



Long-term chloride diffusion in silica fume concrete in harsh marine climates

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ABSTRACT

Supplementary cementitious materials such as silica fume are typically necessary for producing high performance concrete for marine environments in hot regions, such as the Persian Gulf. Silica fume use generally improves the strength and/or durability properties of the concrete. This paper investigates the effects of silica fume on various properties of concrete specimens that were exposed to Persian Gulf conditions. Samples were taken at the ages of 3, 9 and 36 months and analyzed to determine the chloride diffusion coefficient. The results show that partial cement replacement with up to 7.5% silica fume reduces the diffusion coefficient, whereas for higher replacement rates the diffusion coefficient does not decrease significantly. Also time-dependent chloride diffusion and compressive strength of concrete containing silica fume are investigated.

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1. Introduction

The Persian Gulf region is known to be one of the most aggressive environments for reinforced concrete constructions. Chloride-induced corrosion is believed to be the main reason for premature deterioration, and sometimes failure, of reinforced concrete structures constructed in this region [1–3]. The harsh climate of the Persian Gulf region can be described as having high temperatures, high evaporation rates and consequently high salinity of water, which increases the chloride induced reinforcement corrosion rates [4,5]. Typical extreme temperatures range from about 50 °C down to 3 °C and the relative humidity may be as high as 95% or as low as 5% [6].

The quality of concrete, mainly the permeability, nature and intensity of cracks, and the cover thickness, have a great bearing upon the initiation and sustenance of reinforcement corrosion [7]. Poor quality of concrete combined with aggressive environmental conditions has exposed inadequacies in design: some structures have had alarming degrees of degradation within less than a decade [8]. Application of supplementary cementitious materials (SCM) such as silica fume (SF) and fly ash (FA) to extend the service life of infrastructures has significantly increased in Iranian concrete practices of recent decades [9,10].

The SF is an efficient pozzolan that forms reaction products with the lime from hydrated cement and reduces the volume of large pores and capillaries found in cement paste. Consequently a more dense concrete is obtained. Neville [6] suggests a certain maximum

content of 8–10% of cementitious materials for SF content in the Persian Gulf region. As using silica fume induces a tendency for plastic shrinkage in concrete, precautions should be taken to avoid early moisture loss and any ensuing cracking [11].

This paper presents the results of an extensive experiment carried out as a part of a contract with Iran's Management and Planning Organization to develop a new service life design model for durability-based design in the Persian Gulf, which would provide a more realistic prediction of corrosion initiation for RC structures in this region [12]. This model is based on the results obtained from a complete set of field investigations on chloride diffusion, and considers parameters such as water to cement ratio, silica fume content, curing condition, exposure condition, environment temperature and surface coating. Detailed information on the project can be found elsewhere [13,14].

The results of investigations on the long-term effects of silica fume and water to cementitious materials ratio on durability performance of concrete exposed to the tidal zone of Persian Gulf environment are presented. Moreover, the decrease in the diffusion coefficient with time and improvement in compressive strength of concrete are studied.

2. Experimental program

2.1. Materials

The cementitious materials used in this study were Portland cement (PC) equivalent to ASTM Type II, and silica fume (SF) obtained from Azna ferro-silicon alloy manufacture. The chemical and physical properties of these materials are given in Table 1. The

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Table 1
Chemical and physical properties of portland cement and silica fume.

