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Effects of carbonation and chloride ingress on the durability of concrete structures

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Abstract: The durability of concrete structures, which are designed for long-term use is predominantly determined by the resistance to chemical influences, *i.e.*, the concrete's ability to protect the reinforcement steel. The carbonation and the chloride ingress into concrete are the most significant causes of steel corrosion and the potential failure of the structure. The primary goal is to ensure that any significant damage does not occur during the structure's service life, primarily achieved by selecting an adequate thickness of the concrete cover. The issue is approached through calculations based on performance analyses, and the use of appropriate models for these chemical phenomena. The paper provides a brief overview and the methodology for analysing the impact on the durability of concrete structures in accordance with the leading international normative documents. The emphasis is on the recent changes introduced in second generation of European Eurocode standards. The consequences of the analysed phenomena are presented through the results of field tests conducted at salt factories, coke industries, and thermal power plants, and through laboratory tests. The tests were performed in order to develop a rapid prediction method for the measure of chloride ingress into concrete without the stimulating chloride ion migration by electricity, as an alternative to standardized tests.

Keywords: service life; deterioration, field tests; laboratory tests; Eurocodes; depassivation.

INTRODUCTION

Concrete structures are exposed to different environment conditions and are vulnerable to damage from corrosion. The progressing corrosion is the major cause of deterioration of concrete structures. The durability of concrete structures receives significant attention in many international normative documents and recommendations. The Euro-international Committee for Concrete (fib) has published a Design Guide,[1] while International Union of Laboratories and Experts in

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Construction Materials, Systems and Structures (RILEM) has published a monograph.[2] The theoretical foundations of durability design are presented in the monographs by Richardson (2002)[3] and Alexander *et al.* (2017),[4] while the fib's "Structural concrete" textbook,[5] in Chapter 5, describes the durability of concrete structures. In some standards, durability is associated with the service life design of concrete structures,[6] while it is treated separately in the American Concrete Institute's approach.[7,8] European structural standards are based on a probabilistic approach,[9] and for concrete structures, there are designated standards[10] for specification, performance, production and conformity.[11] Like in ACI recommendations, in the fib's guide,[12] the provisions for service life are separated (in the Model Code,[13] this is outlined in clauses 7.8.2 and 7.8.3). The fundamentals analysis of concrete durability design is a subject of the paper by Folic (2009),[14] while modelling and structural assessment in durability design are considered by Folic *et al.* (2010).[15] The method of designing service life according to the provisions of the fib Model Code MC 2010 and its implementation into ISO 16204 is analysed by Helland (2013).[16] A comprehensive review of the literature on the introduction of carbonation and chlorides into the analysis of corrosion in reinforced concrete structures is the subject of the study by Zhou *et al.* (2014).[17] A similar literature review on durability and service life, with a critical approach to modelling, is presented in the article by Alexander *et al.* (2019).[18] Demis *et al.* (2019)[19] consider the issues and perspectives of designing the durability of concrete structures while Helland (2022)[20] describes the performance-based service life as it is implemented in the 2021 Eurocodes. In Europe, the design for the durability of new reinforced concrete structures is currently based on a prescriptive approach. However, designers must understand the basic deterioration mechanisms and the potential types and rates of damage development, as it is given in next generation of European standards for different type of corrosion.[21] Since 2014, EN1992[10] and EN206[11] are under the main revision definition of exposure class for the new generation standards system to specify durability.[20,22] The new tendencies in designing durability of concrete structures, and state of the art issues, are subject of paper by Folic and Brujic,[23] where pre-normative CEN documents,[24–27] which will be adopted as the second-generation Eurocodes, are discussed.

The group of authors, Folic, Zenunović and Rešidbegović, published articles[28–31] related to the investigations of carbonation and chloride corrosion, in situ and in laboratories. Within the context of two case studies of complex industrial structures located in an aggressive environment, the issue of durability of concrete has been considered, the recommendations regarding structural durability analysis are considered by the means of verification of the limit state of depassivation and the reinforcement corrosion induced by chloride ingress, and the models for predicting chloride ingress in concrete samples and in existing concrete structures have been suggested.

