Numerical technique for chloride ingress with cover concrete property and time effect

Bang Yeon Lee[1a], Mohamed A. Ismail[2b], Hyeok-Jung Kim[3c],
Sung-Won Yoo[4d] and Seung-Jun Kwon[*5]

[1]*School of Architecture, Chonnam National University, 77 Yongbong-no, Buk-gu, Gwangju, 61186, Republic of Korea*
[2]*Department of Civil and Construction Eng., Faculty of Engineering and Science,
Curtin University Malaysia, CDT 250, 98009 Miri, Sarawak, Malaysia*
[3]*Elastomer TS&D Team, Kumho Petrochemical R & BD Center, 15577 Yuseong-daero,
Yuseong-gu, Daejeon, 34044, Republic of Korea*
[4]*Department of Civil and Environmental Eng., Gachon University, 1342 Seongnam Daero, Seongnam-Si 13120, Republic of Korea*
[5]*Department of Civil and Environmental Eng., Hannam University, 133 Ojeong-dong, Daejeon, 34430, Republic of Korea*

Abstract. Durability problems initiated from steel corrosion are unseen but critical issues, so that many researches are focused on chloride penetration evaluation. Even if RC (Reinforced Concrete) structures are exposed to normal environment, chloride ingress varies with concrete surface conditions and exposed period. This paper presents an analysis technique for chloride behavior evaluation considering time effect on diffusion and surface conditions assumed as double-layered system. For evaluation of deteriorated surface condition, field investigation was performed for concrete pavement exposed to deicing agent for 18 years. In order to consider enhanced surface concrete, chloride profiles in surface-impregnated concretes exposed to chloride attack for 2 years from previous research were investigated. Through reverse analysis, effectively deteriorated/ enhanced depth of surface and the related reduced/enlarged diffusion coefficient in the depth are simulated. The proposed analysis technique was evaluated to handle the chloride behavior more accurately considering changes of chloride ingress within surface layer and decreased diffusion coefficient with time. For the concrete surface exposed to deicing agent, the deteriorated depth and enlarged diffusion coefficient are evaluated to be 12.5~15.0 mm and 200% increasing diffusion coefficient, respectively. The results in concrete containing enhanced cover show 10.0~12.5 mm of impregnated depth and 85% reduction of chloride diffusion in tidal and submerged conditions.

Keywords: diffusion; durability; double-layer; deicing agent; chloride attack; surface impregnation

1. Introduction

Concrete, a unique construction material with stable material supply, high durability, and cost-benefit, is one of the most attractive materials. It allows very small ultimate strain and low tensile strength compared with structural steel, so that the reinforcements with ductility and high tensile strength are required in the tensile region. The embedded steel in concrete is usually protected from corrosion due to high alkalinity of passive film (Broomfield 1997, RILEM 1994), however durability problem starts with corrosion when RC structures are exposed to harsh environments like chloride attack and carbonation. Among the causes of deterioration due to steel corrosion, chloride attack is one of the most critical causes since chloride ions from sea, underground water, and deicing agent directly

affect steel corrosion through pitting on the steel, which propagates to cracking, delamination of concrete cover, and significant threat to structural safety (Broomfield 1997, Jung *et al.* 2003, Kwon and Kim, 2012). The mechanism of chloride transport is very complicated since the dissolved chloride ions come into concrete through diffusion due to concentration gradient and convection due to moisture pressure gradient (Maekawa *et al.*2003, Park *et al.* 2012). During the penetrating process of chloride ions, some of them can be absorbed in cement hydrates as bound chlorides and the others can intrude as free chlorides which affect corrosion directly (Ary *at al.* 1990). Several numerical models based on Nernst-Einstein equation have been proposed with adopting diffusion and permeation of free chloride, isotherm of bound chloride, varying exterior chloride ions, and early-aged concrete behavior such as porosity and saturation (Ishida and Maekawa 2003, Maekawa *et al.* 2003, Song *et al.* 2006). The models have been recognized to be very effective but the mechanisms like ion condensation under freezing/ thawing actions, threshold of pore size which can block the ion transport, and quantification of varying local condition in concrete are still under discussion. The numerical approaches also have been attempted for evaluation of chloride behavior in concrete with different mix conditions (Tsao *et al.* 2015,

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*Corresponding author, Ph.D.
  E-mail: jjuni98@hannam.ac.kr
[a] E-mail: bylee@jnu.ac.kr
[b] E-mail: m.abdelkader@curtin.edu.my
[c] E-mail: ceasare@kkpc.com
[d] E-mail: imysw@gachon.ac.kr

Paul *et al.* 2014) and weak region like cracked concrete (Yang *et al.* 2017, Park *et al.* 2012). Furthermore, the numerical technique on chloride evaluation has been developed considering non-steady boundary condition of surface chlorides (Ye *et al.* 2015) and integration of mechanical behavior of concrete cracking due to steel corrosion (Hossenini *et al.* 2015).

Fick's second Law is usually adopted for an evaluation of chloride behavior and this technique provides very simple but clear answer of total chloride amount even if it cannot consider isotherm of free chloride, variation of humidity, and local conditions of surface chlorides (Kwon and Kim 2012). The governing equation based on Fick's second Law assumes steady-state condition of mass transport, so that several simple parameters are adopted such as time effect on diffusion, built-up of surface chloride content, and skin effect on diffusion for simulation of the actual chloride behavior (Tang and Joost 2007, Thomas and Bamforth 1999, Poulsen 1993, Zhu *et al.* 2016).

The performance of concrete surface can be enhanced or deteriorated depending on its exposure conditions. When concrete surface is exposed to deicing agent under freezing and thawing attack, it permits micro cracks and pop-out of aggregates, and then enlarged chloride ions dissociated from deicing agent ($CaCl_2$ or $NaCl$) can penetrate into concrete through the deteriorated surface (Colombo *et al.* 2015, Jiang *et al.* 2015). The damages from combined deterioration with chloride attack and freezing/thawing action are reported to be getting severe since concrete cover contains enlarged chloride diffusion due to micro cracks and surface scaling (Yang *et al.* 2006, Metha and Monteiro 1993).

