

Cement and Concrete Research 32 (2002) 935-941



Modeling carbonation for corrosion risk prediction of concrete structures

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Abstract

Damage of reinforced concrete structures is often caused by corrosion of steel reinforcements due to carbonation. Although literature on carbonation has become vast, a comprehensive numerical model for quantitative prediction of the corrosion risk that allows for environmental influences such as temperature and humidity seems lacking. The aim of the present paper is the development of a theoretical model to predict carbonation of concrete structures. The model describes movement and retention of heat, moisture and carbon dioxide (CO_2) by means of balance equations and diffusion laws. The balance equations are coupled and take into account the interaction between different transport and storing processes. A new mathematical formulation of the function of moisture in balance faithfully represents the moisture-storing properties of a porous media. The evolutionary equation of the reaction of CO_2 is derived from reaction kinetics that are described by the Arrhenius' function. The model is solved by means of an efficient numerical method using a Finite Element concept and numerical time integration techniques. It is verified by using results from experimental tests reported in the literature. Taking into account changing atmospheric conditions, structures are investigated with respect to the corrosion risk of steel reinforcements. Together with threshold values taken from the literature, the numerical results give the corrosion risk of reinforced concrete structures. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Carbonation; Transport properties; Durability; Mathematical modeling; Finite element analysis

1. Introduction

Concrete itself is a porous compound material consisting of mineral aggregates and the cement matrix that form a durable structure. The porosity of the concrete renders possible movement and retention of water and other substances. Concrete generally protects steel reinforcements from corrosion. The protection is due to the high alkalinity of the pore solution that causes passivation of the steel, i.e. the buildup of an extremely thin inert layer on the steel's surface. Chemical reactions of penetrating carbon dioxide (CO₂) with the cement matrix reduce the alkalinity of the pore solution. Thus, the protecting property of the concrete diminishes, and the steel becomes susceptible to corrosion.

The risk of corrosion of a steel reinforcement is given by the distribution in time and space of relevant state variables with respect to corrosion. Aggressive substances such as

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chlorides in marine environments will not be dealt with in this study. Thus, relevant state variables are temperature, moisture and degree of carbonation. Threshold values for relevant state variables taken from the literature may be used as criteria for the evaluation of the corrosion risk.

The identification of the carbonation as important factor for the durability of reinforced concrete structures dates back to the very beginnings of concrete research, when the experimental basis has become vast. In 1965, the German Research Institute of Cement Related Industry conducted a large-scale study on existing concrete buildings measuring depths of carbonation [7]. Laboratory studies on transport and reaction of CO₂ in various cement pastes were carried out by Kropp [10]. Bier [1] tried to realkalize carbonated ordinary portland cement (OPC). Experimental studies on carbonation of various fly ash concretes under laboratory and atmospheric conditions were conducted by Thomas and Matthews [17]. Further studies on concrete samples under atmospheric conditions were carried out by Bunte and Rostásy [3]. Recent accelerated carbonation and weathering studies were reported by Roy et al. [12]. A model for the kinetics of the carbonation reaction was set up by Brieger and Wittmann [2]. It was

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combined with a one-dimensional diffusion model for heat, moisture and CO₂ flow by Saetta et al. [13] who subsequently proposed a two-dimensional extension [14]. Bunte and Rostásy [3] develop a method based on weather-measuring techniques and statistical interpretation for the lifetime assessment of concrete structures with respect to carbonation. For carbonation of high-volume fly ash concrete that differs considerably from that of OPC concrete, a mathematical model is proposed by Jiang et al. [9].

The new model combines results of extensive studies by Bunte and Rostásy [3] on diffusion of CO_2 in different types of concrete and the modeling of the reaction kinetics of carbonation by Saetta et al. [13] with a comprehensive coupled temperature and moisture model for concrete. The study focuses on the crucial influence of the environmental conditions a concrete structure is exposed to on the progress of carbonation.

2. Carbonation of concrete

 CO_2 penetrates into the concrete mainly gaseous by diffusion through air filled pores. The diffusion through water-filled pores and the convection within water that moves in the pores may be neglected. The balance of mass describes that the sum of all changes in concentration of free CO_2 κ must be equal to changes of flux q_K . The reaction of carbonation withdraws free CO_2 from the mass balance. Thus, the reaction of CO_2 is taken into account by a sink term in the balance equation. Denoting the concentration of reacted CO_2 by κ_T , the balance equation is

$$\dot{\kappa} + \dot{\kappa}_{\rm r} = -\text{div}\boldsymbol{q}_{K}.\tag{1}$$

Transport and reaction of CO₂ depend on temperature, relative pore humidity and degree of carbonation, as shown in the following sections.

