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Time and Crack Effect on Chloride Diffusion for Concrete with Fly Ash

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Abstract: Nuclear power plants are constructed very close to the marine environments for cooling water and the structures are more susceptible to chloride induced corrosion. Cracking in RC structures in mass concrete is unavoidable when they are exposed to chloride contaminated chemical environments. This study is focused on the evaluation of crack and time effect on chloride diffusion rate. Two types of concrete strength grade were taken for nuclear power plant construction and the crack was induced with varying from 0.05 to 1.35 mm of width. The tests for chloride diffusion coefficients from steady-state condition were performed. The influence of crack width on the chloride transmission behavior was discussed and analyzed over an exposure period to one year. The diffusion coefficients due to growing crack width increase in crack width but they decrease with increasing curing period, which yields 57.8–61.6% reduction at the age of 180 days and 21.5–26.6% of reduction at 365 days. Through the parameters of age and crack width which are obtained from regression analysis, the evaluation technique which can consider the effect of crack and time on diffusion is proposed for nuclear power plant concrete.

Keywords: nuclear power plant concrete, crack, chloride diffusion, steady-state condition, time effect.

1. Introduction

Concrete is an attractive construction material which has many engineering merits like cost–benefit, stable supply system, fire resistance, and high durability despite of considerable CO_2 emission during manufacturing of cement (Back et al. 2011). Concrete has an excellent radiation shielding performance and low thermal conductivity, so that it has been used for nuclear power plant construction (Lee et al. 2016a, b; ACI Committee 1996, 2009). The prestressed concrete (PSC) or reinforced concrete (RC) members are the commonly used in the nuclear industry, however these structures are affected by reinforcement corrosion when exposed to coastal environment (Broomfield 1997; Song et al. 2006). Nuclear power plants are usually located near to the sea due to the cooling water-intake and drainages. Even if the structures are not submerged or directly exposed to sea water, the chlorides intrude into concrete through diffusion and permeation, and it causes rapid steel corrosion (Broomfield 1997; Song et al. 2006; Win et al. 2004; Nguyen et al. 2017). Nuclear power plant system has a large dimension with massive system and concrete is placed at a large scale, so called, mass concrete. In the mass concrete, cracks easily occur due to the dimensional instability caused by drying shrinkage and hydration heat at early age (Lee et al. 2016a, b; Song et al. 2001; Kwon et al. 2009; Yang et al. 2016). The crack on the surface of the concrete allows more rapid intrusion of chlorides into the concrete and accelerates the initiation of corrosion. Many researches have performed the quantification of chloride intrusion in cracked concrete considering crack width (Yokozeki et al. 1998; Gerard and Marchand 2000), torturity of crack shape (Yang et al. 2017a, b), and the complicated models enhanced by the material behaviors in early aged concrete such as porosity and saturation have been proposed (Maekawa et al. 2003; Park et al. 2012a, b; Ishida et al. 2009).

The diffusion and permeation of chlorides decrease with time. The main reasons for reduction of chloride ingress can be summarized reduction of porosity which can block the chloride ion intrusion (Kwon et al. 2009; Thomas and Bamforth 1999), reduction of diffusion which makes the ion transport slow into the concrete (Yoon et al. 2017; Park et al. 2009), enhanced hydration which can make more absorption of chloride ions (Ishida et al. 2009; Leng et al. 2000), and decreasing gap of chloride ion concentration between the inner and outer boundary conditions which reduces the concentration gradient (Welty et al. 1989). The time-dependent diffusion has been studied for a long time with considering changing properties of concrete but they are focused only on crack-free concrete (Thomas and Bamforth

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1999; Lingjie et al. 2017). Crack width also changes with time due to the steady reaction of cement hydration and auto-healing enhanced by relative humidity (Japan Society of Civil Engineering 2002).

The governing mechanism for chloride behavior can be formulated with Nernst–Einstein Equation and Fick's 2nd Law, representatively. In the former formulation, the reduction in porosity and moisture transport are considered in flux term which handles diffusion and convention. The enhanced absorption of free chloride ions due to cement or supplementary cementitious material hydration is considered in ion-mass conservation system (Maekawa et al. 2003; Ishida et al. 2009). Unlike the equation, Fick's 2nd Law assumes steady-state condition which means constant inlet of chlorides from the surface chloride condition. In the equation, time-exponent is proposed for expressing the decreasing diffusion with time, which contains a referential period and diffusion coefficient, and effect of mineral admixtures like Fly Ash (FA), Ground Granulated Blast Furnace Slag (GGBFS), and Silica Fume (SF) (Thomas and Bamforth 1999; Lingjie et al. 2017; Thomas and Bentz 2002).

In the present work, two concrete mix proportions for nuclear power plant are prepared and the accelerated chloride diffusion tests under steady-state condition are performed for concrete containing various crack width and curing periods up to 1 year. The results from the accelerated diffusion test are quantitatively analyzed with different crack width and curing period.

2. Outline of Test for Chloride Diffusion with Cracks

2.1 Mixing Proportions and Used Materials

Two different mix proportions with 4000 psi (N: 28 MPa) and 6000 psi (H: 41 MPa) are prepared and shown in Table 1. Tables 2 and 3 show the properties of Ordinary Portland Cement (OPC) and FA, and the aggregates used for the investigation, respectively.