In this paper, the theoretical foundations related to their implementation into the analysis of the durability of reinforced concrete structures were discussed. The primary goal of this study is to illustrate the examples of applying fundamental sciences, especially chemistry, in the practice of designing concrete structures, particularly in terms of their durability.

Carbonation and chloride ingres in standards (CODES)

In international standards, structural reliability is defined as the "ability of structure to fulfil the specific designed requirements during the design service life". Degradation (mainly in structural materials), which affects the structural durability, accumulates slowly over time and gradually leads to the deterioration of structural performance. For the performance assessment, it is necessary to investigate the material degradation caused by long-term effects of chemical, physical and other factors, and their effects on structural resistance. In the case of concrete structures, the investigation of structural durability, and its testing, primarily involves the analysis of corrosion of reinforcing steel. The designers should have a comprehensive understanding of fundamental deterioration mechanisms, as well as the potential types and rates of damage development (different types of corrosion cause very different damage developments, some of which reduce the structural safety). The parameters which influence durability are the cement type and quality control of early age cracking, the limitation of crack width, the environmental aggressiveness, etc. The models for the description of the deterioration mechanisms must integrate knowledge from a wide range of different fundamental and applied disciplines.

In deterministic design of durability, actions (S), resistance (R) and service life are used as deterministic quantities. The design formula compares two quantities for the target service life, t_g:

$$R(t_\mathrm{g}) - S(t_\mathrm{g}) > 0 \tag{1}$$

In reality, S and R are time dependent functions. In the case of the service life the principal design formula is: $t_\mathrm{L} - t_\mathrm{g} > 0$, where t_L is the service life function.

In stochastic design method the distributions of S, R and service life are taken into account. Probability that the service life of a structure is shorter than the target life is smaller than a certain allowable failure probability is written as:

$$P\{\mathrm{failure}\}_{t_\mathrm{g}} = P\{t_\mathrm{L} < t_\mathrm{g}\} < P_\mathrm{fmax} \tag{2}$$

The problem can be solved if the distribution of service life is known. The design service life is: $t_\mathrm{d} = \gamma_\mathrm{t} t_\mathrm{g}$, where t_d is the design service life, while γ_t is the lifetime safety factor. The service life principle may be written:

$$R(t_\mathrm{g}) - S(t_\mathrm{g}) \geq 0, \quad t_\mathrm{L} - t_\mathrm{g} > 0 \tag{3}$$

The lifetime safety factor must be calibrated with the results of stochastic design methods and value depends on the maximum allowable failure probability.

The resistance $R(t)$ of a structure and the applied loads $S(t)$ both are stochastic time functions. At any time, t, the margin of safety $M(t)$ is:[8]

$$M(t) = R(t) - S(t) \tag{4}$$

The function $P_\mathrm{f}(t)$ has the character of a distribution function. Considering the continuous distributions, the failure probability P_f at a certain moment of time can be determined using the convolution integral:

$$P_\mathrm{f}(t) = \left[M(t) < 0\right] = \int_{-\infty}^{\infty} F_R(s) f_S(s)\, ds \tag{5}$$

in which $F_R(s)$ and $f_S(s)$ are probability distribution function of R and density function of S, while s is the common quantity or measure of R and S. It is important to establish a model describing action and the resistance with acceptable reliability. Steel in concrete is protected against corrosion by passivation, due to the alkalinity of concrete (the pH of the pore water runs up to greater than 12.5). If the pH of concrete around reinforcement drops below 9 or chloride content exceeds a critical value, the passive film and the corrosion protection will be lost.[1]

The verification of design requires definition of the limit states, and identification of the required design service life and reliability. Recently, besides the serviceability limit state (*SLS*) and the ultimate limit state (*ULS*), some codes introduce the condition limit state (*CLS*), *i.e.*, "use of depassivation as a limit state of durability".[19] For the depassivation, the suggested probability for failure is 10^{-1}, and reliability coefficient is $\beta = 1.3$, while for ULS it is between 10^{-4} and 10^{-6}, depending on the consequences of potential failure.[12]

In ACI,[8] the corrosion models for reinforced concrete are based on a general deterioration model (0) that has been developed to predict the service life of reinforcing steel (Tuutti, 1982). Time moment at the transition from the initial to the propagation phase is introduced as a depassivation point.