	Cement	Silica fume
<i>Physical tests</i>		
Specific surface, blaine (m ² /kg)	290	
Initial setting time (min)	145	
Final setting time (min)	210	
Compressive strength of 50 mm cube (MPa)	3-day	18
	7-day	29
	28-day	40
		–
Autoclave expansion (%)	0.2	
<i>Chemical analyzes (%)</i>		
Silicon dioxide (SiO ₂)	20.03	93.16
Aluminum oxide (Al ₂ O ₃)	4.53	1.13
Ferric oxide (Fe ₂ O ₃)	3.63	0.72
Calcium oxide (CaO)	60.25	–
Magnesium oxide (MgO)	3.42	1.6
Sodium oxide (Na ₂ O)	–	–
Potassium oxide (K ₂ O)	–	–
Sulfur trioxide (SO ₃)	2.23	0.05
Loss on ignition	1.37	1.58
<i>Bogue potential compound composition (%)</i>		
Tricalcium silicate C ₃ S	49.9	
Dicalcium silicate C ₂ S	23.5	
Tricalcium aluminate C ₃ A	6.6	
Tetracalcium aluminoferrite C ₄ AF	10.9	

aggregates used were crushed limestone from Metosak plant and were graded according to ASTM C 33 [15]. The coarse aggregate had maximum size of 12.5 mm and specific gravity and absorption values of 2.79% and 1.9%, respectively. The fine aggregate had specific gravity and absorption values of 2.59% and 3.2%, respectively. The fineness modulus of fine aggregates was 3.2. Polycarboxylate ether polymer superplasticizer and lignosulphonate plasticizer were used for the mixes in order to improve the workability of fresh concrete.

2.2. Mixture proportions

Four separate series of concrete mixes were developed at the water to cementitious ratios of 0.35, 0.40, 0.45 and 0.50. All series

included four SF mixes with 5%, 7.5%, 10% and 12.5% silica fume and the control mix without any SF. All series were proportioned to have the same total cementitious materials content of approximately 400 kg/m³ which is typical of those used in the marine concrete structures in Iran. The details of the concrete mixtures are given in Table 2.

2.3. Specimen preparation, casting and curing

The concrete mixtures were prepared in the laboratory of the Construction Materials Institute at the University of Tehran using a 0.1 m³ countercurrent pan mixer. The freshly mixed concrete was tested for air content according to ASTM C 231 [16], pressure method; slump according to ASTM C 143 [17]; and unit weight in accordance with ASTM C 138 [18]. Properties of fresh concrete are summarized in Table 3. Cubes of 150 × 150 × 150 mm and prisms of 150 × 150 × 600 mm in dimension were cast in steel mold and compacted on a vibrating table. The 150 mm cubes were used for the determination of compressive strength while the prisms were used to test chloride diffusion. The molds were covered with burlap kept wet for 24 h after casting. The specimens were removed from the molds and were allowed to cure in water saturated with calcium hydroxide at 21 °C for 28 days. After the curing period the prism specimens were sealed on four sides using epoxy polyurethane coating to ensure one-dimensional diffusion as illustrated in Fig. 1.

2.4. Exposure condition

The specimens were moved to an investigation site located in Bandar-Abbas, Iran and were continuously subjected to tidal zone exposure condition in Persian Gulf (Fig. 2) for the entire period of investigations (36 months). Exposure site is situated at the general water levels of low tide periods, so that specimens are exposed to atmosphere for about 2 h, simulating the tidal zone condition. Due to the location of the site and the specimen dimensions, no significant drying occurs.

The Gulf water is highly saline due to its enclosed condition (mostly surrounded by lands of Iran and Arabian Peninsula) and the high evaporation rate. Chemical analysis of the sea water is

Table 2
Details of the concrete mixtures.