2.2 Test Procedure

2.2.1 Compressive Strength

The concrete samples with 100 mm diameter and 200 mm height were cast and cured for 56, 180, and 365 days. The standard compressive strength at the age of 28 days was planned but there was a delay in attaining the strength, so that strength at the age of 56 days was evaluated. The compressive strength test was performed referred to KS F 2406.

2.2.2 Accelerated Diffusion Coefficient Under Steady-State Condition in Cracked Concrete

(1) Crack inducing

Before the accelerated diffusion test, crack was induced in the concrete disk samples with UTM. The standard samples with 200 mm height was cut to 100 mm and subjected to splitting test. The designed crack width was difficult to obtain through loading level, so that many cracking tests were carried out and the crack

Table 1 Mix proportions for N (4000 psi) and H (6000 psi) grades.

Grade (psi)	G_{max} (mm)	W/B	S/a	W	Binder		Coarse agg.	Fine agg.
					C	FA		
6000 psi-H grade	19	40	44.4	162.75	325.50	81.38	938.77	748.89
4000 psi-N grade	19	50	46.7	162.75	260.64	64.86	938.77	822.01

W/B water to binder ratio, *S/a* sand to aggregate, *W* water, *C* cement, *FA* fly ash, *Agg.* aggregate.

Table 2 Physical and chemical properties of the cement and FA used.

Items types	Chemical composition (%)							Physical properties	
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Ig. loss	Specific gravity (g/cm^3)	Blaine (cm^2/g)
OPC	21.96	5.27	3.44	63.41	2.13	1.96	0.79	3.16	3214
FA	55.66	27.76	7.04	2.70	1.14	0.49	4.3	2.19	3621

Table 3 Physical properties of the aggregates used.

Items types	G_{max} (mm)	Specific gravity (g/cm^3)	Absorption (%)	F.M.
Fine aggregate	–	2.58	1.01	2.90
Coarse aggregate	25	2.64	0.82	6.87

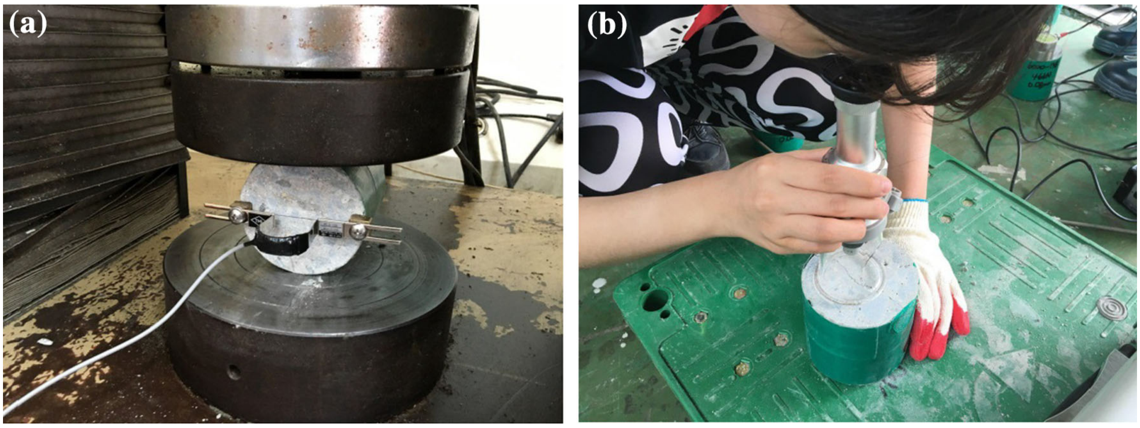

Fig. 1 Photographs for crack inducing and crack measurement. **a** Crack inducing and gauge setup. **b** Crack width measurement.

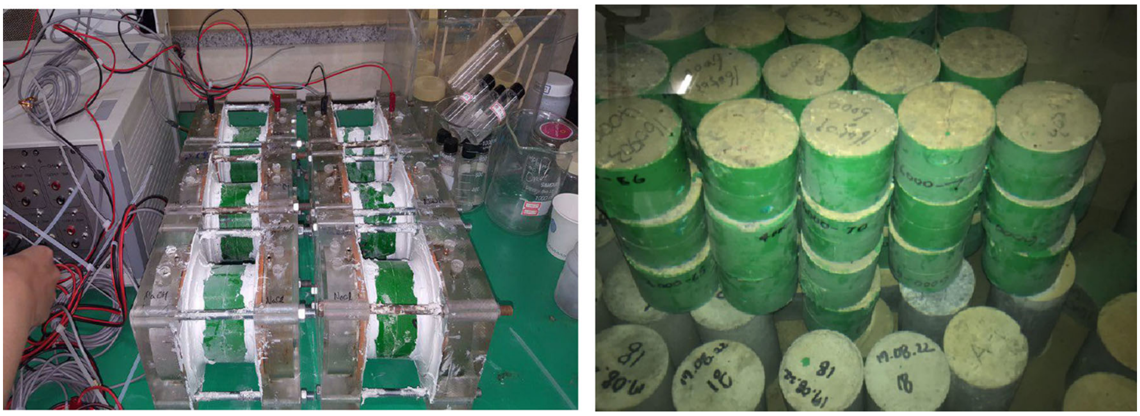

Fig. 2 Photographs of accelerated diffusion test setup and concrete samples under curing.

