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Model uncertainty in diffusion coefficient for chloride ingress into concrete

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Abstract

Diffusion coefficient for the chloride ingress into concrete is a key parameter describing the resistance against chloride penetration into the concrete. Consequently, it is an important aspect of the durability of reinforced concrete structures endangered by chloride-induced corrosion. The diffusion coefficient may be used for the direct assessment of the concrete quality with respect to chloride ingress and with respect to the numerical analysis of the durability of reinforced concrete structures. To estimate the diffusion coefficient, the computation based on the Second Fick's Law model and approximation of chloride profile from destructive penetration tests is usually utilised. Semi-destructive or non-destructive electrochemical tests provide estimates of the diffusion coefficient. The study reveals that (a) the resistivity readings provide a higher estimate for diffusion coefficient in comparison to short-time field exposures that seem to be associated with large uncertainty for exposure periods shorter than one year; (b) electrical resistivity measurements show significantly lower variation com-pared to the estimates based on chloride profiles.

Keywords: diffusion coefficient; chloride ingress; reinforced concrete structure; electro-chemical tests; calibration;

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1. Introduction

Diffusion coefficient for the chloride ingress into concrete is a key parameter describing the resistance against chloride penetration into the concrete. Further, it is a key parameter for estimating durability of reinforced concrete (RC) structures exposed to environments with significant concentrations of chlorides and endangered by chloride-induced corrosion. These include many types of structures as parts of road networks where de-icing salts are applied or structures located nearby seacoasts. Their maintenance and durability are an urgent topic for civil engineers around the globe. The diffusion coefficient may be used for the direct assessment of the concrete quality with respect to chlorides. It is an essential input for numerical modelling and predictions of durability (service life) of RC structures. In order to obtain the diffusion coefficient, the computation based on the Second Fick's Law model and approximation of chloride profile from destructive penetration tests (ASTM C1543, 1996; Nordtest NTBuild 443, 1995) is usually conducted. Semi-destructive (ASTM C1202, 2012) or non-destructive electrochemical based tests (AASHTO T358, 2013) allow for the indirect computation of the diffusion coefficient.

The assessment of reinforced concrete durability is related to application and evaluation of the diffusion coefficient (Chen et al., 2019; DuraCrete, 2000; Konecny and Lehner, 2017). The main concern is usually the quantification of uncertainty in the applied analytical or numerical model (Holický et al., 2016; JCSS, 2006) and uncertainty in testing procedures (Faber et al., 2006; Jcgm, 2008; Kessler and Gehlen, 2010). The reported values for the scatter of diffusion coefficient expressed by the coefficient of variation provide the range of values. The indicative values start from 0.05 (Faber et al., 2006; Faber and Sorensen, 2002; Schneider et al., 2015) or from a range 0.05-0.1 (Malioka, 2009) and reach a high value of 0.2 (Vu and Stewart, 2000).

Even though the analysis of the laboratory experiments of 32 HPC mixtures (with a reference level provided by OPC specimens) provided a range between 0.03 and 0.06 with some mixtures having a coefficient of variation up to 0.1 (Malioka, 2009), a value of 0.05 might be generally considered very low for in-situ cast concretes (see below). It is worth noticing that the study (Malioka, 2009) was based on the correlation of the long-term chloride penetration with electrical resistivity measurement as suggested by some authors even for special concrete mixtures such as binary and ternary mixtures (AASHTO T358, 2013; ASTM C1202, 2012).

However, general experience with in-situ cast concrete suggests that diffusion processes are affected by material properties that might be locally significantly variable, by cracks or by properties of concrete surface – therefore a large model uncertainty is expected. This is indirectly justified also by recent developments of numerical models based on e.g. the finite elements or cellular automaton methods, aiming to improve predictions of diffusion progress (Konecny and Lehner, 2017).

The current study is based on the preliminary evaluation of the diffusion coefficient. Its estimates are obtained by two distinctive approaches:

1. Analysis of chloride profiles,
2. Measurements of concrete electrical resistivity on the samples prepared in the laboratory.

The chloride profiles are obtained by a modified NT Build 443 procedure (Nordtest NTBuild 443, 1995) where the laboratory exposure is substituted by chloride exposure as measured at motorway I/11 near Ostrava, Czech Republic. The electrical resistivity (AASHTO T358, 2013) is evaluated in laboratory conditions on other samples without chloride exposure. Evaluated diffusion coefficients from the two methods are critically compared. Finally, the correlation between the long-term and laboratory measurements is investigated and recommendations for practical applications regarding a separate or combined use of the two methods are provided.