Among the repairing techniques, surface impregnation technique has widely attempted due to its simple application and preventing damage for existing structures (Metha and Monteiro 1993, Park *et al.* 2014). The main advantage of surface impregnation is the full attachment of old concrete without peeling-off since the reproduced CSH (Calcium-Silicate-Hydrates) in impregnated depth is simply filling up the previous pores and this can improve pore connectivity and resistance to harmful ion intrusion (Metha and Monteiro 1993, Moon *et al.* 2007, Alghamri *et al.* 2016). In the surface-impregnated thickness, diffusion coefficient is reduced and the chloride penetration is also hindered due to partial block of surface protection. The skin effect of chloride diffusion with double-layered concrete was already proposed (Andrade *et al.* 1997), however its application is very limitedly performed due to difficulties in comparing the field investigation results. The previous theory can be improved through considering time effect on chloride diffusion (Tang and Joost 2007, Thomas and Bamforth 1999).

In this paper, skin effect theory for double-layered concrete is adopted and improved through constructing time-dependent diffusion characteristics. For applicability of the proposed technique, field investigation is performed and the reverse analysis is conducted for the enlarged diffusion in concrete exposed deicing agent. For the reduced diffusion in enhanced concrete surface, the previous test results of concrete with surface impregnation

was utilized and reverse analysis is also performed. In the work, applicability of the proposed technique with double layer and time dependent diffusion is verified, and the chloride ingresses depending on concrete surface conditions are discussed.

2. Effects of time and surface layer on chloride diffusion

2.1 Time effect on chloride diffusion

Chloride diffusion means an ion transport due to a gradient of concentration difference (Maekawa *et al.* 2009). With hydration of cement, pores and their connectivity are improved, and it impedes chloride penetration. The decreasing diffusion manipulated by time can be explained by the reduced difference of chloride concentration between outer and inner concrete due to reduced porosity, densified pore connectivity, and enlarged cement hydrates which can bind more chloride ions (Maekawa *et al.* 2009, Hussain and Ishida 2011). For the sake of simplicity of modeling, diffusion coefficient is usually assumed to decrease with time as equation (1) (Tang and Joost 2007, Thomas and Bamforth 1999, Al-alaily and Hassan 2016)

$$D(t) = D_{ref} \left(\frac{t_{ref}}{t} \right)^m \tag{1}$$

where $D(t)$ is time dependent diffusion coefficient, D_{ref} is diffusion coefficient in reference time (t_{ref}), and m is exponent constant varying with mineral admixtures (Thomas and Bamforth 1999, Camacho *et al.* 2014). In the steady state condition with a given period, diffusion coefficient in Eq. (1) can be expressed as the Eqs. (2(a)) and (2(b)) considering total equivalent induced chlorides (Tang and Joost 2007, Kwon *et al.* 2009).

$$D(m) = \frac{D_{ref}}{1-m,} \left(\frac{t_0}{t} \right)^m \tag{2a}$$

$$D(m) = \frac{D_{ref}}{1-m} \left(\frac{t_0}{t_c} \right)^m \left[1 - m + m \left(\frac{t_c}{t} \right) \right] \tag{2b}$$

Where $D(m)$ is diffusion coefficient in fixed time and it is usually assumed to be constant after time t_c (=30 years) (Thomas and Bentz 2002).

2.2 Skin dffect on chloride diffusion

In the previous research, diffusion in double-layered concrete can be formulated as per Eqs. (3(a)) and (3(b)) (Andrade *et al.* 1997).

$$C_1(x,t) = C_s \sum_{n=0}^{\infty} \alpha^n \left[erfc \left\{ \frac{2ne+x}{2\sqrt{D_1 t}} \right\} - \alpha \cdot erfc \left\{ \frac{(2n+2)e-x}{2\sqrt{D_1 t}} \right\} \right] \tag{3a}$$
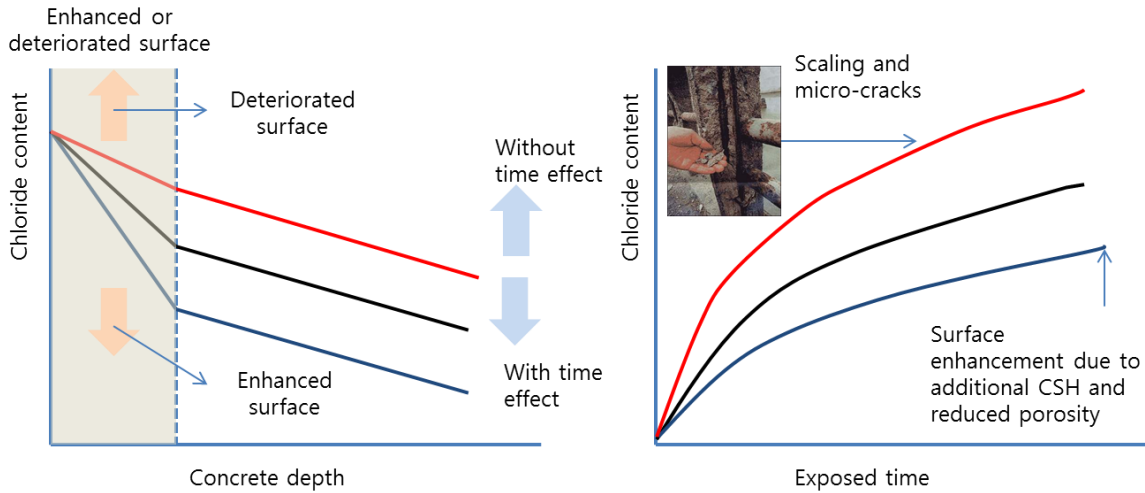
Fig. 1 Schematic diagram of chloride behavior with changing surface conditions

$$C_s(x,t) = \frac{2kC_s}{k+1} \sum_{n=0}^{\infty} \alpha^n erfc \frac{(2n+1)e+k(x-e)}{2\sqrt{D_1 t}} \quad (3b)$$

where C_1 and C_2 are chloride contents in surface and inner concrete, D_1 and D_2 are diffusion coefficients in surface and inner concrete. k and α are $\sqrt{D_1/D_2}$ and $(1-k)/(1+k)$, respectively.

This formulation can consider the skin effect which can handle enlarged or reduced diffusion, however ignoring of time effect on diffusion can lead to an overestimation of induced chloride contents.

3. Chloride diffusion in deteriorated and enhanced concrete surface

3.1 Governing equation of chloride penetration

Governing equation for explaining chloride penetration can be expressed as Eq. (4) based on Fick's 2nd Law (RILEM 1994, Al-alaily and Hassan 2016).

$$C(x,t) = C_s \left\{ 1 - erf\left(\frac{x}{2\sqrt{D\ t}} \right) \right\} \quad (4)$$

where C_s is surface chloride content and *erf* is error function.

