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A Modified Analytical Model for Simulating Chloride Ingress into Concrete Structures Compared with the Experimental LIBS Based Profiles

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Abstract: This article presents an attempt to compute chloride profiles of concrete structures exposed to chloride ions scanned by the laser-induced breakdown spectroscopy (LIBS) method using a modified analytical model. Five concrete samples were taken from a parking garage in the Swiss Alps. Subsequently, 2D profile images from these drilled samples were analyzed and processed to obtain diffusion coefficients for the chloride ingress and arithmetic data under the form of 1D chloride profiles. The modified analytical model was utilized by adopting a Monte Carlo (MC) simulation technique into the analytical formula of Crank's solution of the diffusion equation. Simulated results were quantitatively compared with those of LIBS measurements using the root mean square-error tool.

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Keywords: analytical model, chloride profile, concrete structures, diffusion coefficient, LIBS, Monte Carlo simulation.

Mathematics SubjectClassification: AMS-MOS: 65-11 Research data for problems pertaining to numerical analysis

PACS: 64.60.De Statistical mechanics of model systems (Ising model, Potts model, field-theory models, Monte Carlo techniques, etc)

1. Introduction

Durability assessment of reinforced concrete (RC) structures exposed to chloride ions can be performed by the analysis of 2D chloride profile images (Figure 1) due to the availability of modern scanning facilities such as LIBS technique [1-4].

Besides, the modeling of chloride ingress to RC structures can be conducted through both analytical and numerical models. Most of the analytical models are based on the application and development of Fick's law of diffusion [5-8]. In the meantime, numerical solutions for the equation of Fick's law in combination with finite differences or with a finite element method (FEM) scheme offer new

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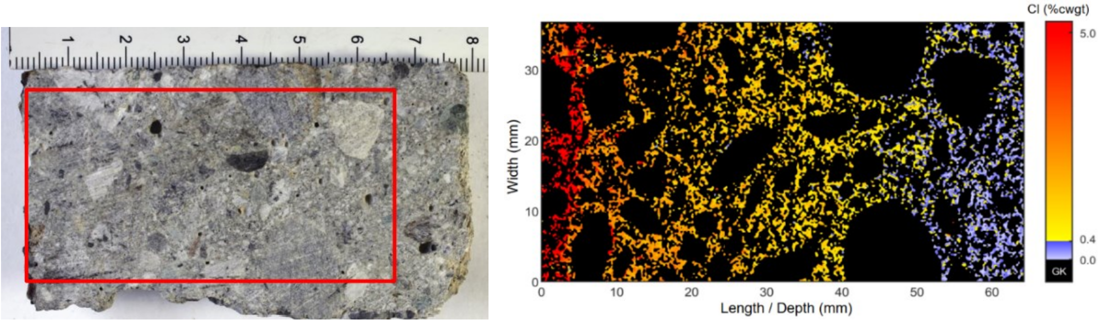
opportunities to solve more complex problems with more complex boundary conditions. Many numerical models have been developed and introduced for the simulation of chloride ingress into concrete, e.g. [9-15].

It is worth mentioning that the lack of statistical data of laboratory experiments is a general problem for estimating suitable input parameters for these numerical models especially more complex ones involving the spatial distribution of material parameters [16-19]. Even though, there are normative documents such as *fib*, *fib* Model Code 2010 [20] and related *fib* bulletin 76 [21] that provide indicative values of the diffusion coefficient and their variations. Moreover, the experimental campaign conducted on 32 high performance concrete mixtures provided a range of coefficient of variation of diffusion coefficient, V_{Dc}, between 0.03 and 0.06 with several extremes ranging up to $V_{Dc} = 0.1$ [22]. Other researcher [23] investigated a range of mixtures in laboratory conditions and found $V_{Dc} = 0.05$ as a lower bound (increasing V_{Dc} with increasing concrete strength and decreasing water/cement ratio). However, if the data would be available then it may be used for numerical modeling. In addition, numerical simulations require computing resources and are time consuming. By now, most of the above mentioned analytical models are deterministic and therefore the randomness of their input parameters was not considered.