Instead, in fib, a multilinear dependence of corrosion on time is introduced for propagation (0).

The length of the initial period is controlled by the rate of the chloride ions in concrete. The one-dimensional diffusion process follows Fick's second low of difusion,[8] here presented in a modified form, as applied in the analysis of concrete structures in the industrial zone of Tuzla:

$$\frac{\partial C(x,t)}{\partial t} = D_\mathrm{c}\frac{\partial^2 C(x,t)}{\partial t^2} \tag{6}$$

where C is a concentration of chloride ions, and D_c is a coefficient of diffusion at distance x from surface at time t. This is further discussed in ACI's Report on service life prediction,[8] theoretical background are given,[14–22] while its application is implemented in the next generation Eurocode text.[27]

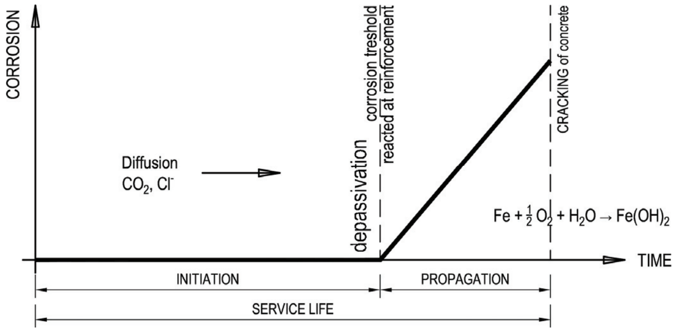

Fig. 1. Schematic of conceptual model of corrosion of steel reinforcement in concrete.

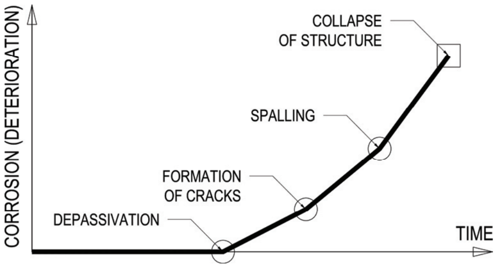

Fig. 2. Determination of service life and limit states with respect to reinforcement corrosion.

With the assumption of a constant chloride concentration on the surface, C_S, of concrete, the typical solution to the previous expression is in the form:

$$C(x,t) = C_\mathrm{S}\left(1 - erf\frac{x}{2\sqrt{D_\mathrm{t}}}\right) \tag{7}$$

Carbonation

The passive ferric oxide film on embedded steel may be disrupted by a reduction in the alkalinity (pH < 10.5) of the concrete by carbonation (CO_2 penetrate in concrete) or by the presence of aggressive ions such as chlorides and sulphates. The chemical reaction may be described as:

$$\mathrm{Ca(OH)_2} + \mathrm{CO_2} \xrightarrow{\text{Water}} \mathrm{CaCO_3} + \mathrm{H_2O} \tag{8}$$

The empirical formulae for depth of carbonation (x) may be adopted as:

$$x = k\sqrt{t} \tag{9}$$

where k is the factor depending on diffusivity, reserve alkalinity, carbon dioxide, concentration, and expose condition. Some formulae modification and theoretical verification of the square root relationship, and the application of models to service life prediction are introduced and discussed by Richardson.[3]