Series	Code	w/cm	SF (%)	Water (kg/m ³)	Binder (kg/m ³)		Fine aggregates (kg/m ³)	Coarse aggregates (kg/m ³)	Superplasticizer (kg/m ³)
					Cement	SF			
1	SF0W1	0.35	0	140	400	–	936	973	7.2
	SF1W1	0.35	5	140	380	20	931	968	–
	SF2W1	0.35	7.5	140	370	30	931	968	6.0
	SF3W1	0.35	10	140	360	40	906	968	6.4
	SF4W1	0.35	12.5	140	350	50	929	964	4.8
2	SF0W2	0.40	0	160	400	–	836	1022	2.2
	SF1W2	0.40	5	160	380	20	833	1018	3.6
	SF2W2	0.40	7.5	160	370	30	832	1017	3.2
	SF3W2	0.40	10	160	360	40	830	1014	3.6
	SF4W2	0.40	12.5	160	350	50	829	1012	3.8
3	SF0W3	0.45	0	180	400	–	815	997	2.4*
	SF1W3	0.45	5	180	380	20	810	990	1.2
	SF2W3	0.45	7.5	180	370	30	808	998	2.0
	SF3W3	0.45	10	180	360	40	807	985	1.8
	SF4W3	0.45	12.5	180	350	50	806	983	2.0
4	SF0W4	0.50	0	200	400	–	778	956	1.2
	SF1W4	0.50	5	200	380	20	793	991	0.8
	SF2W4	0.50	7.5	200	370	30	784	959	1.2
	SF3W4	0.50	10	200	360	40	820	1020	1.8
	SF4W4	0.50	12.5	200	350	50	782	955	2.0

* In this mix lignosulphonate plasticizer was used instead of polycarboxylate ether polymer superplasticizer.

Table 3
Properties of fresh concrete.

Series	Code	Density (kg/m ³)	Slump (mm)	Air content (%)
1	SF0W1	2467	936	2.8
	SF1W1	2460	931	3.5
	SF2W1	2400	931	3.6
	SF3W1	2370	906	3.1
	SF4W1	2340	929	3.0
2	SF0W2	2422	836	2.9
	SF1W2	2420	833	3.0
	SF2W2	2430	832	3.0
	SF3W2	2445	830	2.9
	SF4W2	2370	829	3.1
3	SF0W3	2440	815	3.1
	SF1W3	2410	810	2.6
	SF2W3	2390	808	2.6
	SF3W3	2400	807	2.1
	SF4W3	2350	806	2.6
4	SF0W4	2370	778	2.7
	SF1W4	2350	793	3.6
	SF2W4	2355	784	1.6
	SF3W4	2390	820	2.5
	SF4W4	2255	782	2.4

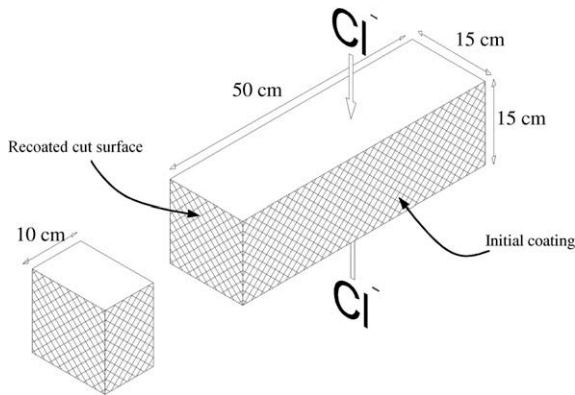


Fig. 1. Initial concrete prism specimens + cuts for laboratory tests.

shown in Table 4. The climatic factors of region include large fluctuations in daily and seasonal temperature and humidity regimes. Temperature can vary by as much as 30 °C during a typical summer day and relative humidity can range from 40% to 100% within 24 h [19].

2.5. Sampling and testing

Sampling was carried out at the ages of 3, 9 and 36 months. Each time, 100 mm was cut from end of the prism specimens. The cut surface of the remaining part was coated as illustrated in Fig. 1 and the specimen was moved back to the exposure condition for future sampling. The 150 × 150 × 100 mm slices were taken to the laboratory for the determination of chloride diffusion at the above ages.

A nominal 45 mm diameter core was taken from each slice for chloride concentration profiles. Each core was grinded in 8 increments from the finished surface to an estimated depth of chloride penetration. The fine particles from each ground layer were collected and analyzed separately for acid-soluble chloride according to ASTM C 1152 [20] (Standard Test Method for Acid-Soluble Chloride in Mortar and concrete) and ASTM C 114 [21], part 19. The cross-sectional area of a 45 mm diameter core is large enough to represent the concrete so that there is no need to be concerned about variations from sample to sample due to varying aggregate



Fig. 2. CMI exposure site in Persian Gulf tidal zone specimens.