(a) (b)

Figure 1: Photograph of one sample (a) and its 2D LIBS image of the cement based chloride distribution over the cross-sectional area (b) with the exclusion of aggregate (GK = aggregate; %cwgt = weight percentage of cementitious material).

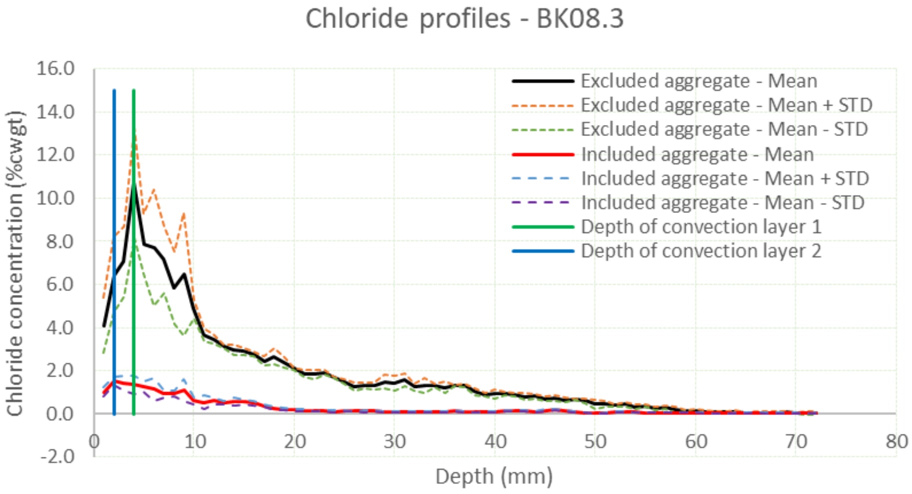
This research evaluates the diffusion coefficient based on the data from LIBS measurements, as indicated on Figures 1 (a) and (b). It aims to simulate chloride ingress into RC structures using a modified probabilistic analytical model and compare the results with field data obtained by LIBS technique. This model is built based on the application of the analytical formula of Crank's solution for the Fick's 2nd law of diffusion equation with the inclusion of MC simulation techniques. Simulated results are comparatively evaluated with respect to the LIBS measurements.

2. LIBS based test and resulted chloride profiles

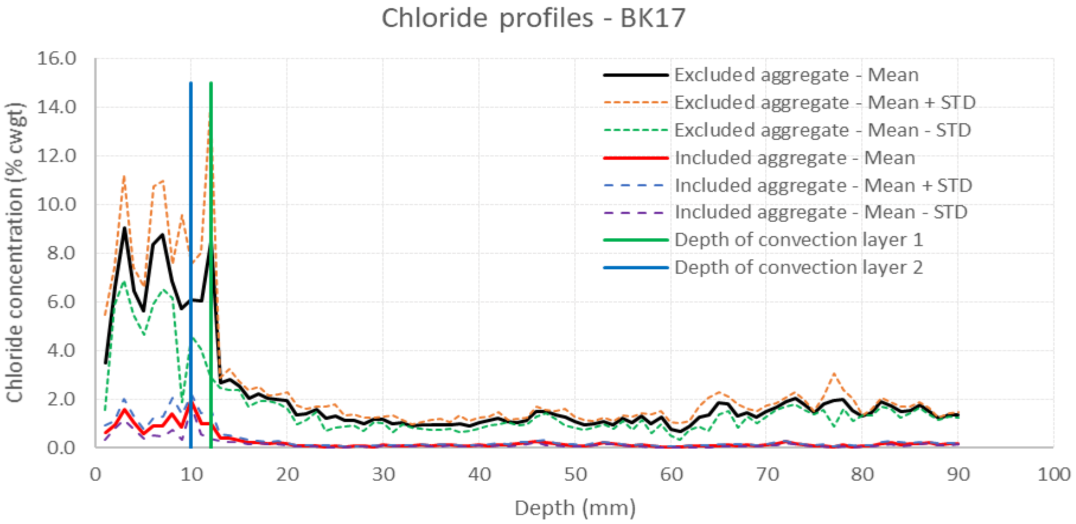
The principle of LIBS is the determination of the chemical composition of laser evaporated material exposed to a high energy laser pulse. Procedure of the technique is as follows: the radiation of the pulse is focused on the target surface via a quartz lens, plasma is generated due to a very high energy density produced at the focal point, through an optical fibre the light of the plasma is collected and guided to the entrance slit of the spectrometer, at the exit slit of the spectrometer the spectrally dispersed light is detected by the optical multichannel analyzer and finally, the data are collected in a time window by the computer [24]. The LIBS technique is able to evaluate the chloride concentration per mass of cementitious material as indicated in Figure 1 (b) considering multi-phase concrete structure (cement paste and matrix). Contrary traditional approaches based on grinding or drilling the concrete sample to a powder get the average amount of concrete and chlorides [25, 26].