widths were grouped with 0.1 mm interval. The crack gauge was attached on the concrete surface perpendicular to cracking direction (loading direction), and the crack width was monitored with loading. The crack width increases with loading and slightly decreases after unloading. The crack width after fully-unloading was measured and the side was coated with epoxy resin except the top and bottom side for one-dimensional flow of chloride transport. The photographs of crack-inducing and width measurement are shown in Fig. 1.

(2) Accelerated diffusion test under steady state condition
Diffusion coefficient from non-steady state condition is

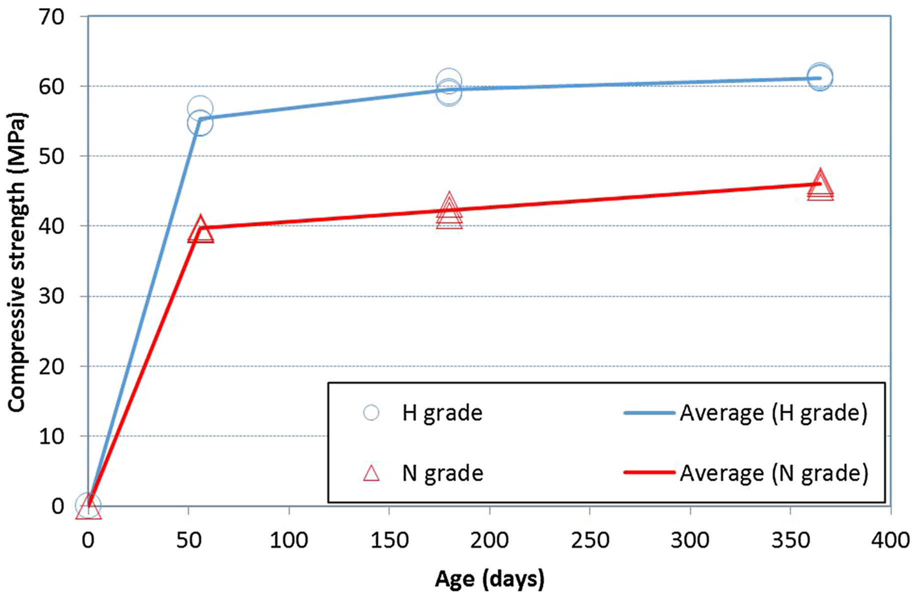

Fig. 3 Changes in compressive strength with different curing ages.

very vulnerable to crack width since it reflects the local chloride penetration depth in the crack width, so that the enlarged diffusion coefficient reaches to 160 times for normal condition (Park et al. 2012a, b). In the work, the increasing diffusion coefficient from the total cracked concrete is to be monitored. Accelerated diffusion test in concrete with single crack was performed referred to ASTM 1202, under the steady-state condition. Diffusion coefficient was calculated through the results adopted by Andrade (1993). For the test, a 0.5 M solution of NaCl and 0.1 M solution of NaOH was used as the catholyte and anolyte in the migration cell, respectively (Andrade 1993). The diffusion coefficient was calculated using the following Eq. (1).

$$D_{eff} = \frac{RTit_{cl}l}{nF^2\Delta EAC_{cl}Z} \tag{1}$$

where R is the universal gas constant (8.314 J/mol), T is the absolute temperature (K), i, is the current (Amp), t_{cl} is the transference number (0.4336 in the case of 0.1 M NaOH and 0.5 M NaCl), l, is the thickness of the sample (m), n is 1.0, F is Faraday's constant (9.648×10^4 J/V mol), ΔE is the applied potential (30 V), A is the cross-section of the concrete sample (m^2), Z is the ion valence, and C_{cl} is the concentration of the chloride ions in the diffusion cell (mol/L). Figure 2, shows the photograph of diffusion test setup and concrete cylinders for chloride diffusion test under curing. The measured current rapidly increases with test initiation and keep stable after 1–2 days. The stable measurement is adopted and considered for diffusion coefficient calculation.