2. Methodology

The critical comparison of the two selected methods for chloride ingress-related diffusion coefficient is based on test results obtained by (a) chloride profiles from in situ exposure as codified in (ASTM C1543, 1996; Nordtest NTBuild 443, 1995), and (b) electrical resistance according to the AASHTO standard (AASHTO T358, 2013). In approach (a), the chloride penetration test allows studying the chloride concentration profiles from the laboratory or in situ experiments. The profiles of chloride concentrations are established based on the extraction of concrete powder containing chlorides. The concentration of chloride ions in the concrete dust is then determined by potentiometric titration (ČSN EN-14629, 2008). After determining chloride concentrations at given depths, the measured chloride profile is approximated via equation (2) in (Lu, 1997), with the diffusion coefficient estimated by the method of least squares (Birge, 1932; Kožar et al., 2019). The so-called second Fick's law reads:

$$C(x,t) = C_0\left\{1 - erf\left(\frac{x}{\sqrt{4 \cdot D_c \cdot t}}\right)\right\}, \tag{1}$$

where $C(x,t)$ [mass %] is the chloride ion concentration at a distance of x [m] from the surface of the concrete in time t [s], C_0 [mass %] is a surface concentration of chloride ions, erf is the error function; and D_c [m^2/s] is the effective diffusion coefficient, characterizing the ability of concrete to resist the penetration of chlorides. The procedure and the calculation process are described in detail in (Collepardi et al., 1972).

All available studies consider relationship (1) as unbiased but consider a wide range of coefficients of variation of uncertainty related to Fick's law as discussed above.

Regarding approach (b), the procedure for the diffusion coefficient estimation from the electrical resistivity readings is described in (Horňáková et al., 2020). It is based on the volumetric resistivity ρ_{BR} that is calculated from the measurements of electrical resistivity (AASHTO T358, 2013). For a porous material such as concrete, the diffusion coefficient is computed according to the Nernst-Einstein equation (Ghosh, 2011; Horňáková et al., 2020):

$$D = \frac{RT}{Z^2F^2} x \frac{t_i}{\gamma_i C_i \rho_{BR}}, \tag{2}$$

where D denotes the diffusion coefficient [m^2/s], $R = 8.314$ is the universal gas constant [J/K.mol], T absolute temperature [K], Z valence of ions [-], F Faradays constant [C/mol], t_i transport number of chloride ions [-], γ_i activity coefficient of chloride ions [-], C_i the concentration of chloride ions [mol/m^3], ρ_{BR} volumetric resistivity [Wm]. The coefficient of activity γ_i may be computed according to (Ghosh, 2011) yielding the value of 0.692 as applied in the presented article.

Average diffusion coefficient at concrete age t may be estimated from equation (3) according to (Thomas and Bamforth, 1999):

$$D_{c,nom,t} = D_{c,nom,28}\left(\frac{t_{28}}{t}\right)^m, \tag{3}$$

where $D_{c,nom,28}$ is a nominal diffusion coefficient in [m^2/s], measured at concrete age t [years]; t_{28} [years] is a reference period of measurement for an age of 28 days (0.767 y.); and m is the aging factor.

3. Experimental data

A concrete mixture was prepared; intentionally the mixture was designed to achieve low-strength concrete that is often found in old bridges. The concrete mixture contained 255 kg/m^3 of Cement type I 32.5, 170 kg/m^3 of water, 944 kg/m^3 of natural crushed aggregate (0/4), 391 kg/m^3 of natural crushed aggregate (4/8) and 553 kg/m^3 of natural crushed aggregate (8/16). Water/cement ratio (W/C) was 0.67, corresponding to normal-strength concretes produced around 1950s (Thomas and Bamforth, 1999). The samples were cast in a laboratory and strength class determined at 28 days was C12/15.

3.1. Electrical resistivity from laboratory

Files Resistivity measurements were performed continuously on cylindrical samples at selected time intervals of 7, 14, 28, 56, 91 and 161 days after casting. From the set of measured values from different times it is possible to determine the value of the m-factor from equation (3). The procedure is described in detail in (Konečný et al., 2020). The testing was conducted by a Wenner Array probe that can be used to measure surface resistance. The device offers measurements at four points at a constant distance (approximately 5 cm). The method is non-destructive, and tests can be repeated to determine characteristics of the time-dependent diffusion process to evaluate the *m*-factor in Equation (3). However, the heterogeneity of material and possible non-uniform contact conditions may result in large test uncertainty.

3.2. Chloride profiles from in situ measurements

All Samples of approximate size 100 × 100 × 60 mm (plates) were prepared for long-term exposure under the bridge over the first-class road I/11, near Hrabyně (HR) and the bridge over motorway D1 near Ostrava-Svinov (SV). The traffic intensity under the HR bridge in 2016 was approximately 15 000 vehicles per day and the traffic intensity under the SV bridge in 2016 was 23 015 vehicles per day and the intensity above the bridge is 37 000 (IPSOS s.r.o., 2016). The samples were placed under both bridges approximately 15 meters from the road on November 1, 2018. Besides the top surfaces, all surfaces of the samples were protected by an epoxy coating to prevent the ingress of chlorides. The first set of samples (three plates) was removed on 1 May 2019, i.e., 225 days after concreting and 181 days since the beginning of exposure (approximately half a year). The second set of samples (three plates) was removed on 1 November 2019, i.e., 410 days from concreting, with 365 days of exposure (one year).