Five concrete samples were taken from a parking garage built in 1980 in Switzerland. The parking garage was exposed to chloride from the beginning because it is located in the Swiss Alps. The samples were measured at VALTEST AG (Switzerland, Lalden) in December 2018. At the Faculty of Civil Engineering of the VSB – Technical University of Ostrava, chloride profiles were processed to obtain arithmetic data and scaled back to the depth of the samples. It is important to mention that the sample BK17 contains a crack. Thus, it is expected its chloride profile curves to be a significant outlier in comparison to those of the other four samples. In addition, the statistical properties of the diffusion coefficient and the surface chloride concentration, i.e. their mean value (Mean) and standard deviation (STD), of sample BK17 are expected to deviate from those of the other profiles.

In this study, depths of boundary of convection layers of all chloride profiles are determined as described in [27] and indicated as vertically continuous lines in Figure 2. In addition, certain filters and algorithms based on particular element ratios such as Ca/O are applied to analytically separated the binding agent from the aggregates after the LIBS measurement so that solely the chemical composition of the cement can be determined (Figure 2).



(a)



(b)

Figure 2: Processed chloride profiles of (a) sample BK08.3 and (b) sample BK17, including the position of depths of the convection layers (Mean = mean value, STD = standard deviation).

For each chloride profile of every sample, 1D approximation of 2D image is created where the image was divided into 64 to 90 layers (columns) based on the available resolution in pixels of each image. Data from each of the column were statistically processed in order to obtain Mean and Mean±STD values. The authors believe that the actual concentration in cement matrix obtained by LIBS and presented in Figure 1 (b) shall be used with caution when considering reference chloride concentration threshold values in concrete that is typically described as “homogeneous material”. This means it is necessary to examine if the obtained values are chloride concentration per weight of concrete or chloride concentration per weight of cementitious material because the traditional methods evaluate the concentration of the powder including the aggregate. Therefore, the chloride concentration data is processed in two ways: (i) including aggregate (aggregate contribution with zero chloride concentration, reducing the chloride ion concentration), (ii) excluding aggregate (analyzing chlorides only in the paste). Then, chloride profiles are created with two respective values of surface chloride concentrations: Mean+STD and Mean-STD, i.e., each sample is described by three chloride profiles (Figure 2). Moreover, it is worth mentioning that the chloride concentration is measured as a portion of mass of soluble chlorides to mass of cementitious material (%cwgt). For that reason, analysis with the effect of aggregate is related to practical application due to similarity with homogenized concrete approach in case of chloride threshold limits (see e.g. [28]). The analysis without the effect of aggregates would be study on the concentration in a cement paste.

Subsequently, representative parameters of these new profiles iteratively approximated by the method of least square curve fitting through equations (1) and (2) to obtain new representative values for diffusion coefficient (D_c) and surface chloride concentration (C_0):

$$S = \sum_{n=2}^{N} \Delta C^2(n) = \sum_{n=2}^{N} \left(C_m(n) - C_c(n)\right)^2 \qquad (1)$$

where S is the sum of squares to be minimized (percent by mass of total cementitious materials)^2, N is the number of concrete layers ground off, $\Delta C(n)$ is the difference between the measured and the calculated chloride concentration of the n'th concrete layer (percent by mass of total cementitious materials), $C_m(n)$ is the measured chloride concentration of the n'th concrete layer and $C_c(n)$ is the calculated chloride concentration in the middle of the n'th concrete layer using equation (2) and i'th iteration of D_c and C_0.