2.1. Diffusion of CO₂

The diffusion coefficient of CO₂ in concrete cannot be measured experimentally, because carbonation takes place already during testing and falsifies the measurements. Therefore, oxygen is used as an inert gas to determine the diffusion coefficient. Oxygen diffusion coefficients for concrete can experimentally be measured in function of cement type, cement content and curing [3].

The diffusion of CO₂ is described by Fick's law. The diffusion flux depends on the concentration gradient of free CO₂, thus,

$$\mathbf{q}_K = -\mathbf{\lambda}_K \cdot \operatorname{grad}\kappa,$$
 (2)

where the diffusion coefficient is a function of temperature, relative pore humidity and degree of carbonation

$$\lambda_K = D_O f_T f_H f_k I. \tag{3}$$

Gertis et al. [8] describe the dependency on temperature ϑ with a function (Eq. (4))

$$f_T = \frac{\vartheta + 273.0}{293.0}. (4)$$

High humidity hinders diffusion. From experimental tests on OPC pastes, the empirical function (Eq. (5))

$$f_H = a_1 + \frac{a_2}{1 + (a_3 - a_3 \varphi)^{a_4}} \tag{5}$$

can be derived for the dependency on humidity φ [3]. For OPC, its parameters are identified as a_1 =0.993, a_2 =-0.974, a_3 =3.621 and a_4 =5.750. This function that is shown in Fig. 1 is crucial for the carbonation progress. The reaction of carbonation changes the pore structure of the cement paste and thus the transport properties of the material, which will be discussed in detail in Section 2.2 dedicated to the reaction of CO₂. The influence of the reaction on the transport coefficients may be described by function

$$f_k = \exp[c_1 k] \tag{6}$$

with material parameter c_1 , where k denotes the degree of carbonation, i.e. the actual maximum reacted CO_2 ratio [3].

2.2. Reaction of CO₂

During hydration, the anhydrous calcium oxide of the cement, CaO, forms calcium silicate hydrate, mCaO-SiO₂·nH₂O, denoted by C-S-H, and calcium hydroxide, Ca(OH)₂. Whereas C-S-H is insoluble, the alkaline Ca(OH)₂ may dissolve in the pore liquid. Additional alkalis are supplied in form of sodium and potassium oxides of the cement. They dissolve in the pore liquid during hydration, forming sodium, potassium and hydroxyl ions, respectively. As these ions do not take part in the formation of the major cement hydration products, they accumulate in the pore solution and thus render calcium very insoluble. The pH of such pore fluids may well be greater than 12.4.

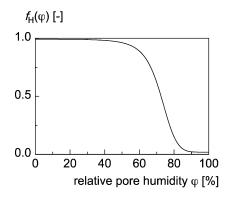


Fig. 1. Humidity effect on diffusion coefficient of CO₂.

The CO_2 that penetrates into concrete reacts with the alkaline hydration products of the cement paste. The reaction is called carbonation. To some extent, C-S-H also reacts with CO_2 ; however, this reaction is difficult to figure out in detail because of the variety of C-S-H phases. Exact stoichiometric relations are unknown. The formation of crystalline calcium carbonate and amorphous hydrous silica can be simply described (Eq. (7))

$$C-S-H+CO_2 \xrightarrow{H_2O} CaCO_3 + SiO_2 \cdot nH_2O.$$
 (7)

This reaction, however, does not affect the alkalinity of the pore liquid and may be neglected. After hydration, sodium hydroxide and potassium hydroxide are completely dissolved in the pore liquid and therefore react readily with penetrating CO₂ that dissolves in the pore liquid. After neutralization of the alkali ions in form of carbonates, Ca(OH)₂ dissolves in the pore solution to restore the chemical equilibrium. As long as calcium ions are available, the formed sodium and potassium carbonates are unstable and reorganize with the dissolved Ca(OH)₂ to calcium carbonate, which crystallizes into stable calcite and metastable aragonit. The calcium carbonate is almost insoluble. Both dissolution of alkalis and solution of CO₂ require free water in the pores.