A sampling of concrete powder in selected layers to obtain chloride profiles was performed by drilling. The first 10 mm of the chloride profile were disregarded – it is a convection zone that is significantly influenced by many other chloride transport mechanisms other than diffusion (absorption, capillary suction, convection including washing-off by rain) or local micro-defects of the surface layers. Whereas generally depth of convection layers should be verified by in-situ measurements (DuraCrete, 2000), the recommendation provided in (François and Arliguie, 1999) is adopted here - depth of 5 - 15 mm for sound concrete may be assumed and thus 10 mm is considered as a representative value for the environmental conditions under investigation. Note that the range of 5 to 15 mm does not apply in the marine environment (Ye et al., 2012).

3.3. Ratio between diffusion coefficients based on electrical resistivity and chloride profiles

The diffusion coefficients from both locations (HR and SV) are estimated by the two selected methods and recomputed to the reference age of 28 days using the equation (2) in order to have a consistent maturity level. The comparison is made by means of the correlation factor - ratio $D_{c,NT443,28}$ / $D_{c,res,28}$. $D_{c,NT443,28}$ represents the diffusion coefficient from the chloride ponding test (ASTM C1202, 2012) and $D_{c,res,28}$ from concrete resistivity (AASHTO T358, 2013). The results from chloride ponding were computed from three samples per each location and particular age of exposure (three samples for exposure of half a year and three samples for exposure of one year). The measurement for the HR_A sample for the exposure one year was identified as an outlier with two data points only and was eliminated from further analysis. For each sample, four readings of electrical resistivity were evaluated. The concrete was produced in one batch for both HR and SV samples; therefore, the resistivity estimates apply to all the samples.

4. Results

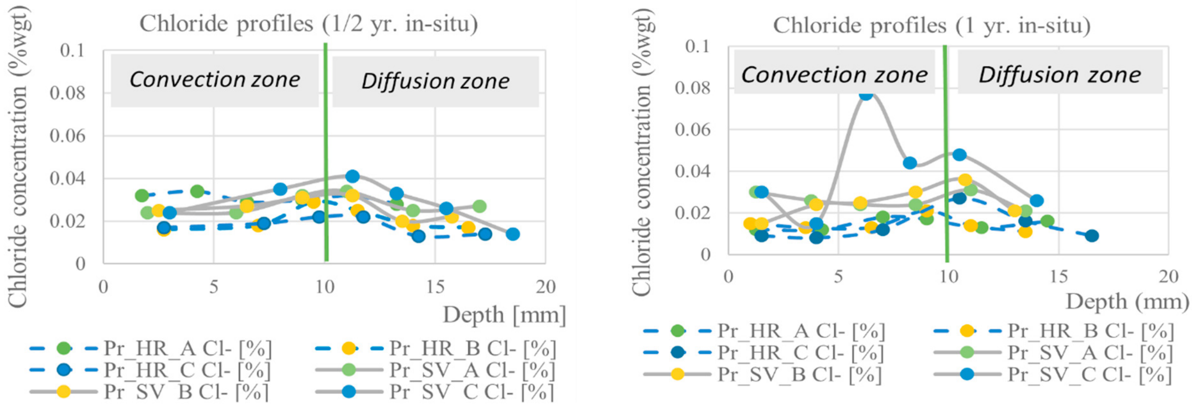
The elementary statistical analysis provides mean *m*, standard deviation *s* and coefficient of variation *CoV* of the estimates of the diffusion coefficient. The resulting diffusion coefficients recalculated to the concrete maturity of 28 days are given in Tab. 1.

Table 1. Diffusion coefficients of the resistivity $D_{c,res,28}$ and chloride profile $D_{c,NT443,28}$ (recalculated to a reference level of 28 days).