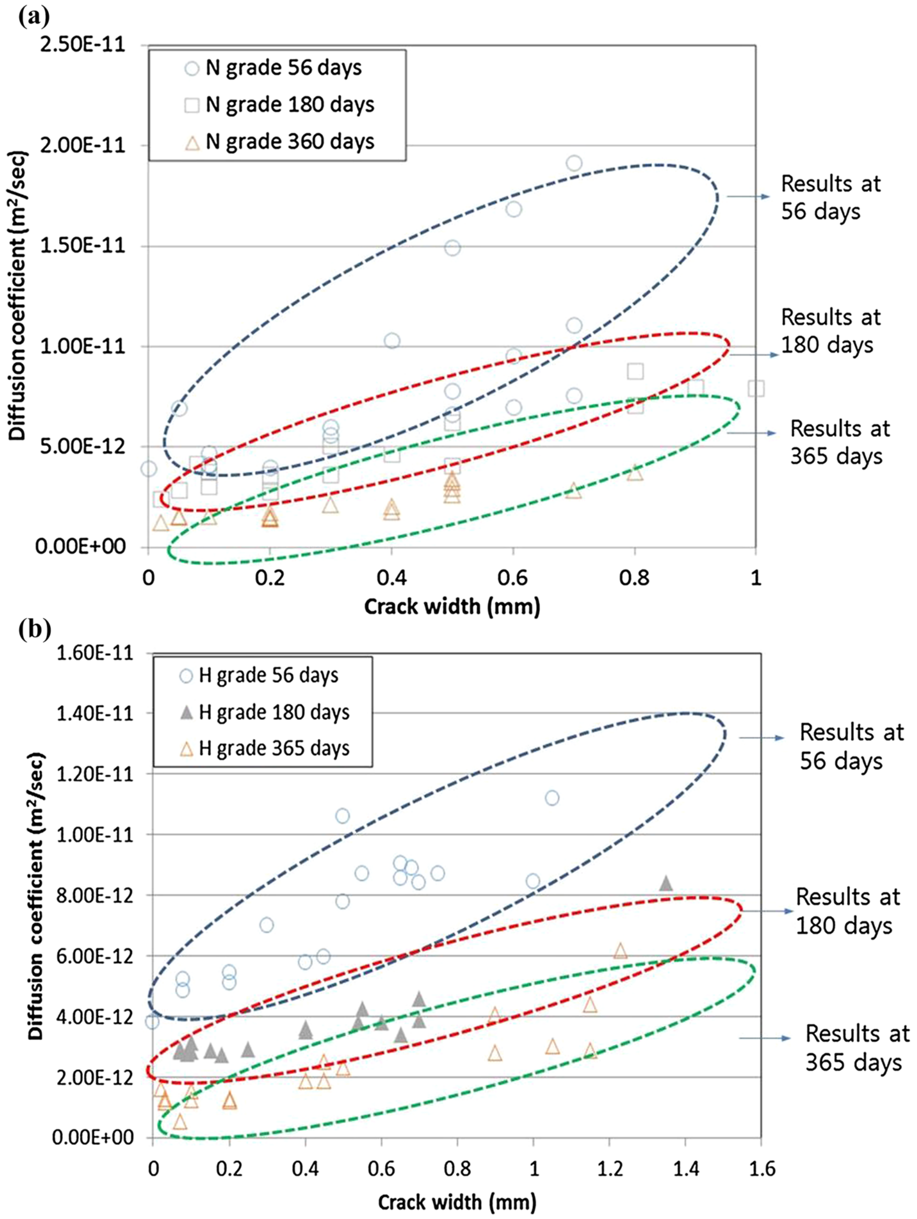

Fig. 4 Changes in diffusion coefficient under steady state condition with various crack width. **a** Enlarged diffusion coefficient in cracked concrete (N grade). **b** Enlarged diffusion coefficient in cracked concrete (H grade).

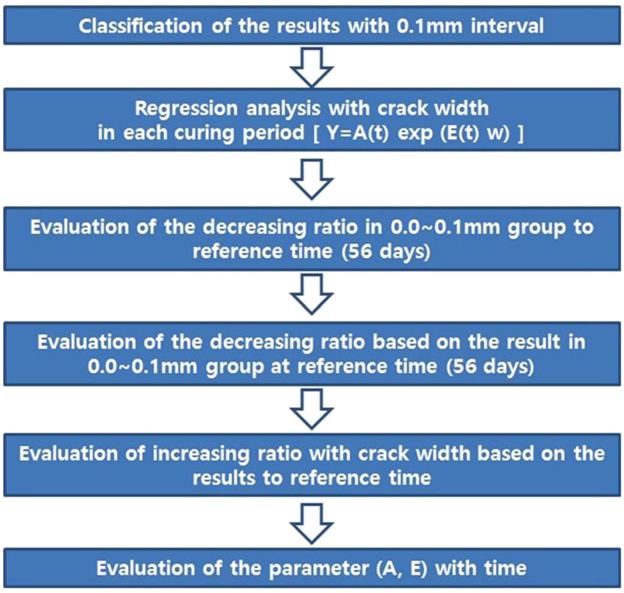

Fig. 5 Flowchart for quantification of crack and age effect on diffusion.

3. Evaluation of Time and Crack Effect on Diffusion

3.1 Changes in Compressive Strength with Ages

The compressive strength test results up to 1 year are shown in Fig. 3. Triplicate samples were tested and the average results were plotted. The N grade concrete has the compressive strength value of 39.7, 42.3, and 46.1 MPa after 56, 180, and 365 days, respectively. Those in the H grade have the compressive strength 56.8, 59.2, and 61.2 MPa at each curing period.