Only when the soluble Ca(OH)₂ is completely bound in CaCO₃ are the sodium and potassium carbonates stable. Because the concentration of soluble Ca(OH)₂ in the cement paste is far greater than those of sodium hydroxide and potassium hydroxide, it is the governing factor for the alkalinity of the pore liquid. Thus, carbonation may simply be described by the reaction of Ca(OH)₂, as stated also by Taylor [16]. Neglecting the minor alkalis, the chemical reaction of carbonation may, for the sake of simplicity, be reduced to

$$Ca(OH)_2 + CO_2 \xrightarrow{H_2O} CaCO_3 + H_2O.$$
 (8)

The reaction of carbonation sets free water that was combined in Ca(OH)₂. As a rough estimation, the molecular masses $M_{\rm H_2}{\rm O} = 18.0$ g/mol and $M_{\rm CO_2} = 44.0$ g/mol yield 0.4091 g water set free by the reaction of 1 g CO₂. Thermogravimetric experiments on carbonation of cement pastes conducted by Kropp [10], however, reveal only a mass of 0.17–0.23 g water set free per 1 g CO₂. The difference may be due to carbonation of other alkalis that react without setting free water.

The chemical reaction results in structural changes of the cement paste. Moreover, the volume of the reaction products may change with respect to the initial volume of the reactants. For carbonation of Ca(OH)₂, Enculscu [6] identifies a volume increase of 11% for calcite and 3% for aragonite formation, respectively, which is confirmed by De Ceukelaire and Van Nieuwenburg [4] who indicate a volume increase of approximately 11% for the formation of calcium carbonate from Ca(OH)₂. The carbonation influence on the

concrete porosity, however, is discussed controversially. Bunte and Rostásy [3] observe no significant influence of carbonation on the porosity accessible under atmospheric pressure. Kropp [10] and Bier [1] measure a decrease in total porosity and pore volume of OPC concrete as a result of carbonation. It is widely accepted that concrete made of OPC cement becomes denser when carbonating, which again is confirmed by a recent study by Dias [5], whereas concrete made of blast–furnace cement becomes more permeable. These effects are taken into account in the definition of the diffusion coefficient by the function involving the carbonation degree (see Eqs. (3) and (6)).

2.3. Model equations for carbonation processes

For analyzing carbonation numerically, the chemical reactions according to Eq. (8) must be modeled. The reaction velocity depends on temperature and concentration of the involved species water, CO₂ and Ca(OH)₂. Extending a formulation by Brieger and Wittmann [2] based on the Arrhenius' function, Saetta et al. [13] introduce the reaction velocity

$$\mathbf{v}_{\rm r} = \dot{\kappa}_{\rm r} = \alpha A \exp\left[-\frac{E_0}{RT}\right] f_H f_K f_{K\rm r}. \tag{9}$$

The intensity of activation E_0 denotes the energy required to start the reaction. The impact number A is the number of collisions of reactive particles per second. Factor α indicates that the reaction takes place in the open system of the cement paste. These parameters are identified from material tests by Saetta et al. [13]. The influence of humidity on the reaction is given by function (Eq. (10))

$$f_{H} = \begin{cases} 0 & : & \varphi \leq \varphi_{\min} \\ \frac{5}{2} (\varphi - \varphi_{\min}) & : & \varphi_{\min} < \varphi \leq 0.9 \,. \\ 1 & : & 0.9 < \varphi \end{cases}$$
 (10)

The chemical reaction requires a certain amount of water in the pores since CO_2 and $Ca(OH)_2$ must dissolve and enter in contact. The minimum water content required for the reaction is unknown. A minimum relative pore humidity of $\phi_{min}\!\approx\!50\%$ can be estimated. Furthermore, it may be assumed that the reaction velocity reaches its maximum when the relative pore humidity exceeds 90%. The reaction velocity depends also on the concentration of free CO_2 in the pores that is modeled by the function (Eq. (11))

$$f_K = \frac{\kappa}{\kappa_{\text{max}}},\tag{11}$$

where the maximum concentration κ_{max} of free CO_2 in the concrete is equal to the atmospheric concentration $\bar{\kappa}_{\infty}.$ Finally, the reaction velocity depends on the concentration of Ca(OH)_2 dissolved in the pore liquid. Simplifying, it may be assumed that the concentration of available Ca(OH)_2 is proportional to the concentration of CO_2 $\kappa_{\scriptscriptstyle T}$ already reacted

with the $Ca(OH)_2$. As a rough estimation, the linear function (Eq. (12))

$$f_{Kr} = 1 - \frac{\kappa_{\rm r}}{\kappa_{\rm rmax}} \tag{12}$$

gives the dependency on the available $Ca(OH)_2$. The maximum concentration of CO_2 that may combine with reactive alkalis is denoted by $\kappa_{\rm rmax}$. From experimental tests on carbonated portland cement paste, a relation between the calcium oxide concentration C of the cement and the maximum concentration of reacted CO_2 $\kappa_{\rm rmax} = 0.0052z(C-3.2)$ can be derived [3], where z denotes the cement content of the concrete.