Because the five samples were taken from one structure, the statistical properties of D_c and C_0 can be averaged for all the samples and considered as one representation for the complete structure. These statistical properties include Mean and STD values (Table 1).

Table 1: Statistical properties of D_c and C_0 for every sample and the complete structure

Samples	Statistical parameters	Without aggregates		With aggregates	
		D_c ($\times 10^{-12}$ m^2/s)	C_0 (%cwgt)	D_c ($\times 10^{-12}$ m^2/s)	C_0 (%cwgt)
BK06	Mean	0.9502	2.4	0.9416	0.6
	STD	0.0077	0.3	0.0369	0.1
BK08.3	Mean	0.8393	3.0	0.6970	0.4
	STD	0.0447	0.6	0.0100	0.1
BK12	Mean	0.9918	2.7	0.7586	0.6
	STD	0.0052	0.3	0.0186	0.1
BK14	Mean	0.9653	3.8	1.0726	0.4
	STD	0.0503	0.9	0.0299	0.1
BK17	Mean	1.9306	3.4	1.2808	0.4
	STD	0.1981	0.8	0.0602	0.1
Complete structure	Mean	1.1354	3.1	0.9501	0.5
	STD	0.4483	0.6	0.2372	0.1

As can be seen from Table 1 that D_c and C_0 of sample BK17 are outlier as expected due to the above mentioned physical reason.

Variation coefficients of diffusion coefficient V_{Dc} of the five LIBS samples are provided in Table 2. It can be observed from Table 2 that the V_{Dc} of the five LIBS samples range from 0.014 to 0.047 if aggregates are taken into account. Meanwhile, the range of V_{Dc} is more extreme (both at lower and upper bounds) if aggregates are not considered, between 0.005 and 0.103.

Table 2: Coefficient of variation of diffusion coefficient for every LIBS sample

Sample		BK06	BK08.3	BK12	BK14	BK17
V_{Dc}	Without aggregates	0.008	0.053	0.005	0.052	0.103
	With aggregates	0.039	0.014	0.024	0.028	0.047

It is worth noticing that since the samples were taken from the structure with respect to evaluation of the chloride ingress and the spread of resulting coefficient of variation indicate within-batch, batch-to-batch, and location variability. However, the limited set of samples is hardly to be used in order to identify the source of variability.

3. Modified analytical model and its application in computing chloride profiles

An homogenized 1D model of the 2D chloride distribution of Figure 1b can be expressed by the analytical Crank's solution [5] of the diffusion equation:

$$C_{x,t} = C_0 \left\{ 1 - erf\left(\frac{x}{\sqrt{4D_c t}} \right) \right\} \tag{2}$$

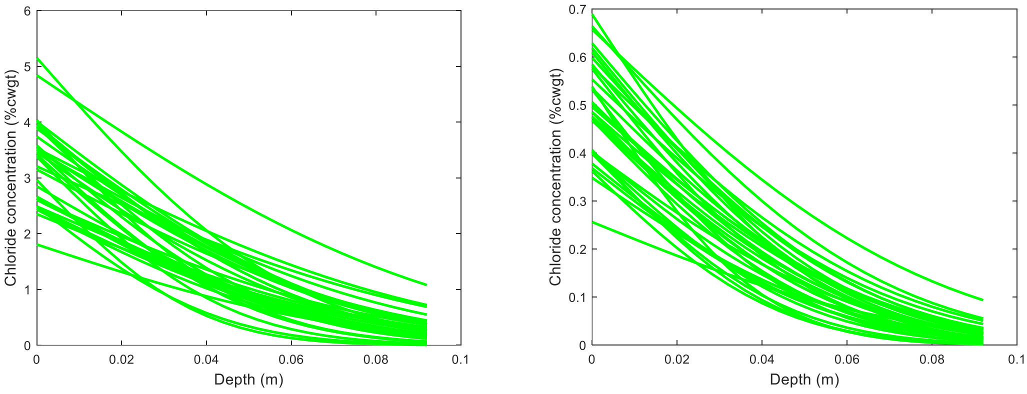
where $C_{x,t}$ is the concentration of chlorides (percent by mass of total cementitious materials) at time t (years) and at depth x (m), C_0 is the chloride concentration on the exposed surface of concrete (percent by mass of total cementitious materials), erf is the error function and D_c is the apparent diffusion coefficient (m^2/year).