Penetration of chlorides into concrete

Chlorides (originating from sea water or de-icing salt used in winter) may penetrate through the pores to the interior of the concrete. The decrease in diffusion coefficient is due to the effects of penetrated chlorides, from de-icing salts, leading to an ion exchange with subsequent blocking of pores in the surface layer. The critical chloride content, indicating the incipient danger of corrosion and subsequent cracks and the spalling of concrete, depends on various parameters. In the region of cracks, carbonation and chlorides tend to penetrate faster towards the the reinforcement than in uncracked concrete. Fick model with apparent diffusion coefficient (Eq.(6) and Crank's solution (Eq.(7)) are given in the literature.[3] During the 1980s, Schiessl examined the use of finite difference models, as well as other models.[3] For the corrosion of reinforcement, as a simplification, two single processes may be separated: the cathodic and the anodic process (0).

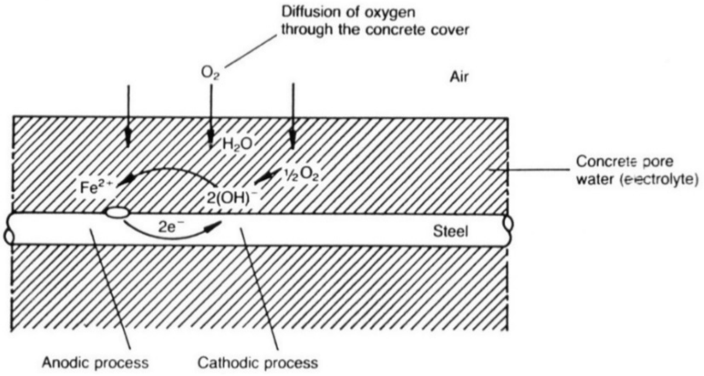

Fig. 3. Simplified model for corrosion of reinforcement in concrete.

In June 2010 fib Technical Council approve start of activities on the MC2020 project to undertake the provisions for the design of concrete structures and will provide them for new and for existing structures, including promotion of structural safety, service ability and durability (limit state of depassivation). Empirical models for corrosion initiation due the carbonation and chloride ingress are defined by fib[12,16] (deemed to satisfy provision and the application of modelling methods). Providing partial factors for carbonation assuming normal serviceability limit state reliability requirements. Like in EN[10], fib[13] introduces the limit state of depassivation – the following requirement shall be fulfilled:

$$P\{\} = P_{\mathrm{dep}} = P\left\{\mathrm{c} - x_{\mathrm{c}}(t_{\mathrm{SL}}) < 0\right\} < P_0 \tag{10}$$

where $P\{\}$ is the probability that depassivation occurs, c is the concrete cover; $x_c(t_{\mathrm{SL}})$ is the carbonation depth at time t_{SL} in mm, t_{SL} is the design service life in years, while P_0 is the target failure probability. The variables c and $x_{\mathrm{c}}(t_{\mathrm{SL}})$ need to be quantified in a probabilistic approach.

The design model, the ingress of the carbonation front, may be assumed to obey the following:

$$x_{\mathrm{c}}(t) = W(t)k'\sqrt{t} \tag{11}$$

where k’ is a factor reflecting aspects like the execution, basic resistance of the chosen concrete mix (like water-cement ratio, cement type, additions) under the reference conditions and the influence of the basic environmental conditions (like mean relative humidity and CO_2 concentration) against the ingress of carbonation, and $W(t)$ is a weather function taking the meso-climatic conditions due to wetting events of the concrete surface into account. For the partial safety factor format the following limit state function shall be fulfilled:

$$c_{\mathrm{d}} - x_{\mathrm{c,d}}(t_{\mathrm{SL}}) \geq 0 \tag{12}$$

where index c,d denotes the design value of the concrete cover (c_{nom} – safety margin), $x_{\mathrm{c,d}}(t_{\mathrm{SL}})$ is a design value of the carbonation depth at time t_{SL} in mm, while c_{nom} is nominal value for the concrete cover. The design value of the carbonation depth at time t_{SL} is calculated as follows:

$$x_{\mathrm{c,d}}(t_{\mathrm{SL}}) = x_{\mathrm{c,c}}(t_{\mathrm{SL}})\gamma_{\mathrm{f}} \tag{13}$$

where $x_{\mathrm{c,c}}(t_{\mathrm{SL}})$ is characteristic value of the carbonation depth, *e.g.*, mean value of the carbonation depth, and γ_f is partial safety factor of the carbonation depth.