3.2 Changes in Diffusion Coefficient with Various Ages and Crack Width

The diffusion coefficients with various crack widths and curing periods are plotted in Fig. 4a, b considering strength level. From the figures, it is evaluated that N and H grade concrete show significantly increasing diffusion coefficients with growing crack width. The diffusion coefficient in sound concrete was found to be 3.93×10^{-12} m^2/s, and it increased to 1.03×10^{-11} m^2/s and 1.91×10^{-11} m^2/s when the crack width was 0.4 and 0.7 mm at the age of 56 days respectively. The above-mentioned chloride diffusion coefficients at 0.4 and 0.7 mm are 2.62 and 4.86 times greater than the control cases. By considering the curing period, the diffusion coefficients decreased to 2.43×10^{-12} m^2/s at 0.02 mm of crack width, 4.64×10^{-12} m^2/s at 0.4 mm of crack width, and 7.93×10^{-12} m^2/s at 1.0 mm crack width at the age of 180 days of curing periods respectively. When the curing period is extended to 365 days, the diffusion coefficient gradually decreased to 1.27×10^{-12} m^2/s in sound concrete, 2.05×10^{-12} m^2/s at 0.4 mm of crack width, and 3.78×10^{-12} m^2/s at 0.8 mm of crack width respectively. Even if the curing period is extended, the significantly enlarged diffusion coefficient is observed with growing crack width due to rapid chloride penetration in the crack width.

In the H grade at the age of 56 days, the diffusion coefficient in the sound concrete is observed to be 3.84×10^{-12} m^2/s and it gradually increases with crack width. Between 0.4 and 0.5 mm and at 1.0 mm of crack width crack width, the chloride diffusion coefficient increased to 2.7 and threefold increase was observed at the age of 56 days. The increasing trend with curing period is similar as that in N-grade concrete. At the age of 180 days, 1.3 and 2.9 times increase in diffusion coefficients were

Table 4 Normalized crack width and diffusion coefficient.

Averaged crack width	Increasing ratios- H grade			Increasing ratios- N grade		
	56 days	180 days	365 days	56 days	180 days	365 days
0.05	1.000	0.616	0.215	1.000	0.579	0.267
0.15	–	0.625	0.260	0.792	0.626	0.291
0.25	1.141	0.630	0.231	0.736	0.599	0.283
0.35	1.514	–	–	1.070	0.800	0.399
0.45	1.263	0.773	0.388	1.897	0.854	0.352
0.55	1.946	0.873	0.432	1.801	0.945	0.566
0.65	1.906	0.778	–	2.049	–	–
0.75	1.846	0.915	–	2.316	–	0.524
0.85	–	–	–	–	–	0.695
0.95	–	–	0.633	–	1.464	–
1.05	2.120	–	0.561	–	1.459	–
1.15	–	–	0.670	–	–	–
1.25	–	–	1.138	–	1.522	–
1.35	–	2.946	–	–	–	–