3. Governing equations and numerical solution procedure

The balance of mass, Eq. (1), the diffusion law, Eq. (2), and the evolutionary equation, Eq. (9), hold for CO_2 . The balance equation for CO_2 is coupled with those for thermal energy and water by modeling the interaction of all processes [11]. Since temperature and moisture affect transport and reaction of CO_2 , both the diffusion law, Eq. (2), and the evolutionary equation, Eq. (9), contain coupling terms related to temperature and humidity. Similarly, the balance equation for water contains a coupling term to take into account the influence of the reacted CO_2 $\kappa_{\rm r}$, i.e. the water set free by the reaction.

For thermal energy and moisture, represented by temperature ϑ and relative pore humidity ϕ , respectively, the balance equations are

$$\begin{split} & \left[\rho_{\rm C} c_{\rm C} + w c_{\rm W} + r \left(\psi - \frac{w}{\rho_{\rm W}} \right) \frac{\varphi}{RT} \left(\frac{\partial p_{\rm s}}{\partial \vartheta} - \frac{p_{\rm s}}{T} \right) \right] \dot{\vartheta} \\ & + r \frac{p_{\rm s}}{RT} \left(\psi - \frac{w}{\rho_{\rm W}} - \frac{\varphi}{\rho_{\rm W}} \frac{\partial w}{\partial \varphi} \right) \dot{\varphi} + \text{div} \boldsymbol{q}_T^T + \text{div} \boldsymbol{q}_T^H = 0, \end{split}$$

$$\tag{13}$$

$$\left(\psi - \frac{w}{\rho_{W}}\right) \frac{\varphi}{RT} \left(\frac{\partial p_{s}}{\partial \vartheta} - \frac{p_{s}}{T}\right) \dot{\vartheta}$$

$$+ \left[\frac{\partial w}{\partial \varphi} + \frac{p_{s}}{RT} \left(\psi - \frac{w}{\rho_{W}} - \frac{\varphi}{\rho_{W}} \frac{\partial w}{\partial \varphi}\right)\right] \dot{\varphi} + c_{H}^{K} \dot{\kappa}_{T}$$

$$+ \operatorname{div} \boldsymbol{q}_{H}^{T} + \operatorname{div} \boldsymbol{q}_{H}^{H} = 0. \tag{14}$$

They are specified by diffusion laws for the fluxes

$$\mathbf{q}_{T}^{T} = -\left(\hat{\lambda}_{T} + r\mathbf{D}_{g}\frac{\partial p_{s}}{\partial \vartheta}\varphi\right) \cdot \text{grad }\vartheta,$$
 (15)

$$\mathbf{q}_T^H = -r\mathbf{D}_{\mathbf{g}}\,p_{\mathbf{s}} \cdot \operatorname{grad}\,\varphi,\tag{16}$$

$$\mathbf{q}_{H}^{T} = -\mathbf{D}_{g} \frac{\partial p_{s}}{\partial \vartheta} \varphi \cdot \text{grad } \vartheta,$$
 (17)

$$\mathbf{q}_H^H = -(\mathbf{D}_{\mathbf{g}} p_{\mathbf{s}} + \mathbf{D}_f) \cdot \text{grad } \varphi. \tag{18}$$