In Europe, design for the durability of new reinforced concrete structures is currently based on a prescriptive approach, *i.e.*, deemed-to-satisfy design: within this approach a trading-off of geometrical (concrete cover to reinforcement), material parameters (indirectly linked to the diffusion and binding characteristics) and execution aspects (compaction and curing) is applied.

The following limit state function shall be fulfilled (see Eq. (10)):

$$P\{\} = P_{\mathrm{dep}} = P\left\{C_{\mathrm{crit}} - C(c, t_{\mathrm{SL}}) < 0\right\} < P_0 \tag{14}$$

where C_{crit} is the critical chloride content to achieve depassivation of the reinforcement, and $C(c,t_{\mathrm{SL}})$ is the chloride content at depth c and time t_{SL}. The variables c, C_{crit} and $C(c,t_{\mathrm{SL}})$ shall be quantified in a probabilistic approach.

The area of durability design is regulated by a number of normative documents applied in Europe[9–11] and USA,[7,8] and the documents of the fib,[12,13] which are intended to represent the state-of-the-art in theory and practice. Traditionally, in Europe, as well as in most others leading codes worldwide (even in present Eurocodes related to concrete structures EN 1992-1-1 and EN 206), although ensuring durability is nominally based on performance-based approach, the durability design and verification of performance follows the, so called "prescriptive" or "deemed-to satisfy" (DtS) rules. In the literature[23] some selected aspects and their theoretical background related to the European perspective on performance-based durability design for reinforced concrete structures, which are implemented in new, second-generation, Eurocode FprEN 1992-1-1[26] are mentioned. The exposure classes in Eurocodes refer to categories that classify the anticipated environmental conditions to which a structure will be exposed over its intended design life. The exposure classes are defined for the most common environmental exposure conditions: 1) the corrosion of embedded steel induced by carbonation (XC), when reinforced concrete is exposed to air and moisture; 2) the corrosion of the embedded steel induced by chlorides (XD; de-icing salts usually) or 3) chlorides from sea water (XS); 4) chemical attack (XA1 to XA3). The environmental exposure classification has retained the same form as in present Eurocode 2, only some of the descriptions have been modified for the classes related to the corrosion of reinforcement. Since the selection of a specific exposure class is a designer's decision, in the second-generation of Eurocodes, the definition of exposure classes has been copied from EN 206[25] to the main text.[26] The exposure Resistance Classes (ERC) are used to classify concrete with the respect to resistance against the corrosion induced by carbonation (class XRC) and by chlorides (class XRSD) were covered only through limiting values for mix compositions (similar to DtS approach). Briefly, the exposure resistance classes link the minimum concrete cover required for durability, $c_{\mathrm{min,dur}}$, to exposure class and design service life. ERC is, thus, a set/group of requirements for concrete which are needed to resist the type of exposure associated to an exposure class. The adequate durability may be assumed against the corrosion caused by carbonation or chloride ingress, where cover to reinforcement is selected appropriate to the exposure class, exposure resistance class and the design service life and not less than the minimum cover for durability $c_{\mathrm{min,dur}}$.

EXPERIMENTAL INVESTIGATIONS ON SITE AND IN LABORATORY

The experimental investigations were conducted in the facilities of the Salt Factory, Coke Industry and the Thermal Power Plant in Tuzla. By examining the pollution of air and water in the area, the presence of chemical compounds aggressive in contact with concrete and reinforcement, such as CO, CO_2, SO_2, SO_3,…, has been determined. The comprehensive field research and laboratory testing have been conducted in order to determine the content of chlorides and sulphates, as well as the depth of carbonation. It has been established that the primary cause of the corrosion process in concrete is the presence of chloride (consequences are shown in 0).