The Nernst-Einstein equation which can consider migration, permeation, and chemical reaction of chloride ion can explain complicated chloride behavior more precisely based on the models of early-aged concrete behavior such as hydration, pore structure development, and saturation hysteresis (Maekawa *et al.* 2009, Ishida and Maekawa 2003). The integrated model is accurate, however it should have complicated framework for calculating ion transport, isotherm for binding capacity of chloride ion, and data allocation such as porosity, pore pressure, saturation,

and free/bound chloride ion (Mabrouk *et al.* 2004).

In this work, the simplistic approach using apparent diffusion coefficient is adopted for easy application of the proposed model. The schematic diagram for changing chloride behavior due to surface concrete conditions is shown in Fig. 1.

The Eqs. of (3(a)) and (3(b)) present the formulations on double-layered concrete. For considering time effect on diffusion coefficient, they can be expressed as Eqs. (5(a)) and (5(b)) through constructing Eqs. (2(a)) and (2(b)), respectively. Several assumptions are required for merging them such as 1] constant time effect on diffusion both for skin and inner concrete, 2] without loss of total chloride content between surface and inner concrete, and 3] steady-state condition of surface chloride content.

$$\begin{cases} C_1(x,t) = C_a \sum_{n=0}^{\infty} \alpha^n \left[erfc\left\{ \dfrac{2ne+x}{2\sqrt{\dfrac{D}{1m}\left(\dfrac{t_0}{t}\right)^m t}} \right\} - \alpha \cdot erfc \dfrac{(2n+2)e-x}{2\sqrt{\dfrac{D}{1-m}\left(\dfrac{t_0}{t}\right)^m t}} \right] \\ \\ C_2 = \dfrac{2kC_a}{k+1} \sum_{n=0}^{\infty} \alpha^{\setminus n} erfc\left[\dfrac{(2n+1)e+k(x-e)}{2\sqrt{\dfrac{D}{1-m}\left(\dfrac{t^0}{t}\right)^m t}} \right] \\ \\ (t < t_c) \end{cases} \quad (5a)$$

$$\begin{cases} C_1(x,t) = C_a \sum_{n=0}^{\infty} \alpha^n \left[erfc\left\{ \dfrac{2ne+x}{2\sqrt{\dfrac{D}{1-m}\left(\dfrac{t_0}{t_c}\right)^m \left[1-m+m\left(\dfrac{t_0}{t}\right)\right]t}} \right\} - \alpha \cdot erfc \dfrac{(2n+2)e-x}{2\sqrt{\dfrac{D}{1-m}\left(\dfrac{t_0}{t_c}\right)^m \left[1-m+m\left(\dfrac{t_c}{t}\right)\right]t}} \right] \\ \\ C_2 = \dfrac{2kC_a}{k+1} \sum_{n=0}^{\infty} \alpha^n erfc\left[\dfrac{(2n+1)e+k(x-e)}{2\sqrt{\dfrac{D}{1-m}\left(\dfrac{t_0}{t_c}\right)^m \left[1-m+m\left(\dfrac{t_c}{t}\right)\right]t}} \right] \\ \\ (t \geq t_c) \end{cases} \quad (5b)$$

(a)   (b)

Fig. 2 Surface scaling on concrete pavement: (a) Scaling of concrete pavement and (b) Scaling of guard rail concrete
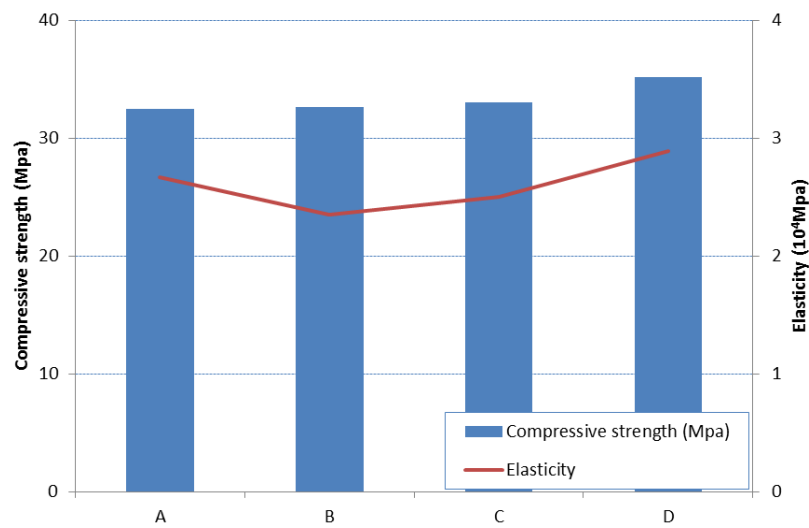

Fig. 3 Compressive strength and elasticity of the concrete core

Table 1 Measured diffusion coefficient and surface chloride content

Type	A	B	C	D	Average
Surface chloride content (kg/m^3)	3.65	3.63	3.78	3.97	3.76
Diffusion coefficient ($\times 10^{-12}$ m^2/sec)	1.14	0.90	1.31	0.92	1.07

3.2 Chloride behavior in concrete containing deteriorated surface

Field investigation was performed for concrete pavement exposed to deicing agent for 18 years. For 4 target pavements, concrete cores were obtained, and several tests were performed such as chloride profiles, compressive strength, and elasticity. Through AgNO$_3$ indicator, chloride penetration depth was measured to 10~25 mm and the deteriorated depth was 10~15 mm through scaling measurement. The deteriorated surface contains damaged area with scaling and micro cracks. The photos for damaged concrete from freezing and thawing are shown in Fig. 2.