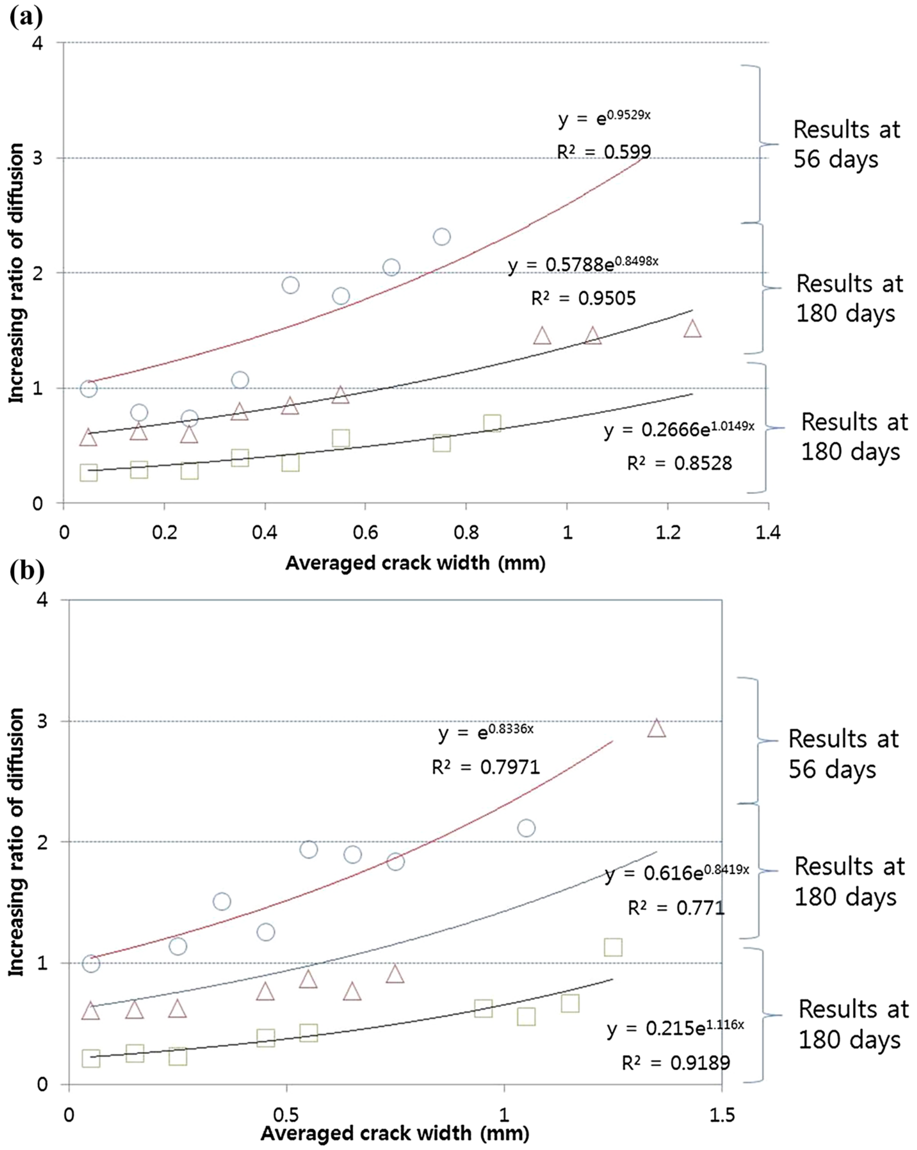

Fig. 6 Diffusion normalization with time and crack width. **a** Result normalization of N grade. **b** Result normalization of H grade.

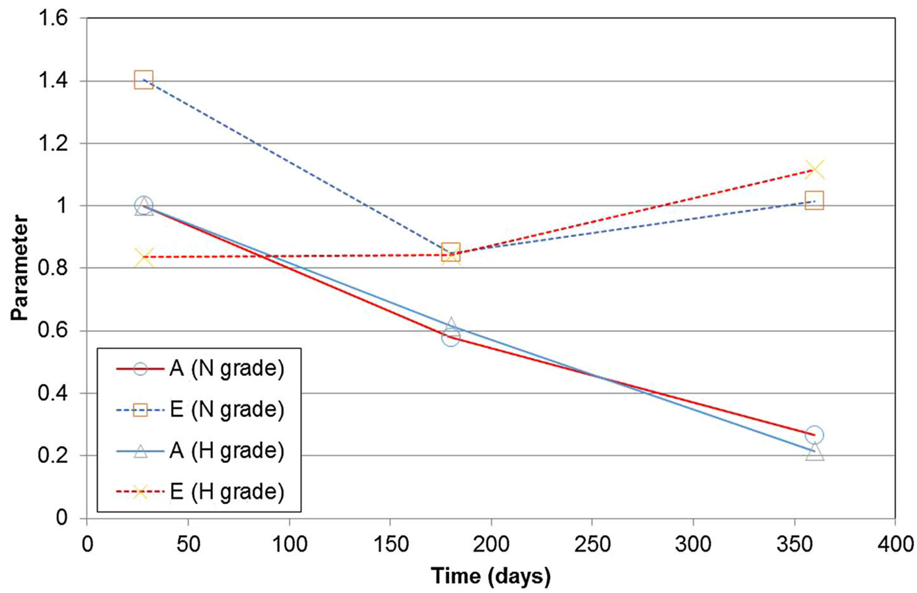

Fig. 7 Regression analysis on parameters (A and E).