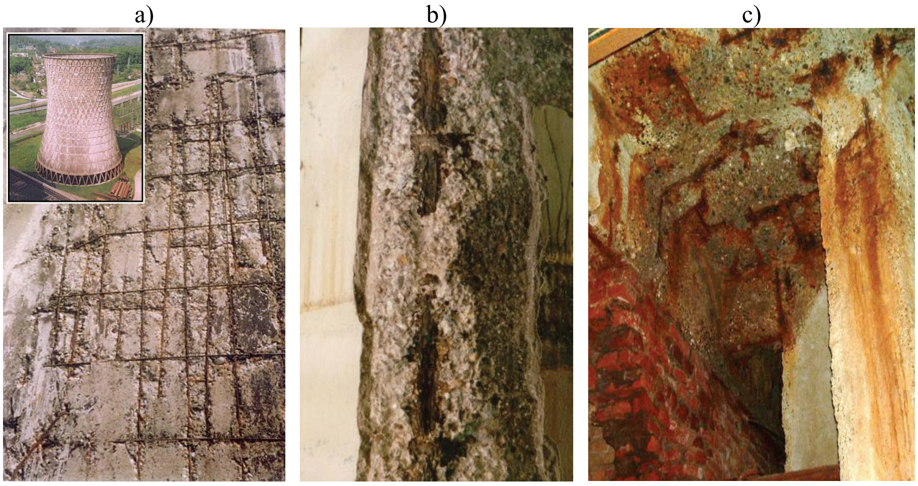

Fig. 4. Effects of chloride ingress: a) cooling tower in power plant; b) salt factory; c) nitrogen factory in the coke industry.

The reinforcement carbonation depth is determined by testing with 0.1 % alcohol phenolphthalein and 0.1 % alcohol thymolphthalein. The procedure is performed by making a cut on the concrete surface, on which the solution is sprayed. The healthy concrete depth is shown through the violet colour of the solution. The chloride concentration is determined by applying two substances, 1 % $AgNO_3$ and 5 % K_2CrO_4, one after another. The indication of chloride presence is the brown colour of the substances.

The amount of chloride and sulphate in samples of concrete removed was determined by laboratory testing. The amount of chloride was determined by filtering the sample in distilled water with $K_2Cr_2O_7$ solution, which is subsequently added to the standard solution of $AgNO_3$. The amount of sulphate was determined by filtering with additions to the solution HCl and $BaCl_2$.

In order to define a rapid method for determining the chloride penetration profile in concrete without the initiation of chloride ions by electrical current, the laboratory research was conducted in the laboratories of the University of Tuzla. The chloride diffusion coefficients through the concrete surface layer exposed to salted water with and without pressure were analysed. Three concrete mix formulas were used. The samples were tested by immersing them into salty water (bulk diffusion test – BDT) and pressuring them with salted water (pressure penetration test – PPT). The term permeation coefficient was introduced, which represent a difference of the transport mechanisms during BDT and PPT. The PPT procedure was applied to define the chloride profile of sampled concrete from the Salt Factory. The procedure of determination of the chloride content by depth is described in detail in the literature.[29,30] The sampling was performed by grinding samples by depth for the purpose of determining chloride concentration and using the indicator colour to determine the chloride penetration depth. After the chloride content in the supplied concrete samples was determined, they were prepared for the PPT examination in the way presented in 0. The samples are named the stepped samples.

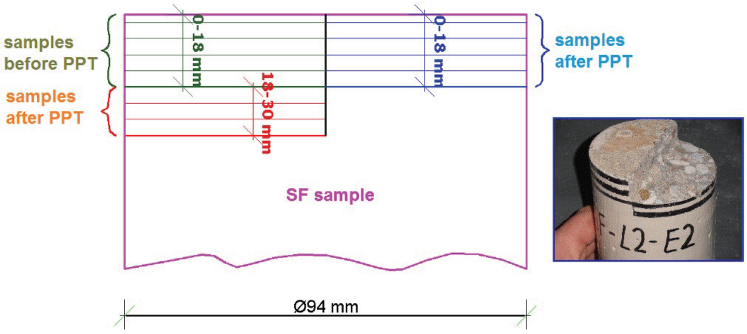

Fig. 5. Stepped samples.