On the one hand it is clear that the analytical solution cannot govern the stochastic nature of the input parameters because C_0 and D_c are constant. On the other hand C_0 strongly depends on initial and exposition conditions while D_c varies with concrete age. To consider the variation of D_c and C_0, a MC simulation-based approach is utilized.

MC simulations are based on a process of repeated random sampling to obtain numerical results by generating a large set of samples of limit state evaluations. Subsequently, the true value of the probability of failure is approximated by identifying the number of samples falling into the failure domain [29, 30]. With MC simulations, the generation of random variables and a desired distribution of a parameter can be modeled using the following formula [31]:

$$N(\mu,\sigma) = \mu + \sigma \times N(0,1) \tag{3}$$

where μ is a specified mean value, σ is a specified standard deviation, $N(0,1)$ represents random numbers from the normalized normal distribution, $N(\mu,\sigma)$ represents random numbers from the generated normal distribution.



(a) (b)

Figure 3: Chloride profiles simulated by model (2) using MC technique in two cases: (a) without aggregate and (b) with aggregate.

In order to facilitate the presentation of results in figures and tables, the analytical model based on the application of MC simulation is named as AMC model. It is worth emphasizing that AMC is a result of Monte Carlo simulation process with utilization of analytical formula to compute chloride concentration. Variations of D_c and C_0 for AMC simulation are based on a normal distribution with their Mean and STD values adopted from computed statistical properties of the LIBS results (Table 1). It is worth mentioning that the normal distribution was chosen because it is a frequently used probabilistic distribution function and is easy to use. In this study, 33 MC simulations are conducted for independent couples of D_c and C_0 (Figure 3).

Chloride profiles computed by AMC model are compared with those processed from LIBS measurements (Figure 4 and Figure 5). Noteworthy, for a better visualization, the convection zones in those profiles have been removed.

There is not such a good match between the chloride profiles simulated by AMC model and those measured by LIBS. Except for the case of Mean-STD, differences in chloride profiles between the two methods of measurement are larger in case of considering the aggregates. For a better evaluation, the comparison is also carried out through the quantification of simulated errors using the root-mean-square-error (RMSE) tool and the dimensionless term RMSE/C_0 as displayed in Table 3.