Table 4
Chemical analysis of sea water.

Element	Chemical analysis of Persian Gulf water (ppm)
Ca ⁺⁺	480
Mg ⁺	1600
Na ⁺	12,600
K ⁺	470
SO4 ⁻	3300
Cl ⁻	23,400

contents. The 150 mm cubes were tested in compression at 7, 28 and 154 days with accordance with DIN 1048 [22].

3. Results and discussion

3.1. Chloride diffusion in silica fume concrete

An apparent diffusion coefficient for each concrete mix is determined by fitting profiles of acid-soluble chloride concentration versus depths according to Fick's second law of diffusion (Eq. (1) and (2) [23].

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \quad (1)$$

and

$$C(x, t) = C_0 \left(1 - \operatorname{erf} \left(\frac{x}{2\sqrt{D \cdot t}} \right) \right) \quad (2)$$

where D is the chloride diffusion coefficient, C_0 is the equilibrium chloride concentration on concrete surface and $C = C(x, t)$ is the chloride content at the depth of x from the surface at the time t .

Fick's second law for one-dimensional diffusion, as shown in Eq. (1), is a special case of a more generalized model of diffusion where concrete is assumed to be a homogenous material; chloride

concentration at the exposure surface is considered constant; no chemical or physical binding between the diffusing species and material occurs; and the effect of co-existing ions is constant. However, some of these assumptions are violated as concrete is not homogenous, the diffusion coefficient is time-dependant due to the hydration process, and tricalcium aluminates within cement paste may bind chloride ions.

Some of the chloride bound to the cement gel may be released when parameters of pore liquid change, as e.g. pH value, temperature and pressure. Therefore it is most convenient to determine the chloride of the concrete as acid-soluble chloride [24,25]. On the other hand, these limitations of analysis may be neglected as measured data are used for comparison purposes within the same set of exposure conditions.

The evolution of the diffusion coefficient for different silica fume contents at sampling ages of 3, 9 and 36 months is represented in Fig. 3. For each specimen, at the time of testing, a single measurement of chloride concentration and diffusion coefficient has been done. It is clear from the curves of Fig. 3 that the diffusion coefficient of concrete incorporating 5% silica fume decreases up to 50% compared to the control concrete without silica fume. Also it can be found that higher percentages of silica fume do not decrease the diffusion coefficient significantly.

It appears that there is an optimum silica fume content of 7.5% by weight of cement beyond which additional silica fume does not produce additional benefits in line with the additional costs. Tout-

anji [26] cites that the addition of 5% silica fume results in a significant reduction in permeability while the increase of SF content from 5% to 10% had a little effect on the reduction of permeability. According to Detwiler et al. [27], most of the benefit in reducing diffusion coefficient occurs with increase in silica fume content from 0% to 6% to 8% by weight of cement and further increases in SF content provide little additional benefit.

Samples containing higher contents of silica fume do not follow the general trend of decreasing diffusion coefficient. This may be due to fast drying, which in turn leads to propagation of micro-cracks within the fabric of concrete specimens. It can be found from Fig. 3 that variation of diffusion coefficient with time is more significant in higher water to cementitious materials ratio than lower ones.

To calculate the chloride diffusion coefficient with increasing silica fume content, the following equation can be used [23].

$$D_{sf} = D_{pc} \times e^{-m \cdot (SF\%)} \quad (3)$$

where D_{sf} represents diffusion coefficient of silica fume concrete; D_{pc} represents diffusion coefficient of Portland cement concrete; SF represents silica fume content (percent by weight of cement) and m is a coefficient given in Table 5. Thomas has derived a similar equation to calculate the diffusion coefficient of silica fume concrete, in which the value of m is considered to be 0.165 [28].