Based on AASHTO T 260, total chloride contents were measured and the results are shown in Fig. 3. The surface chloride content in the sea-submerged condition usually shows very high value over 18.0 kg/m^3. It is reported that surface chloride content linearly increases to the maximum to 10~12 years and keeps almost constant (Thomas and Bentz 2002, JSCE 2002). In the test, surface chloride content was measured to be very low (be 3.65~3.97 kg/m^3) since deicing agent was spread only in the snowing season and disappeared. Through the regression analysis based Fick's second Law, surface chloride content and the related diffusion coefficients were derived as listed in Table 1. The average of compressive strength and Young's modulus values were evaluated to be 32.5 MPa and 2.60 $\times 10^4$ MPa, respectively. The results of physical properties from the cores are shown in Fig. 3 which shows typical range of strength and elasticity.
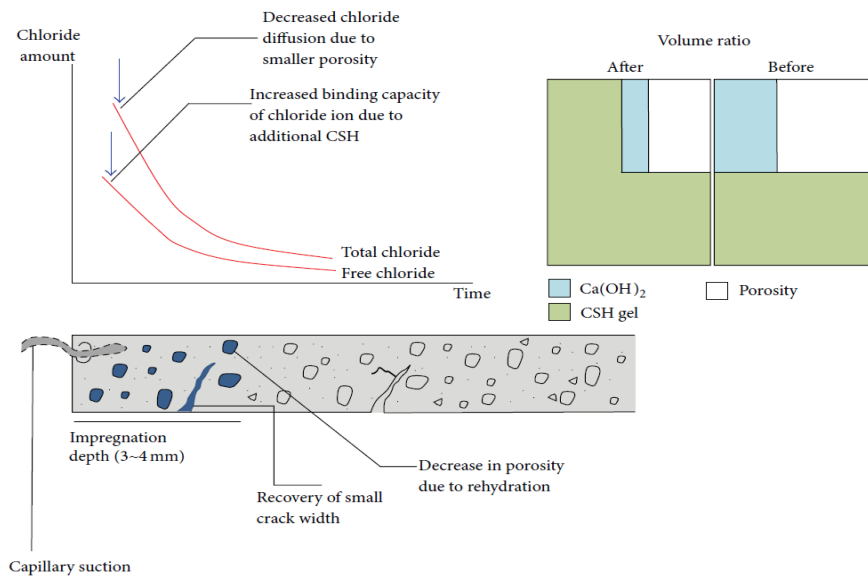
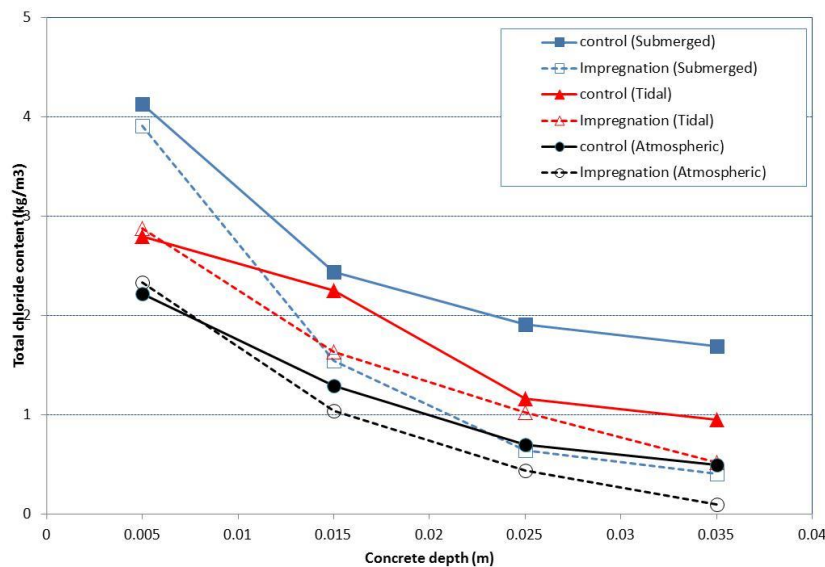Fig. 4 Durability improvement in surface-impregnated concrete (Park et al. 2014)

Fig. 5 Long-term exposure test for impregnated concrete (Park *et al.* 2014)

3.3 Chloride behavior in concrete containing enhanced surface

In this section, the previous test results were investigated (Park *et al.* 2014). Surface- impregnated concrete with sodium silicate was exposed to several sea exposure conditions for 2 years. Compressive strength was 21 MPa with impregnated depth of 8~12 mm. The chemical compositions and the related enhancing mechanism are well explained in the previous literatures (Park *et al.* 2014, Kwon *et al.* 2007). The used mix proportions are listed in Table 2 and the schematic diagram for surface impregnation is shown in Fig. 4.

The long-term exposure tests were conducted at atmospheric, tidal, and submerged zone for 2 years, and the chloride profiles for each exposure are represented in Figure 5. The surface impregnation is reported to be so effective that reduced chloride content is evaluated inside concrete (Park *et al.* 2014, Moon *et al.* 2007, Ohama 1997). The surface chloride content is decreasing from submerged to atmospheric condition due to low chloride concentration in salt spraying area (JSCE 2002).

Through regression analysis, surface chloride content and diffusion coefficient are evaluated as shown in Table 3. The diffusion coefficients were obtained from Fig. 6 through regression analysis, so that they are different from the results based on the theoretical models with material properties.
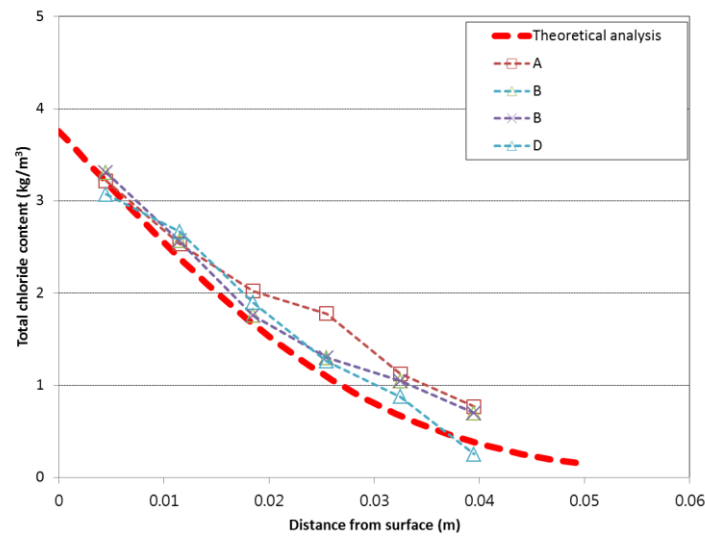
Fig. 6 Comparison of results between field investigation and theoretical analysis

Table 2 Mix proportions for base concrete for surface impregnation (Park *et al.* 2014)

Strength (MPa)	Slump (mm)	W/C* (%)	S/a* (%)	Unit weight (kg/m³)			
				W*	C*	S*	G*
21	150	55.4	45.8	166	267	810	979

* W/C: Water to Cement, S/a: Sand to total aggregate ratio, W: water, C: Cement, S: Sand, G: Gravel

Table 3 Diffusion coefficient and surface chloride content from concrete cores

Type	Atmospheric	Tidal	Submerged
Surface chloride content (kg/m³)	2.49	3.27	4.18
Diffusion coefficient ($\times 10^{-12}$ m²/sec)	4.50	6.87	9.53

4. Analysis technique for chloride penetration with time and surface effect

4.1 Simulation of chloride penetration for concrete with deteriorated surface

Analysis conditions should be reasonably assumed for chloride ingress evaluation in double-layered concrete. The concrete pavement contains w/b (water to binder) ratio of 0.4 and FA (Fly Ash) replacement ratio of 25%, which are allowable properties for concrete with 32 MPa. The mix proportions also agree with recommendation of Domestic Specification (KCI 2012). The apparent diffusion coefficient at the reference time (28 days) and time exponent (*m*) are recommended as Eqs. (6) and (7), respectively (Thomas and Bentz 2002).

$$D_{ref} = 1 \times 10^{12.06 + 2.40 w/c} \qquad (6)$$

$$m = 0.2 + 0.4(FA/50 + SG/70) \leq 0.6 \qquad (7)$$

where *FA* and *SG* mean replacement ratios of fly ash and slag to binder.

