80
2	7.59	8.00	0.66
3	14.00	2.60	1.00
3	14.00	4.00	0.93
3	14.00	8.00	0.78

TABLE 8

Effect of Curing Temperature on Pore Solution Composition

Cement No.	C_3A Content of Cement (% by weight)	Curing Temperature (°C)	Total Cl^- Addition (% by wt. of cement)	Pore Solution Composition			
				Cl^- (mM/L)	OH^- (mM/L)	pH	Cl^-/OH^-
1	2.43	20	0.3	69.7	258	13.41	0.27
		70	0.3	140.6	120	13.08	1.17
		20	0.6	209.9	265	13.42	0.79
		70	0.6	279.0	128	13.11	2.18
		20	1.2	529.9	254	13.30	2.09
		70	1.2	571.2	116	13.06	4.92
2	7.59	20	0.3	35.0	385	13.59	0.09
		70	0.3	129.2	150	13.18	0.86
		20	0.6	109.0	391	13.59	0.28
		70	0.6	267.7	160	13.20	1.67
		20	1.2	342.0	413	13.62	0.83
		70	1.2	561.6	162	13.21	3.58
3	14.00	20	0.3	14.8	524	13.72	0.03
		70	0.3	125.5	222	13.35	0.57
		20	0.6	50.9	503	13.70	0.10
		70	0.6	260.1	198	13.30	1.31
		20	1.2	216.1	534	13.73	0.40
		70	1.2	533.6	194	13.29	2.75

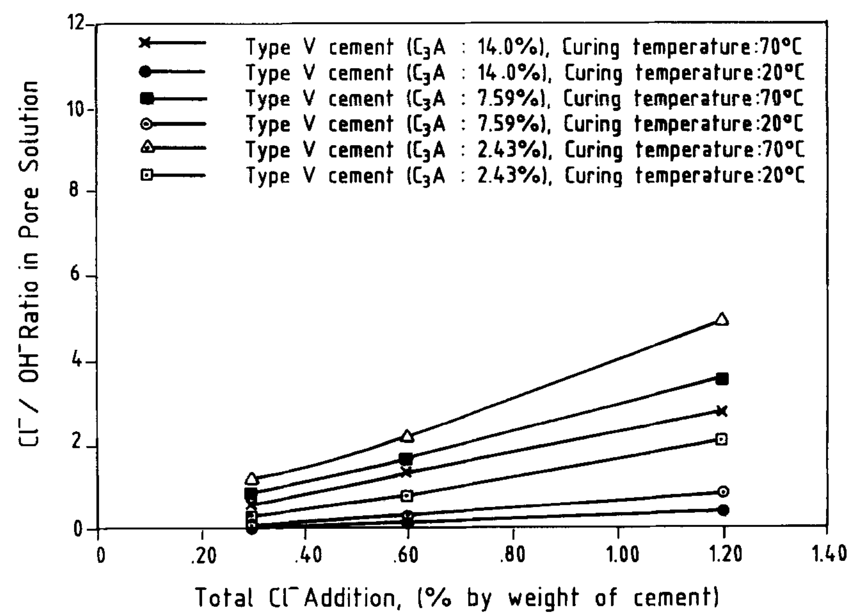

FIG. 5. Effect of Temperature on Cl^-/OH^- Ratio in Pore Solutions of Different C_3A, Cement Treated with Different Levels of Chloride.

TABLE 9

Effect of Exposure Temperature on Threshold Chloride

Cement No.	C_3A Content of Cement (% by weight	Exposure Temperature	Threshold Chloride Content (% by weight of cement)
1	2.43	20	0.35
1	2.43	70	0.04
2	7.59	20	0.62
2	7.59	70	0.09
3	14.00	20	1.00
3	14.00	70	0.19

by cement hydrates. The net effect is a slight increase or decrease in the Cl^-/OH^- ratio and the threshold chloride value. The other implications of alkalies in cement are their effect on alkali silica reactivity (ASR) and setting time. In order to avoid any potential risk of ASR, codes restrict the cement alkali content to a maximum of 0.6% (Na_2O equivalent). Considering this fact and its relatively marginal effect on corrosion resistance, use of high alkali cements to mitigate reinforcement corrosion does not appear advisable.

Data on sulfate-chloride interaction show that the effect of sulfates on threshold chlorides is not consistent for all cements tested in this investigation. In 2.43% C_3A cement, increase of SO_3 content raises the threshold chloride content by about 50% whereas in case of 14% C_3A cement, threshold chloride is reduced by about 25% when SO_3 content is increased to 8%. As shown in our earlier paper (13), this inconsistent behavior is attributable to the alkali and C_3A contents of cement. Using a conservative approach, it may be presumed that the presence of sulfates along with chlorides in concrete reduces the threshold chloride and hence increases the risk of corrosion. However, the magnitude of this increase in corrosion risk is moderate. It should be noted that

TABLE 10

Effect of Various Parameters on Threshold Chloride

Cement No.	Cement Type	Parameter	Threshold Chloride Content (% by weight of cement)
3	I	0.65% Alkali content	1.00
3	I	1.20% Alkali content	0.90
1	V	1.71% SO_3 content	0.35
1	V	4% SO_3 content	0.53
1	V	8% SO_3 content	0.54
3	I	2.6% SO_3 content	1.00
3	I	4% SO_3 content	0.93
3	I	8% SO_3 content	0.78
1	V	20 ^oC Exposure Temperature	0.35
1	V	70 ^oC Exposure Temperature	0.04
3	I	20 ^oC Exposure Temperature	1.00
3	I	70 ^oC Exposure Temperature	0.19

results of this investigation are valid for sodium sulfate salt. The effect of sulfate ions derived from other salts such as sulfates of magnesium or calcium may be quite different.

The factor which has been found to affect threshold most is the exposure temperature of concrete. Threshold chloride contents are reduced by at least five times for all three cements tested in this investigation. The effect of temperature on threshold chloride is twofold. On the one hand, increase in temperature reduces OH^- concentration of the pore solution, on the other hand, it increases the free chloride concentration of the pore solution by causing a decomposition of calcium chloroaluminate and other compounds in which chlorides have been complexed with cement hydrates. The net effect is a sharp increase in the Cl^-/OH^- ratio and also a reduction in threshold chloride value. The other important factor relevant to reinforcement corrosion is rates of chloride ingress as well as corrosion reactions at elevated temperatures. Lower chloride levels required for the onset of corrosion, coupled with increase in the rates of chloride ingress and corrosion reactions, are expected to adversely affect the reinforcement corrosion performance of concrete structures in hot climate environment such as the Gulf region due to prevailing high temperatures.

Conclusions

1. C_3A content of cement has a significant beneficial effect on threshold chloride content and reinforcement corrosion resistance. An increase in C_3A content of cement from 2.43% to 14% raises threshold chloride 2.85 fold.

2. Alkali content of cement also affects threshold chloride content, but the effect is relatively marginal. Due to potential risk of ASR, use of high alkali cement for mitigating corrosion does not seem advisable.

3. Presence of sulfates has a moderate effect on threshold chloride values. Depending upon the chemical composition of cement, presence of sulfates either moderately increases or decreases the threshold chloride content. Adopting a conservative approach, the presence of sulfates along with chlorides in concrete should be considered to have lowered the threshold chloride values by about 25%.

4. Exposure temperature has a very strong influence on threshold chloride values. Increase in temperature from 20° to 70°C causes 5 fold reduction in threshold chlorides. Also, high temperatures are expected to reduce corrosion resistance by increasing the rates of chloride ingress and corrosion reactions.

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