Relative diffusion coefficient versus silica fume content is plotted in Fig. 4. To make a general comparison between several

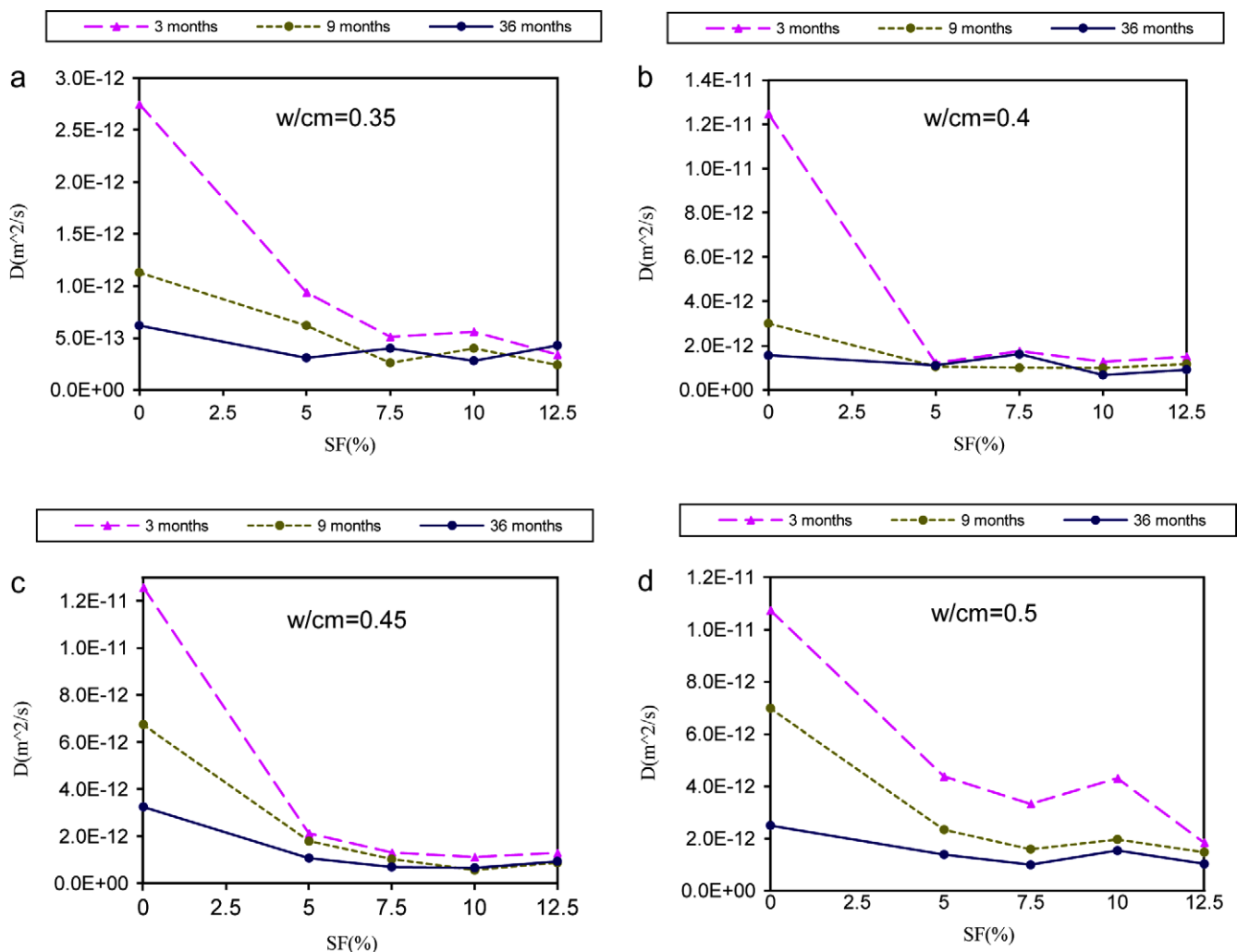


Fig. 3. The evolutions of the diffusion coefficient for different silica fume contents at sampling ages of 3, 9 and 36 months with a w/cm of (a) 0.35. (b) 0.40. (c) 0.45. (d) 0.50.