Material constants are ρ for density, ψ for porosity and c for thermal capacity with indices C and W denoting concrete and water, respectively. R is the gas constant, T the temperature in Kelvin and r the vaporation enthalpy. The saturation pressure is p_s . The transport coefficients of vapor and water, \mathbf{D}_g and \mathbf{D}_f , respectively, are defined more detailed in Steffens [15]. The relationship between the concentration of water w in the concrete and the relative pore humidity is given by the function of moisture in balance (Eq. (19))

$$w = \begin{cases} \rho_W \frac{w_m c \varphi(1+2\varphi)}{1+c\varphi+c\varphi^2} & : \quad \varphi \le 0.5\\ \rho_W \left(a \ln \left[\frac{-4\omega}{\rho_W RT \ln \varphi} \right]^n + b \right) & : \quad 0.5 < \varphi < 1.0, \qquad (19)\\ w_s & : \quad \varphi = 1.0 \end{cases}$$

where ω denotes the surface tension of water at 20 °C and $w_{\rm s}$ the water content at saturation. The material parameters $w_{\rm m}$, c, a, b and n are determined by means of a Levenberg–Marquardt algorithm. The mathematical formulation is verified by experimental data and faithfully represents the moisture storing properties of a porous media.

Coupled balance equations, diffusion laws and the evolutionary equation form a coupled initial value and boundary value problem. Variables are temperature ϑ , relative pore humidity φ , concentration of unbound CO_2 κ and fluxes of temperature, moisture and unbound CO_2 . The concentration of bound CO_2 κ_r is understood as inner variable. Substitut-

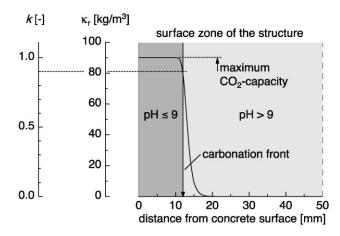


Fig. 2. Concentration of bound CO₂ and carbonation front.

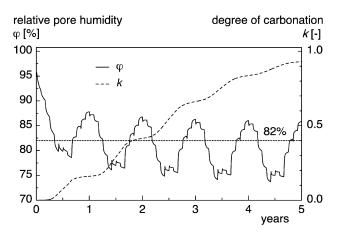


Fig. 3. Development in time of relative pore humidity and degree of carbonation.

ing the diffusion laws, Eqs. (15)–(18), and the evolutionary equation, Eq. (9), into the balance equations, Eqs. (1), (13) and (14), yields a system of coupled differential equations

$$\mathbf{C}\dot{\mathbf{z}} - \operatorname{div}[\mathbf{L} \cdot \operatorname{grad} \, \mathbf{z}] = 0 \tag{20}$$

with the state variable $z = [\vartheta \varphi \kappa]^T$. The numerical solution of Eq. (20) is developed by means of a consistent linearization of the coupled nonlinear partial differential equations and the discretisation by the Finite Element concept. The numerical solution technique is described in detail in Steffens [15].

4. Numerical results

Characterizing quantity for the carbonation is the degree of carbonation k. Because the reaction of CO_2 is much faster than its diffusion, a reaction front of a few millimeters of thickness builds up. As shown in Fig. 2, this front is idealized at k=0.9. The diffusion coefficient of CO_2 decreases con-

depth of carbonation [mm]

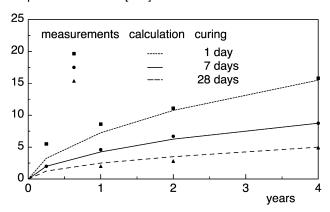


Fig. 4. Carbonation depth: comparison of own predictions with [17].

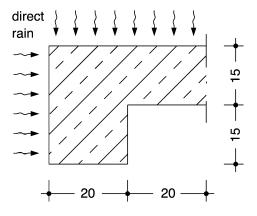


Fig. 5. Geometry of the structure [cm].

siderably if the relative pore humidity exceeds 60%. For relative pore humidity higher than 82%, the diffusion coefficient drops to 10% of its value for dry concrete, as shown in Fig. 1. Thus, the wetting of parts of a structure due to rain significantly affects the development in time of the carbonation depth. The presented model takes into account changing atmospheric conditions by means of coupling of the balance equations. Fig. 3 illustrates the relationship between dry periods and carbonation progress by representing the time development of relative pore humidity and the degree of carbonation, respectively.

To verify the developed model, results of carbonation tests on concretes under laboratory conditions made by Thomas and Matthews [17] are chosen. These material tests focus on

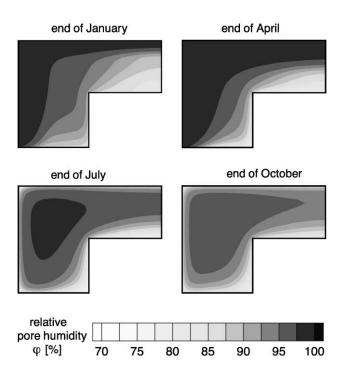


Fig. 6. Relative pore humidity of the structure during the 10th year.