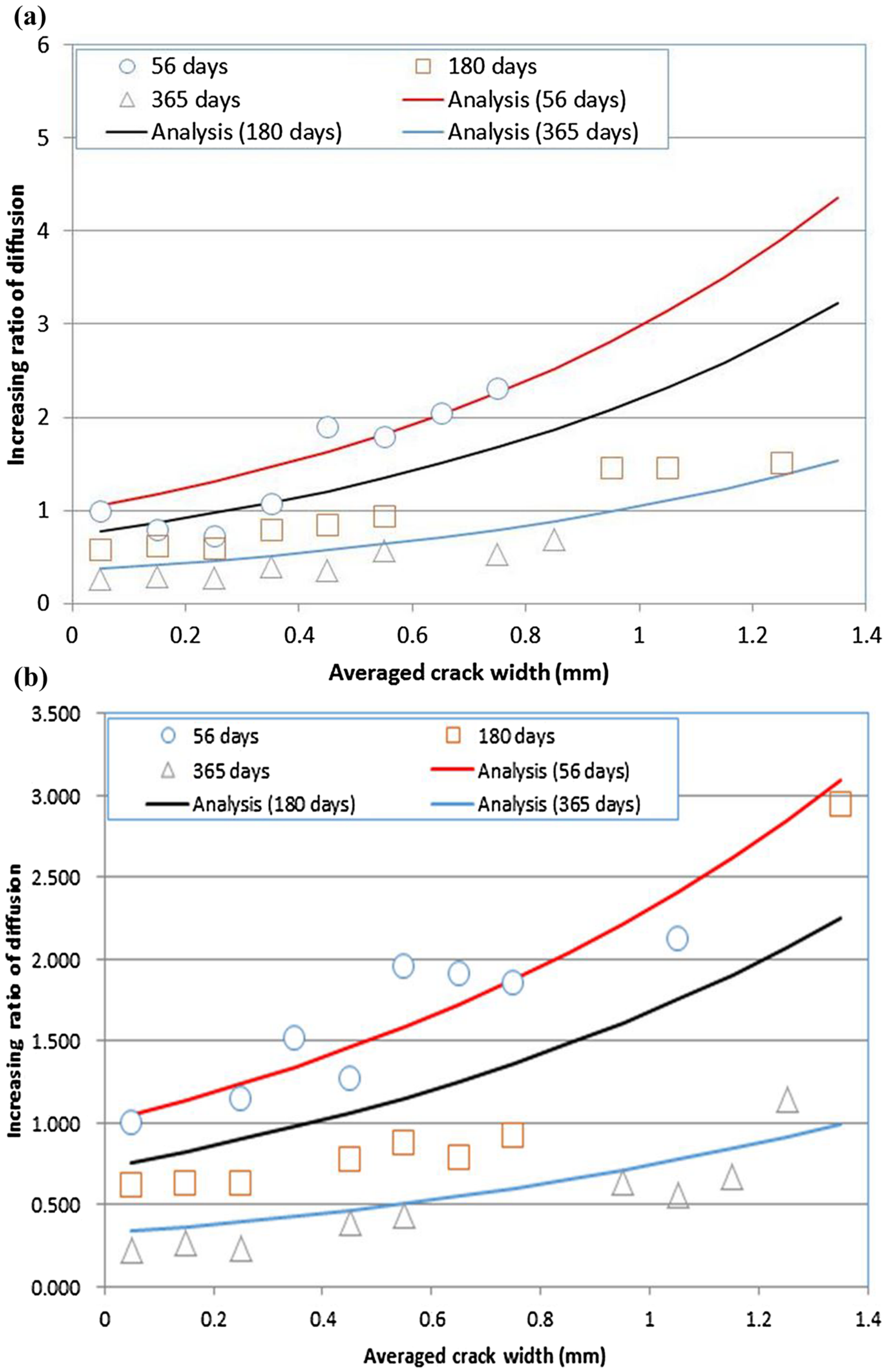

Fig. 8 Diffusion coefficient of test and regression analysis. **a** N-grade. **b** H-grade.

observed with increasing crack width to 0.4 and 1.0 mm. At the age of 365 days, diffusion coefficient is found to be 2.7 times higher than the sound concrete when the crack width reaches over 1.0 mm.

Earlier studies indicated that, the chloride diffusion coefficient was significantly increased to 150–160 times when the crack width reached to 0.4 mm under the non-steady state condition. The results obtained from the non-steady state conditions are not the diffusion over the entire cracked area since the chloride penetration depth is confined in the cracked section only (Park et al. 2012a, b). In the previous test under steady-state condition, 2.7–3.0 times increase in diffusion coefficient was observed, which is consistent with the present results in the concrete with 0.4 mm of crack width.

3.3 Quantification of the Effect of Crack Width and Age on Diffusion

In this section, quantification of crack and age effect on diffusion is performed. During the exposure period, the diffusion is reported to be non-linearly increasing with crack width (Kwon et al. 2009; Yang et al. 2017a, b), so that the increasing ratios are analyzed with crack width in each curing period. Firstly the regression analysis with age is

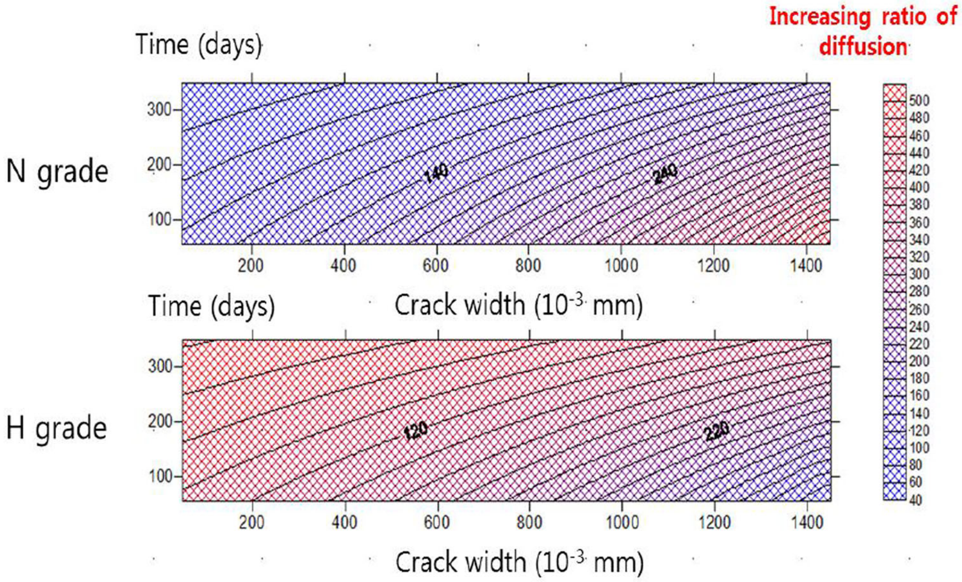

Fig. 9 Diffusion coefficient contours with time and age effect.

performed for the diffusion coefficient in sound concrete. The regression analysis can be drawn as Eq. (2), where $A(t)$ and E show the parameters for age and crack effect on diffusion, respectively. The flowchart for quantification of diffusion behavior is shown in Fig. 5.

$$D(w,t) = D_0 A(t)\exp(Ew) \tag{2}$$

where $D(w,t)$ and D_0 are diffusion in cracked concrete with increasing period and sound concrete at the reference period.