Table 5Values of m coefficient.

w/cm	Chloride exposure period (months)					
	36		9		3	
	m	r^2	m	r^2	m	r^2
0.35	−0.061	0.373	−0.133	0.932	−0.197	0.984
0.40	−0.044	0.444	−0.129	0.793	−0.339	0.957
0.45	−0.175	0.907	−0.244	0.987	−0.310	0.983
0.50	−0.081	0.726	−0.169	0.933	−0.138	0.918

coefficients m , not considering the effect of water to cementitious ratio, an exponential regression analysis of the data is used to determine the value of m . It can be found from Fig. 4 that the value of m decreases as the age of specimens increases. In other words, the effect of silica fume on performance of concrete against chloride ion ingress decreases with time. However, using silica fume at the earlier ages will decrease the diffusion coefficient significantly and thus a more durable concrete would be obtained. It can be seen in Fig. 4 that the coefficient m correlated better with silica fume content at earlier ages of specimens than at later ages. This might be due to that for a constant laboratory error, a smaller diffusion coefficient is less accurate than a greater one.

3.2. Time-dependent chloride diffusion in concrete

The chloride diffusion coefficient of a concrete is time dependent. It will typically decrease as time passes since the capillary pore system will be altered as hydration products continue to form [29]. The time dependency of the chloride diffusion coefficient can be expressed mathematically by a power function [26–30]:

$$D(t) = D_{\text{ref}} \left(\frac{t}{t_{\text{ref}}} \right)^{-a} \quad (4)$$

where $D(t)$ represents diffusion coefficient at time t ; D_{ref} represents diffusion coefficient at some reference time (3 months); a represents a constant, dependent on such variables as the type of cementitious materials used and the mixture proportions, and has been developed to determine the rate of reduction of diffusivity with time [19]. Bentz noted that the value of a is variable with time [30]. Other researchers have reported various values for the reduction coefficient [23].

Table 6 presents the time-dependent reduction coefficient, a , calculated for the twenty mixes.

Relative diffusion coefficient versus time for all of concrete specimens is plotted in Fig. 5. The results show that concrete mixtures with the higher silica fume content have a lower reduction coefficient. However in concrete with w/cm ratio of 0.5, no