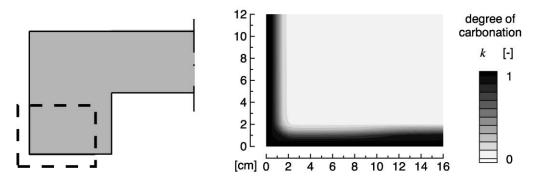


Fig. 7. Section and intensity of carbonation.

the influence of curing on the carbonation progress. The model allows for the curing effect by means of different CO₂ diffusion coefficients $D_{\rm O}=5.00\times10^{-4}$, $D_{\rm O}=1.36\times10^{-4}$ and $D_{\rm O}=0.30\times10^{-4}$ m²/h for curing times of 1, 7 and 28 days, respectively, [3]. The atmospheric CO₂ concentration is $\bar{\kappa}_{\infty}=0.0012$ kg/m³. The model parameters of the reaction $E_0=2.08\times10^6$ N m/kg, $A=3.6\times10^{10}$ h⁻¹ and $\alpha=3.0\times10^{-7}$ are determined experimentally [13]. For the cement, C=64.5% and z=300 kg/m³ hold. Fig. 4 shows good agreement of the numerical results computed with the new model and the measured carbonation depths.

Realistic simulations of transport and reactions in concrete structures are often only possible by means of multidimensional models. The following theoretical example illustrates the applicability of the model to real problems. The concrete structure under consideration is shown in Fig. 5. Two sides of the structure may be hit directly by rain, the remaining surfaces are sheltered. Changing atmospheric boundary conditions are prescribed as monthly constant mean values of temperature and relative humidity. The structure is exposed to the increased atmospheric CO₂ concentration of an industrial site, assuming $\bar{\kappa}_{\infty} = 0.001$ kg/m³. In order to illustrate the effects of carbonation, a rather poor OPC concrete is taken, made with a high water/ cement ratio of 0.7, $z=280 \text{ kg/m}^3$ and insufficient curing that results in an increased CO2 diffusion coefficient of $D_{\rm O} = 2.39 \times 10^{-4}$ m²/h. The model parameters of the reaction are identical with those given above. Fig. 6 shows the variation of relative pore humidity in the structure during the tenth year. Rain regularly wets the unsheltered rims and hinders drying of inner zones. Thus, the sheltered zones are significantly dryer during the whole year. Fig. 7 shows the carbonation of an edge sector of the structure after 50 years. In the zones exposed to rain, the carbonation depth reaches 7 mm, whereas the sheltered zones show already 13 mm of carbonation. Comparison with Fig. 6 illustrates the influence of relative pore humidity and thus the influence of atmospheric conditions on the carbonation progress. The relative pore humidity in the structure does not drop below 65%. Therefore, in the zones exposed to rain corrosion of steel reinforcements is possible within a rim of only 7 mm, whereas in the sheltered zones, corrosion may occur within a rim of 13 mm.

5. Summary and conclusion

In the numerical model, movement and retention of heat, moisture and CO_2 are described by balance equations and diffusion laws. Coupling of the balance equations takes into account interactions between different transport and storing processes. The evolutionary equation of the reaction of CO_2 is derived from the Arrhenius' function governing reaction kinetics. Coupled balance equations, diffusion laws and the evolutionary equation form a coupled initial value and boundary value problem. The numerical solution requires consistent linearization and discretisation by a Finite Element concept. Investigations of arbitrary concrete structures are possible by means of an efficient computer code.

Experimental data provided by thermo-hygrical and carbonation tests is used to verify the developed numerical model. One-dimensional analysis shows the ability of the model to account for interactions between transport and reaction. The analysis of two-dimensional structures demonstrates the applicability of the model to real problems. The distribution in time and space of the relevant state variables with respect to corrosion, which is given by the numerical analysis, is evaluated with regard to the corrosion risk of the steel reinforcements.

The developed numerical method is a thoroughly tested tool to evaluate the corrosion risk of steel reinforcements in concrete. It renders possible the durability rating of reinforced concrete structures during planning and thus may help avoiding damage due to steel corrosion.

Acknowledgments

Thanks are due to the Deutsche Forschungsgemeinschaft (German Research Council) for support under Grant Du 25/50 to the Institute for Structural Analysis, Technische Universität Braunschweig.

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