Recently the diffusion coefficient from Eq. (6) is reported to overestimate actual diffusion coefficient since Eq. (6) was recommended for durability design with high diffusion coefficient (Kwon *et al.* 2009, Lee and Kwon 2012). In particular, concrete with lower than w/c of 0.5 contains much lower diffusion coefficient than the result from Eq. (6), so that an experimental parameter is assumed as 0.5 in order to consider an actual diffusion in concrete. The time dependent exponent of *m* is calculated as 0.4 from Eq. (7). After 18 years, the diffusion coefficient is evaluated to be 0.364×10^{-12} m²/sec and the comparison with the field data is presented in Fig. 6.

As shown in Fig. 6, field test results show enlarged chloride ingress to inside due to increased diffusion from deteriorated surface. Utilizing Eq. (5(a)), simulation of

(a) e = 5.0 mm

(b) e = 7.5 mm
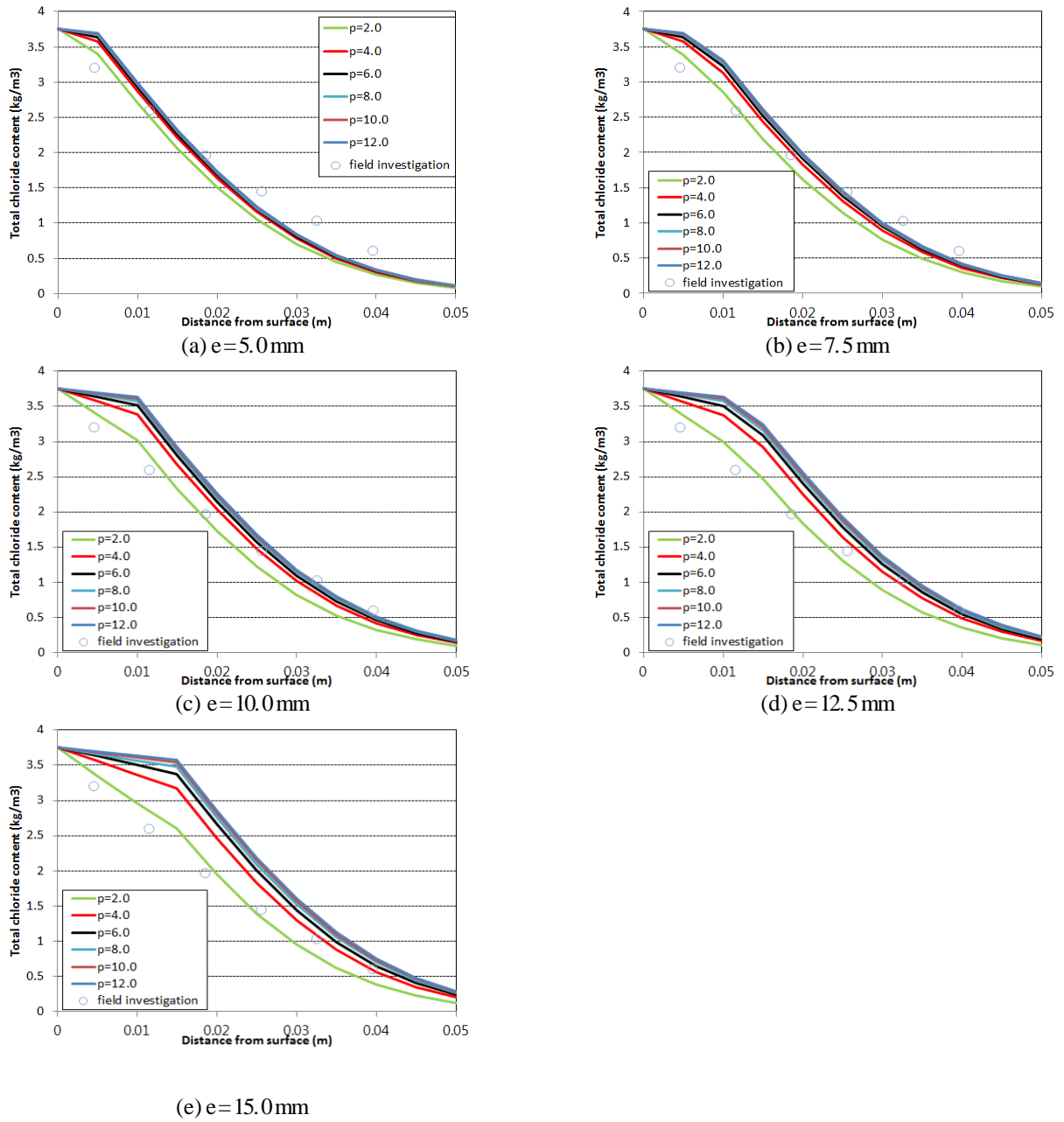
(c) e = 10.0 mm

(d) e = 12.5 mm

(e) e = 15.0 mm

Fig. 7 Simulation of chloride penetration considering deteriorated depth and enlarged diffusion

chloride behavior is performed considering both double-layered concrete and time-dependent diffusion. Here diffusion coefficient in deteriorated surface and its depth are unknown. For reverse analysis, deteriorated depth and enlarged diffusion coefficient are assumed to have 5~15 mm and 1.5~4.0 times of base concrete, respectively as in Table 4.

Relative errors between the simulation and the average of field investigation at the given depth are analyzed. The simulations varying D_1/D_2 and e are shown in Fig. 7. The variations of the relative errors with their contour are presented in Fig. 8. Relative error (R) is calculated as per Eq. (8).

$$R(\%) = \sqrt{(C_{test} - C_{sim})^2} / C_{test} \times 100 \qquad (8)$$

where C_{test} and C_{sim} are fields data and simulation result at the measured depth respectively.