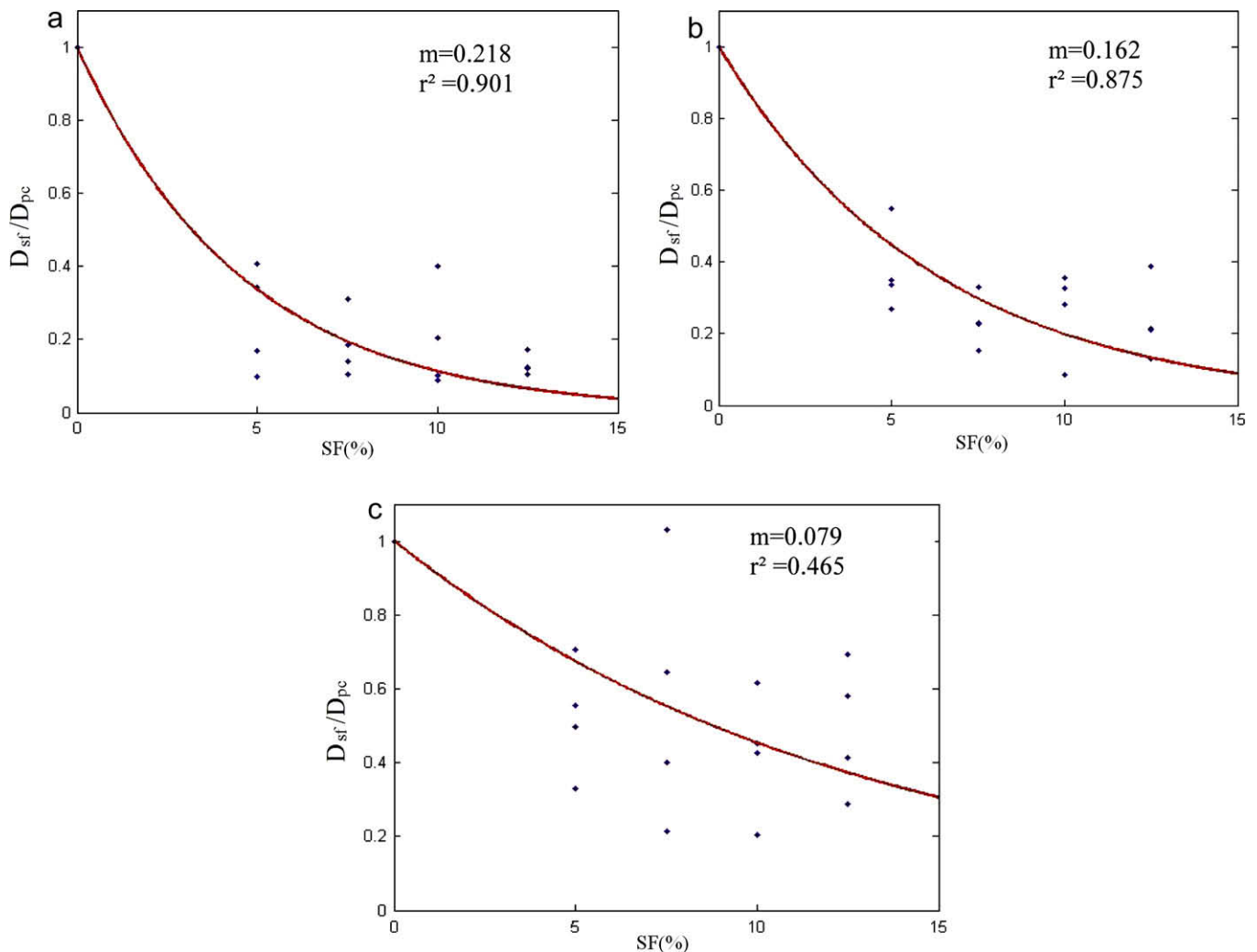


Fig. 4. Relative diffusion coefficients versus silica fume content at (a) 3 months. (b) 9 months. (c) 36 months.

Table 6
Time-dependent reduction coefficient for concrete specimens.

Series	Code	a	r^2
1	SF0W1	0.713	0.984
	SF1W1	0.422	0.993
	SF2W1	–	–
	SF3W1	0.287	0.998
	SF4W1	–	–
2	SF0W2	1.184	0.986
	SF1W2	–	–
	SF2W2	–	–
	SF3W2	0.256	0.999
	SF4W2	0.211	0.993
3	SF0W3	0.555	0.999
	SF1W3	0.239	0.938
	SF2W3	0.241	0.991
	SF3W3	–	–
	SF4W3	–	–
4	SF0W4	0.494	0.972
	SF1W4	0.508	0.989
	SF2W4	0.569	0.975
	SF3W4	0.534	0.924
	SF4W4	0.224	0.993

* No regression was made due to increase in diffusion coefficient at 36 months.