The results of normalization in sound concrete for N and H grade are listed in Table 4 and plotted in Fig. 6, respectively. In the normalization of the results, the data over relative error of 100% is disregarded and calculated again.

From Fig. 6a for N grade, it is found that the gradient of $A(t)$ is reduced from 1.000 at the reference period to 0.579 (at 180 days) and 0.267 (at 365 days) which shows a greater reduction of 26.7% when it reaches to 365 days. The parameter E has relatively small reduction from 0.9529 to 0.8498 (180 days) and 1.0149 (365 days). Figure 6b showing the results of H grade, it is found that $A(t)$ also decreases from 1.000 to 0.616 (180 days), and 0.215 (326 days). Unlike a great reduction of parameter- $A(t)$ from N and H grade, crack parameter-E slightly increases from 0.8366 (reference period: 56 days), 0.8419 (180 days), and 1.1116 (365 days). This indicates that the increase in crack width is not much affected by the curing period. In the previous test results of 3 days and 28 days (Park 2002), the increasing ratio of diffusion was strongly affected by crack width regardless of curing periods even in early aged condition. In order to evaluate the effect of time on diffusion, $A(t)$ and E are plotted with curing period.

In Fig. 7, the effect of age on parameter of $A(t)$ and E are shown, which shows a close linear relation of $A(t)$ with time but relatively inconsistent relation of E regardless of mix proportions. When adopting the results from the regression analysis, $A(t)$ and E can be formulated with curing period. $A(t)_N$ and $A(t)_H$ denote the time effect on diffusion coefficient in N and H-grade concrete, respectively. The parameter E shows no clear relation with ages and has a level of 0.8366–1.4036, so that it is assumed as the average of each grade. E for N-grade and H-grade are evaluated to be 1.09 with 26.1% of coefficient of variation (COV) and 0.93 with 17.1% of COV, respectively. The effect of age can be written as Eqs. (3a) and (3b), which yields final equation like Eqs. (4a) and (4b) with merging Eq. (2).

$$A(t)_N = -0.0021T + 1,\ R^2 = 0.9786 \tag{3a}$$

$$A(t)_H = -0.0022T + 1,\ R^2 = 0.9879 \tag{3b}$$

$$D(w,t)_N/D_0 = (-0.0021T + 1) \times [EXP(1.09w)] \tag{4a}$$

$$D(w,t)_H/D_0 = (-0.0022T + 1) \times [EXP(0.93w)] \tag{4b}$$

The comparison of the test results is plotted in Fig. 8 where Fig. 8a, b shows the comparison of N and H grade, respectively. The diffusion coefficient contours covering age and crack width for N and H grade are shown in Fig. 9.

4. Conclusions

In this work, the effects of crack and age on diffusion coefficient in concrete for nuclear power plant are evaluated and simplified evaluation technique is proposed considering the effects of age and crack width. The conclusions from the study are as follows.

1. At the age of 56 days, the N grade sound concrete without crack has the diffusion coefficient of 3.93×10^{-12} m^2/s and it increases to 1.03×10^{-11} m^2/s and 1.91×10^{-11} m^2/s when the crack width reaches to 0.4 mm and 0.7 mm, respectively. With increasing curing age, the diffusion coefficient decreases to 4.64×10^{-12} m^2/s (0.4 mm of crack width) and 7.93×10^{-12} m^2/s (1.0 mm crack

width) at 180 days of curing period. In the case of 365 days of curing period, they gradually decrease to 2.05×10^{-12} m^2/s (0.4 mm of crack width) and 3.78×10^{-12} m^2/s (0.8 mm of crack width) respectively.

2. In H grade concrete (6000 psi), the diffusion coefficient of sound concrete (at the age of 56 days) is found to be 3.84×10^{-12} m^2/s and it gradually increases with crack width. Further the diffusion coefficient increases to 2.7 and 3.0 times between 0.4 and 0.5 mm and 1.0 mm of crack width, respectively.
3. At the end of 1 year, the effect of age on diffusion can be independent on crack effect since the increasing diffusion due to crack width is not much affected regardless of strength grade and curing period. Even if the diffusion coefficients significantly increase due to growing crack width, they decreases with extended curing period, The enhanced diffusion coefficients due to increase in crack width decrease with similar reduction level with extension of curing period, which yields 57.8–61.6% reduction at the age of 180 days and 21.5–26.6% of reduction at 365 days with similar trend, respectively. Considering the two parameters (age and crack width), simple evaluation technique is proposed for chloride diffusion coefficient in cracked concrete for nuclear power plant.
4. In the work, the results from 56 days are adopted to reference data. If reference diffusion coefficient is set to 28 days, the decreasing pattern is expected to be similar but the constant should be altered. With more data base of mix proportions, crack width, and curing period on diffusion coefficient, more reasonable evaluation technique can be derived considering interaction of crack and age on chloride diffusion.

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