The goals of testing the stepped sample are simultaneous determination of the residual capacity for the chloride absorption in the surface layers and the determination of the chloride penetration depth in the old concrete (0). The procedure is described in more detail in the literature.[31]

TABLE I. The pH value, chloride content, sulfate content and the depth of chloride penetration

Sample	pH	Content, %		Depth of chloride penetration, cm
		Chlorides	Sulfates	
1	11.5	0.113	0.90	1.0
2	11.0	0,098.	0.69	1.0
3	9.5	0.128	0.76	2.0
4	11.0	0.135	0.74	1.0
5	11.0	0.137	1.30	1.0
6	9.5	0.103	0.83	2.5
7	9.5	0.113	0.71	2.5
8	10.5	0.098	0.88	1.0
9	11.0	0.015	0.46	1.0
10	11.0	0.015	0.96	1.0

The investigation of concrete carbonation was conducted by the determination of the pH value of the concrete samples. Depending on the determined values, a 0.1 % alcohol phenolphthalein indicator solution was applied for pH 8.2–9.8, and a 0.1 % alcohol thymolphthalein indicator for pH 9.3–10.5. The determined pH values ranged from 9.0 to 11.0. The depth of carbonation at selected locations of the deteriorated concrete ranged from 15 to 20 mm. The rates of carbonation are the highest in the range from 50 to 70 % of relative humidity. Above 75 % of the relative humidity the influence of water filling the pores becomes significant. The coefficient of variation for measuring the depth of carbonation V = 30 % and the distribution function is shown in 0.

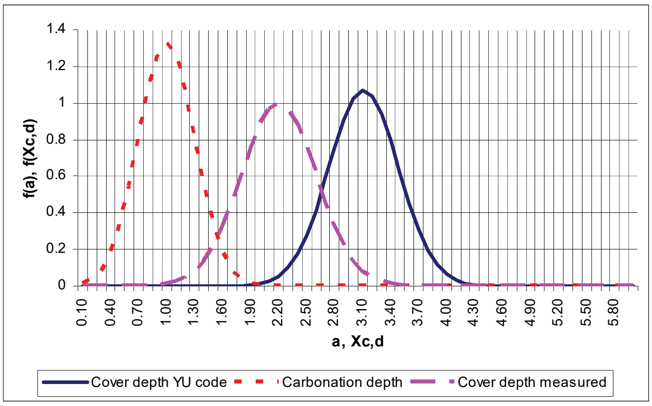

Fig. 6. Functions of distribution of cover concrete depth and carbonation depth.[28]

In the Salt Factory, the predominant impact is notably from chloride, which has penetrated the structure up to the reinforcement. By extracting the samples, the amount of soluble chloride was determined to be between 0.14 and 0.3 %. However, it is important to note that the chloride and sulphate content is significantly higher in concretes where the concrete cover has been spalled due to the reinforcement corrosion, ranging up to 2 % for chlorides and sulphates. This high chloride content has caused significant damage and a decrease in concrete strength of 30 % or more. In the Thermal Power Plant, the chloride content was determined to be up to 0.15 %, and the sulphate content up to 1 %. A more detailed presentation of the results is given in the literature.[28] The sampled concrete from the Salt Factory was used to create a predictive model of chloride penetration using a stepwise sample (0). Here, a representation of one chloride profile determined in the affected concrete from the Salt Factory is provided (0). The obtained diagrams are described in detail in the literature.[31]

The aim of this investigation was to determine the possibility of chloride penetration at greater depths, where the concrete is firmer and less porous, and to assess the remaining capacity for chloride absorption.