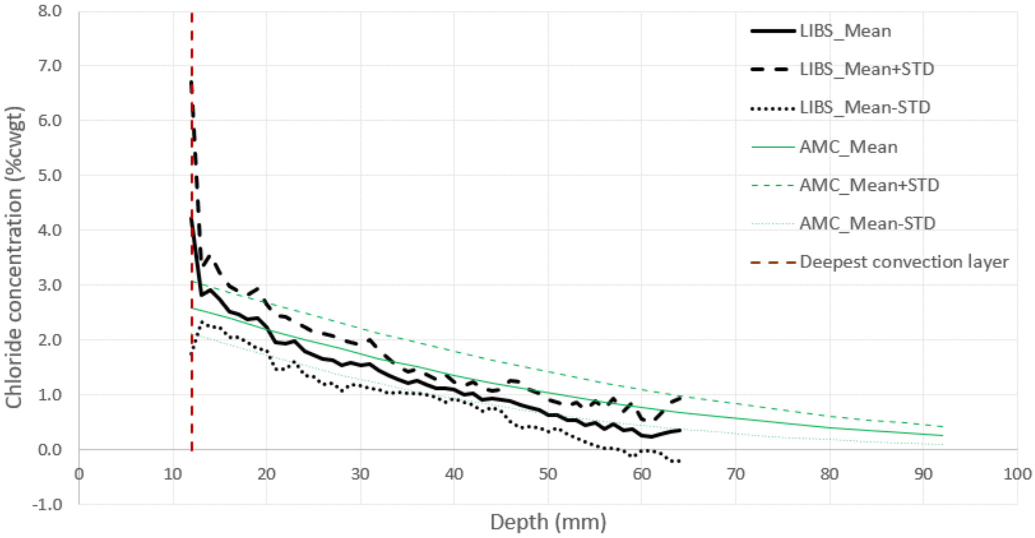


Figure 4: LIBS chloride profiles (without aggregates): Laboratory measurements compared with AMC model.

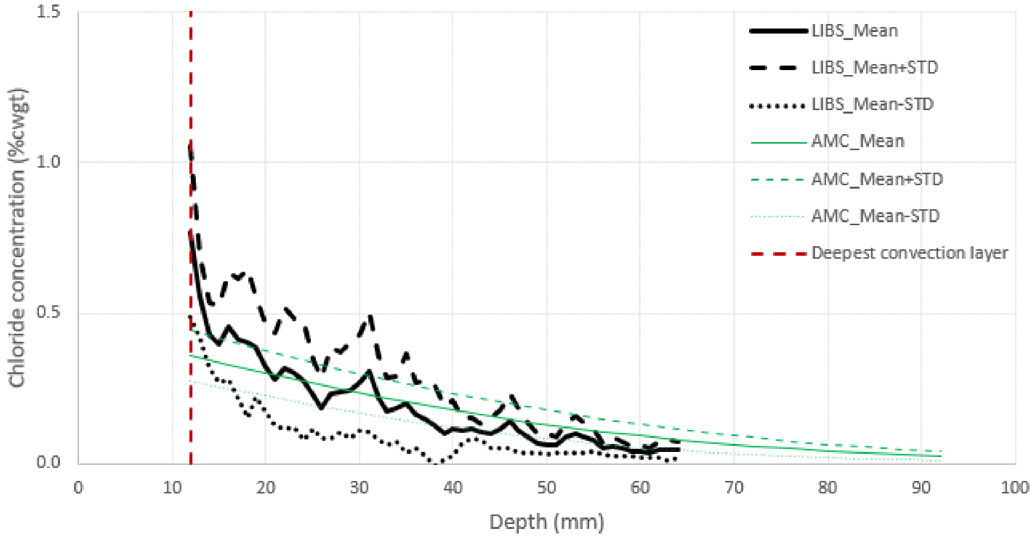


Figure 5: LIBS chloride profiles (with aggregates): Laboratory measurements compared with AMC model.

Table 3 reveals that the AMC model data significantly deviated from the LIBS measurements. Without aggregates the chloride profiles simulated by AMC model deviated for around 16.54% (Mean profile), 28.05% (Mean+STD profile) and 30.13% (Mean-STD profile) from the LIBS data, whereas 26.76%

(Mean profile), 33.44% (Mean+STD profile) and 24.21% (Mean-STD profile) with aggregates. These results confirm the observations from Figures 4 and 5.

Table 3: Computed RMSE/C_0 of AMC model compared to LIBS data

Parameters	Without aggregates			With aggregates		
	Mean	Mean+STD	Mean-STD	Mean	Mean+STD	Mean-STD
RMSE (%cwgt)	0.53	1.05	0.82	0.12	0.18	0.09
RMSE/C_0 (%)	16.54	28.05	30.13	26.76	33.44	24.21

4. Discussion

The major differences between resulted chloride profiles measured by LIBS and those computed by AMC model are concerned. Possible reasons are: (i) averaging data from the LIBS images to arithmetic data and scaling chloride profiles back to depth of samples reduced the maximum values of the original chloride profiles and, (ii) difference in thickness of convection layers of the five samples may have caused deviation in statistical parameters, especially the mean value of surface chloride concentration. It is worth mentioning that the AMC model does not consider the concrete maturity while in reality it affects diffusive flux especially in case of young structures, which is not the case herein. Moreover, this phenomenon is not affecting the difference between LIBS and AMC model because the diffusion coefficient was computed without considering the aging.