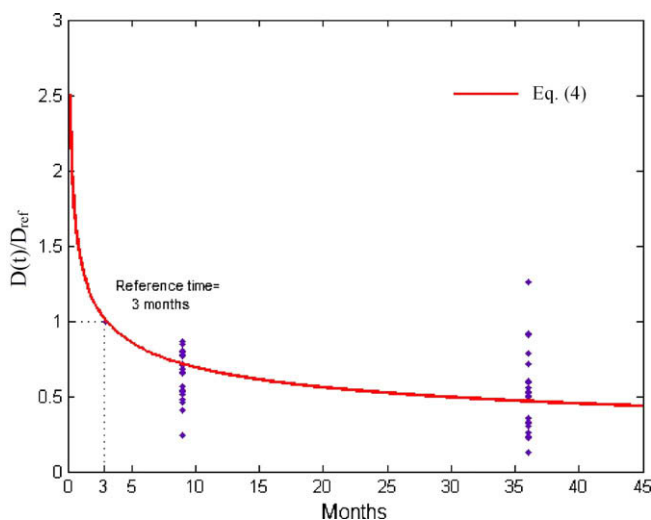


Fig. 5. Relative diffusion coefficient versus time for all of the concrete specimens.

significant variation in reduction coefficient is observed. A reason for this might be that for higher w/cm ratios, the effect of SF on diffusion coefficient is less significant and thus the reduction coefficient would be reduced. To obtain a unique reduction coefficient a power regression analysis of the data is used, which yields a value of 0.307 for a ($r^2 = 0.4988$).

3.3. Compressive strength

Compressive strength of concrete specimens is determined according to DIN 1048. The results are shown in Table 7, where each value is averaged from the results of three cubes. Specimens of 7 and 28 days old were cured in laboratory conditions, while the 154-day specimens were exposed to Persian Gulf conditions after 28 days of curing.

It can be found from the results that using silica fume will increase the rate of strength development at early ages, while it does not improve the final strength of concrete significantly. Results also show that optimum silica fume content for compressive

Table 7
Compressive strength of concrete specimens.

Series	Code	Compressive strength (MPa)		
		7 days	28 days	154 days
1	SF0W1	53.2	51.5	74.6
	SF1W1	49.8	66.8	83.9
	SF2W1	50.6	74.8	89.0
	SF3W1	57.9	79.6	91.3
	SF4W1	51.6	64.6	63.6
2	SF0W2	41.4	50.0	67.3
	SF1W2	47.4	69.8	74.8
	SF2W2	44.0	65.3	70.5
	SF3W2	47.3	71.7	80.7
	SF4W2	43.2	66.9	77.6
3	SF0W3	36.3	44.9	66.5
	SF1W3	38.3	57.9	66.2
	SF2W3	37.8	59.6	62.7
	SF3W3	35.4	55.0	57.7
	SF4W3	37.9	65.9	69.2
4	SF0W4	25.9	33.9	50.6
	SF1W4	37.3	50.4	68.8
	SF2W4	34.4	53.9	61.6
	SF3W4	27.7	44.2	43.6
	SF4W4	24.7	49.1	38.4

strength is 7.5–10% by weight of cement. Appa Rao [31] reported a similar pattern for mortars containing silica fume.

4. Conclusion

The effects of silica fume on resistance to chloride diffusion and compressive strength of concrete were investigated in this study, which considered long-term exposure to Persian Gulf conditions. The following conclusions can be drawn:

1. The partial replacement of cement with silica fume results in a concrete with an appreciably lower chloride permeability than concrete containing only Portland cement as the cementitious material. The chloride diffusion coefficient can be expressed as an exponential function of silica fume content.
2. The reduction in diffusion coefficient is not proportional to the increase in silica fume content. The addition of 7.5% silica fume resulted in a significant reduction in chloride diffusion into concrete. However, an increase of silica fume content from 7.5% to 12.5% exhibited little effect on the reduction in permeability.
3. The time-dependent reduction coefficient a is calculated in this paper by using the power function proposed by Mangat and Molloy [24,26–29]. Results show that the reduction coefficient is lower in concrete mixtures with a higher silica fume content.
4. The rate of strength development is higher in silica fume concrete than concrete containing no SF. However, the final strength does not improve significantly. Results show an optimum silica fume content of 7.5–10% by weight of cement.

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