Table 4 Deteriorated depth and enlarged diffusion coefficient for reverse analysis

Deteriorated depth (e) mm	Diffusion ratio, p(D_1/D_2)
5.0, 7.5, 10.0, 12.5, 15.0	2.0, 4.0, 6.0, 8.0, 10.0, 12.0

<div style="text-align:center">(a)　　　　　　　　　　　　　　　　　　　　　　　　(b)</div>
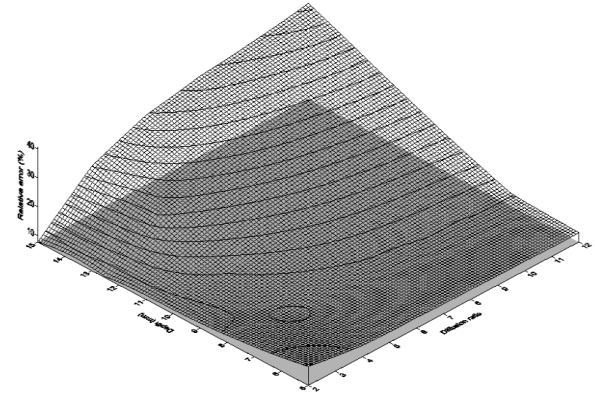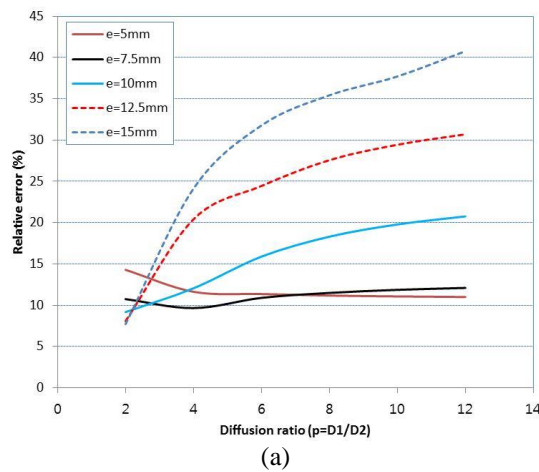
Fig. 8 Variation of relative error and contour: (a) Variation of relative error and (b) Relative error contour
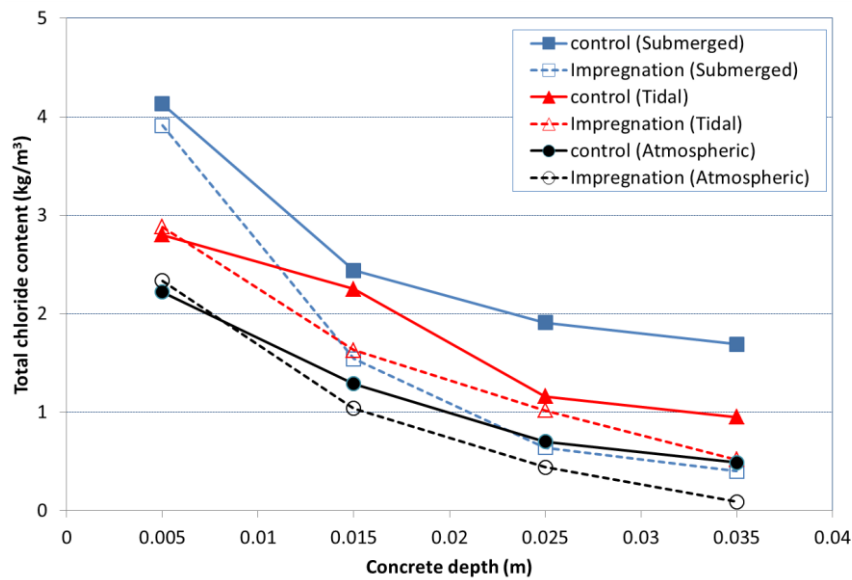

Fig. 9 Comparison of chloride content between surface-impregnated and normal concrete

From the analysis, relative errors show below 10.0% when deteriorated depth and enlarged ratio of diffusion coefficient reach 12.5~15.0 mm and 2.0 times of increment, respectively.

4.2 Simulation of chloride penetration for concrete with enhanced surface

Similar to the process in section 4.1, diffusion coefficient in concrete with Table 2 is calculated to be 18.60×10^{-12} m^2/sec, and it decreases to 3.107×10^{-12} m^2/sec considering exposure period (2 years) and experimental constant (0.5) for actual situation. The enhanced surface impedes the intrusion of chlorides so that the chloride diffusion is reduced within the impregnated depth. The diffusion after depth is the same as the base concrete, so that chlorides move inside rapidly. Actually the different pore structure can cause locally decreasing pressure which can make local condensation of chloride ion, however the local behavior of chlorides is disregarded for the simulation.

The chloride profiles between surface-impregnated and normal concrete show clear reduction of chloride penetration as shown in Fig. 9. The chloride behavior in surface impregnated concrete shows much lower chloride content inside even if it has almost same surface chloride contents. The surface chloride content as listed in Table 3 is decreasing from submerged to atmospheric condition due to exterior chloride concentration and relatively low exposure period of 2 years. In Table 5, enhanced depth and reduced diffusion coefficient are assumed for reverse analysis and the relative errors are calculated through Eq. (8).