CONCLUSIONS

Parameters which influence the durability of a concreate are the cement type and the quality control of early age cracking of concrete, water–cement ratio,

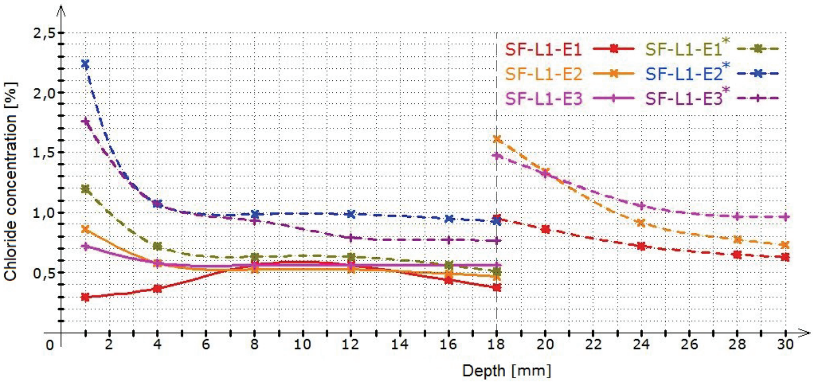

Fig. 7. Chloride profiles in stepped samples – location L1.

limitation of crack width, the environmental aggressiveness, etc. Even if in most cases durability concerns the serviceability, the resistance verification during the design of concrete structures, the inclusion of durability aspects and the deterioration mechanisms are a necessity. The deterioration is a result of external environmental exposure of a concrete, which is most often the carbonation of concrete and chloride corrosion of reinforcement, especially when using de-icing salt and near marine or industrial regions.

The European approach to durability design is in continuous development. To overcome the inconsistencies of the current European prescriptive approach, a new durability design concept has recently been proposed – the “exposure resistance classes”.[21] While the exposure classes given in actual Codes[10,11] refer to categories that classify anticipated environmental conditions, to which a structure will be exposed over its intended service life, the exposure resistance classes relate to the performance of concrete in such environments.

The models for describing the deterioration mechanisms have to integrate the knowledge from a wide range of different fundamental and applied disciplines, especially chemistry. They need to be founded in a reliable manner and to provide some appropriate testing options, which is essential for the design or the assessment of structures. The assessment of the existing concrete structures, which is important before rehabilitation, must be performed timely, by the combination of *in situ* testing and laboratory testing of samples taken from the structures.

The further research is directed towards the investigation and assessing the corrosion of prestressed concrete structures.

ИЗВОД

УТИЦАЈ КАРБОНИЗАЦИЈЕ И ПРОДОРА ХЛОРИДА НА ТРАЈНОСТ БЕТОНСКИХ КОНСТРУКЦИЈА

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Трајност бетонских конструкција, које су предвиђене за дуготрајну употребу, пре свега зависи од отпорности на хемијске утицаје, односно од способности бетона да заштити челик за арматуру. Карбонација и улазак хлорида у бетон су најзначајнији узроци корозије челика и потенцијалног квара конструкције. Примарни циљ је да се у току радног века конструкције не појаве значајна оштећења, пре свега избором адекватне дебљине бетонског покривача. Овом питању се приступа кроз прорачуне засноване на анализама перформанси и коришћењем одговарајућих модела за ове хемијске феномене. У раду је дат кратак преглед и методологија за анализу утицаја на трајност бетонских конструкција у складу са водећим међународним нормативним документима. Акценат је на недавним променама уведеним у другу генерацију европских Eurocode стандарда. Последице анализираних појава приказане су кроз резултате теренских испитивања спроведених у фабрикама соли, коксаре и термоелектрана, као и кроз лабораторијска испитивања. Тестови су спроведени да би се развила метода брзог предвиђања за мерење уласка хлорида у бетон без стимулисања миграције јона хлорида електричном енергијом, као алтернатива стандардизованим тестовима.

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