In addition, AMC simulation is a simple 1D probability-based model without requiring huge time and computational resources that describes concrete as homogeneous material including the probabilistic description of governing parameters. In contrast, more advanced models such multi-phase models [32, 33] or random-field [14], which could describe the heterogeneity of concrete, might be more capable of reproducing LIBS measurements.

The statistical parameters applied for the simulation and provided in Table 1 allows for the probabilistic modeling of the chloride concentration on the surface and the diffusion coefficient. Therefore, the Mean and STD values are evaluated allowing to describe the parameters with normal distribution of the probability of occurrence. Such data are sufficient with respect to the limitation of the prepared numerical model simulating the random field because Gaussian distribution is the only distribution widely available. Therefore, it is applied in the current form of the random field model [15] for the diffusion coefficient. It is worth mentioning that further investigation would be useful to check if the surface chloride concentration followed Gaussian distribution or other distributions as e.g. in data published in [34] and processed for the utilization in probabilistic assessment in [35].

Moreover, when the effect of aggregates is taken into account, values of V_{Dc} available in Table 2 (from 0.014 to 0.047) are slightly lower than those reported in [22]. If aggregates are excluded, resulted value of upper bound V_{Dc} (0.103) is almost equal to that stated in the work of [22] whilst the value of lower bound V_{Dc} (0.005) is ten times lower than that concluded by [23]. This can be explained by the fact that structure and site-specific conditions such as temperature, moisture and terrain related parameters affecting chloride deposition were not taken into account during the investigation of chloride diffusion coefficients. The evaluated concrete samples were taken from different location of one structure and does not represent the variation of concrete within the same concrete mix design or within the set of similar structures built independently.

5. Conclusions

A modified analytical model was constructed by integrating MC simulations into the analytical formula of Crank's solution for the Fick's 2nd law of diffusion equation. Subsequently, the chloride penetration to RC structures was simulated with input parameters obtained from computed statistical properties of D_c and C_0 of five LIBS measurements. Simulated chloride profiles were compared with those analyzed from LIBS images. The comparison was also quantitatively conducted by using a RMSE tool, displaying major differences (16.54% without aggregates and 26.76% with aggregates) in resulted chloride profiles simulated by the proposed model comparing to those scanned by LIBS.

The proposed model proved to be usable for addressing chloride penetration to concrete only with comparative analysis purpose. It does not provide sufficient precision in order to address the heterogeneity of the concrete analyzed by LIBS. To reproduce LIBS measurements, therefore, it is desirable to have an advanced multidimensional model that is expected to have capable of precise capturing chloride penetration to RC structures on which spatial variation of concrete material is explored, especially with respect to the correlation length and the scatter of random input parameters in order to distinguish between the cement paste and aggregate. Such a model might be built according to the scheme of probabilistic method to facilitate the description of spatial variation of material properties or multi-phase discrete particle model.

The LIBS analysis significantly improved the precision of obtained data from concrete samples not only in case of chloride ion ingress. It allows to capture the heterogeneity and evaluate the actual concentration of chlorides in cement paste and matrix. The comparison of homogenized 1D model and LIBS indicates the difficulties in the interpretation of the LIBS results in relation to the standardized chloride threshold levels.

To conclude, authors believe that the proper interpretation of the results related to evaluation of the chloride threshold levels with respect to non-nonhomogeneous 2D or even 3D chloride profile data deserve further attention. It is recommended to draw attention at the type of data of the chloride ion concentration in the LIBS images, i.e. if the results are concentration given as percentage of weight of cementitious material or of weight of concrete. Moreover, it is also important to check if the images give the information about total chlorides or soluble ones.

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