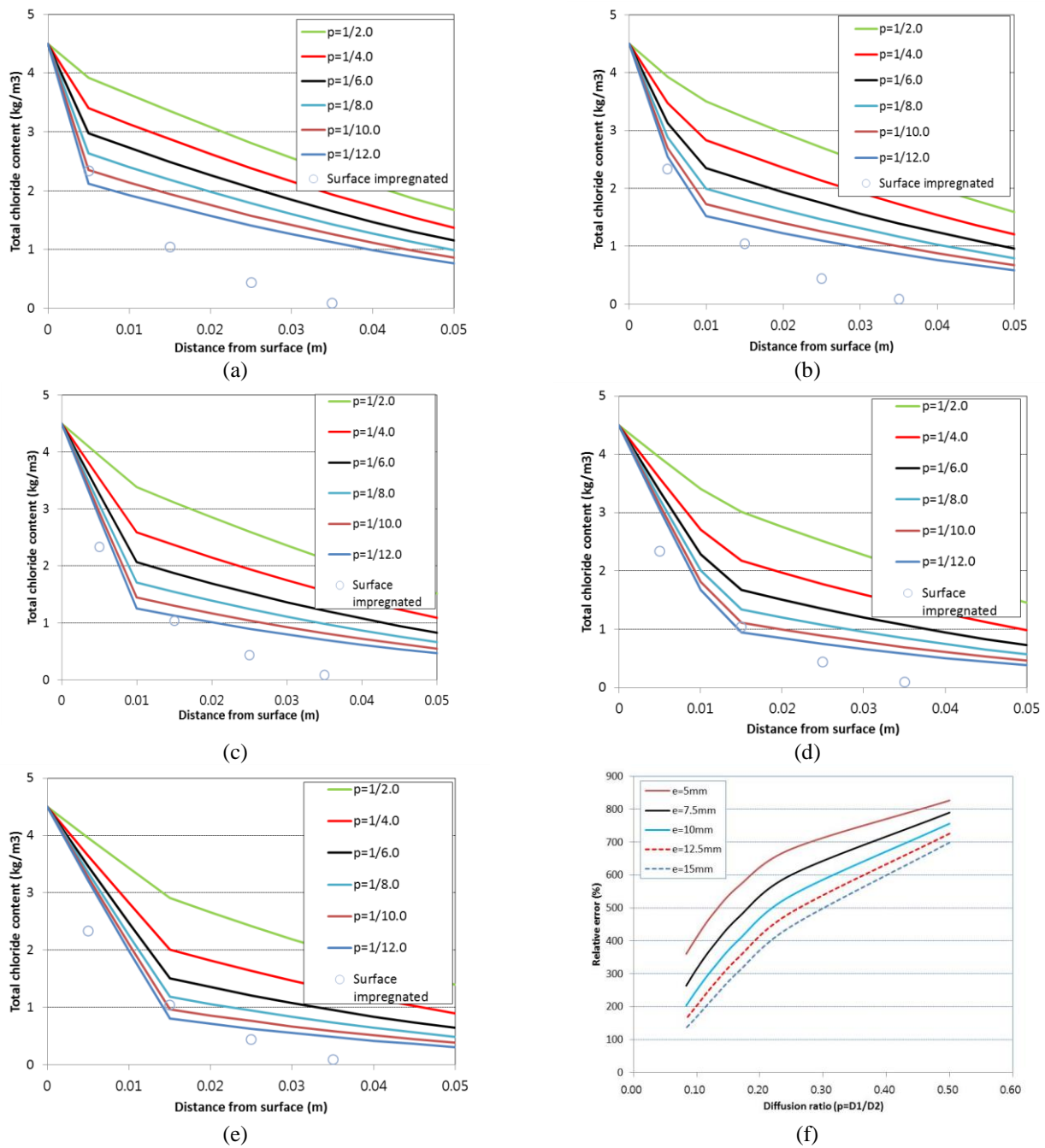The simulation results are shown in Fig. 10 to Fig. 12 with the evaluated relative errors.

Fig. 10 Simulation results with enhanced depth and reduced diffusion – Atmospheric condition: (a) e=5 mm, (b) e=7.5 mm, (c) e=10.0 mm, (d) e=12.5 mm, (f) e=15.0 mm and (g) Varying relative errors

Table 5 Enhanced depth and reduced diffusion coefficient for reverse analysis

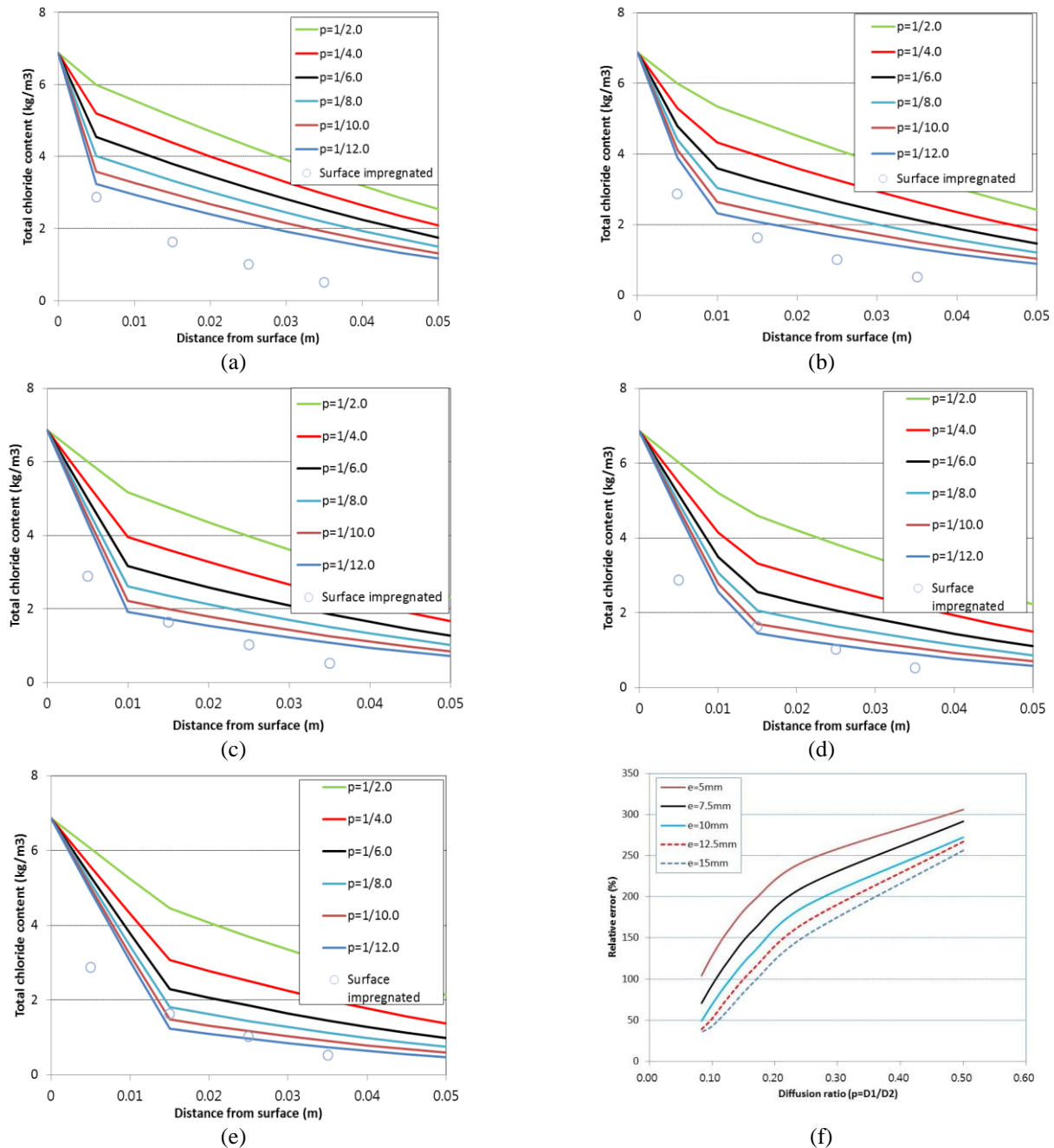| Enhanced depth (e) mm	Diffusion ratio, $p(D_1/D_2)$
5.0, 7.5, 10.0, 12.5, 15.0	1/2.0, 1/4.0, 1/6.5, 1/8.0, 1/10.0, 1/12.0