Sample	Exposure (years)	m factor (-)	Resistivity measurement $D_{c,res,28}$ (m/s^2×10^{-12})			Chloride profile measurement $D_{c,NT443,28}$ (m/s^2×10^{-12})		
			μ	σ	*CoV*	μ	σ	*CoV*
HR	1/2	0.1832	20.143	0.087	0.004	10.21	3.22	0.32
HR	1					10.00	5.55	0.55
SV	1/2					15.23	10.09	0.66
SV	1					5.00	0.73	0.15

It appears that the resistivity measurements lead to on average doubled D_c-values in comparison to the estimates based on chloride profiles. Further, *CoV* of $D_{c,res,28}$ is unrealistically low, much lower than the indicative values 0.05-0.2 (as discussed in the introduction). It seems that the resistivity may provide rough estimates of the diffusion coefficient only. The *CoV* from in-situ exposure indicates much larger variability – the values between 0.15 and 0.66 exceed significantly those presented in the literature. It thus seems that model uncertainty related to in-situ conditions may be large, when real chloride concentrations are unknown and associated uncertainty contributes to the model uncertainty in D_c. It is important to note that the exposure time of ½ or 1 year is quite short to provide sufficient insight into the effect of in-situ climatic exposure.

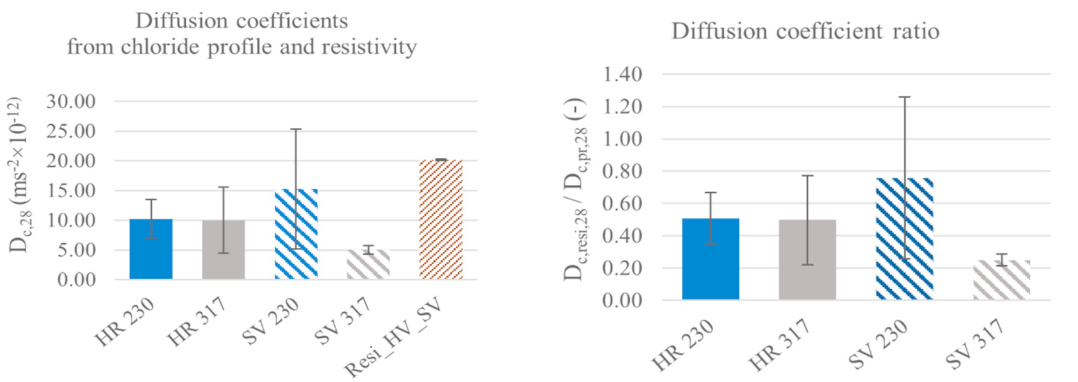
The measured chloride profiles are shown in Fig. 1. The effect of convection zone up to 10 mm of the sample depth is apparent for both exposure times. However, it is more distinctive in Fig. 1(b) for the longer exposure time. The effect of the convection zone is well shown by sample Pr_SV_B (Pr abbreviates profile). In contrast, sample Pr_HR_A – identified as the outlier - has a rather flat chloride profile.



(a) (b)

Fig. 1. Chloride profiles after exposure in situ: (a) ½ year; (b) 1 year.

The comparison of diffusion coefficients given in Fig. 2 (a) confirms that electrical resistivity leads to diffusion coefficients higher than those obtained from chloride profiles. Interesting to observe is that the HR diffusion coefficients are less variable than SV-values. This is judged to be related to the exposure at SV where the bridge is above the motorway with about two-times higher traffic intensity in comparison to HR. The $D_{c,NT443,28}$ / $D_{c,res,28}$ values are shown in Fig. 2 (b). The average ratio is 0.5. The ratio exhibits a similar variability as that observed for the diffusion coefficients.



(a) (b)

Fig. 2. (a) Diffusion coefficient; (b) Ratio between NT Build 443 [2] and electrical resistivity (AASHTO T358, 2013).

5. Summary and conclusions

This contribution investigates the concrete diffusion coefficient with respect to chloride ingress, focusing on measurements obtained by two methods - indirect laboratory measurements of electrical resistivity and direct chloride profile-based evaluation from samples exposed to in situ conditions.

The diffusion coefficients from indirect electrical resistivity are systematically higher in all cases than those based on chloride profiles. The estimates based on electrical resistivity exhibit unrealistically small variability (coefficient of variation of 0.004). In stark contrast, in-situ measurements result in significant variability (coefficient of variation ranging from 0.15 up to 0.66) which is much higher than indicative values provided in the literature.

It is judged that the systematically lower diffusion coefficients obtained from chloride profiles could be attributed to the fact that the requirement on constant surface concentration in these tests was not fulfilled; detailed analysis of the effect of varying chloride concentration on diffusion coefficient is within the scope of further research.

The obtained results suggest that:

- The estimate of diffusion coefficient from chloride profiles obtained from in-situ placed samples is deemed to be associated with large uncertainty in the case of short exposure times (say up to one year).
- The cores should be drilled from the structure and laboratory chloride penetration tests should be performed for the very young bridges in order to compute D_c.

Future research directions include:

- Model uncertainty evaluation for the chloride field exposure of young concrete (with age less than t_{lim}).
- Preparation of the lab-based chloride profiles as a reference to field exposure if possible.
- Study in order to specify t_{lim} with respect to a chloride profile analysis.

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