Fig. 11 Simulation results with enhanced depth and reduced diffusion – Tidal condition: (a) e=5 mm, (b) e=7.5 mm, (c) e=10.0 mm, (d) e=12.5 mm, (f) e=15.0 mm and (g) Varying relative errors

Table 6 Effective enhanced depth and diffusion reduction

Exposure	Enhanced depth - reduction ratio
Atmospheric	10 mm - 92%, 12.5 mm - 88%, 15.0 mm - 87%
Tidal	5 mm - 83%, 7.5 mm - 77%, 10.0 mm - 72%, 12.5 mm- 66%, 15.0 mm - 63%
Submerged	7.5 mm - 92%, 10.0 mm - 88%, 12.5 mm - 86%, 15.0 mm - 84%

For considering the changing of relative errors below 200%, the effective enhanced depth and diffusion reduction can be summarized as Table 6.

Actually the impregnated depth is reported to be approximately 10.0 mm without additional intrusion pressure since capillary suction is the main mechanism of
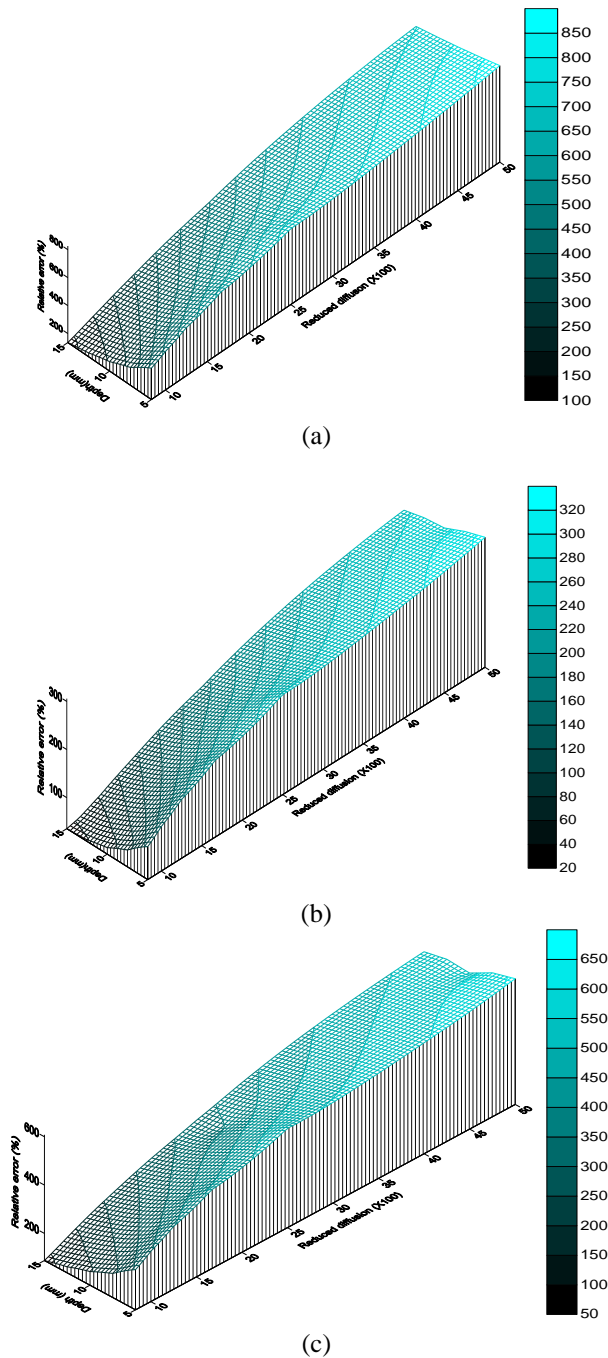
(a)

(b)

(c)

Fig. 13 Contours of relative error with varying exposure conditions: (a) Atmospheric conditions, (b) Tidal conditions and (c) Submerged condition

agent suction. In the atmospheric condition, enhanced depth shows 10.0~15.0 mm with high reduction of diffusion over 87%. In tidal and submerged conditions, 10.0~12.5 mm and around 85% reduction ratio are evaluated for minimum errors. Through the employment of time effect on diffusion and double layer concept, the chloride behavior from the analysis is much closer to the test results. The contours of relative error are shown in Fig. 13 with varying enhanced depth, diffusion reduction, and exposure conditions.

5. Conclusions

The conclusions on numerical technique for chloride ingress with cover concrete property and time effect are as follows.

- Considering double-layered concrete and time-dependent diffusion concept, analysis technique on chloride behavior is proposed considering enhanced and deteriorated surface of concrete. The technique is evaluated to be effective for deteriorated surface exposed deicing agent and enhanced surface due to surface impregnation. The deteriorated depth and enlarged diffusion coefficient are evaluated to be 12.5~15.0 mm and increment by 2 times, respectively. The deterioration of surface concrete and enlarged diffusion cannot be considered in classical Fick's 2nd Law equation but its accuracy is improved through adopting time effect on diffusion and multi-layered system.
- Through comparison with the test results in concrete with surface impregnation, effectively enhanced depth and diffusion reduction are evaluated. The result in the atmospheric condition shows small enhanced depth and high reduction of diffusion due to relatively short exposure period and low surface chloride content. However 10.0~12.5 mm of enhanced depth and 85% reduction of diffusion are clearly evaluated in tidal and submerged conditions.
- The proposed technique assumes steady-state condition of chloride penetration based on Fick's 2nd Law, so that complicated chloride behaviors cannot be considered such as chloride condensation, locally varying exterior conditions, and variation of free chloride ion. But the concrete exposed to steady state condition like submerged sea water or long time exposure condition, the proposed technique can be effectively applied with handling the surface conditions of concrete cover.

Acknowledgements

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