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NUMERICAL SIMULATION OF CHEMO-MECHANICAL DEGRADATION OF CONCRETE AND ITS IMPACT ON RC STRUCTURES BEARING CAPACITY

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Abstract. Durability of engineering structures depends on the structure itself and environmental conditions. Aggressive substances can penetrate concrete matrix causing chemical degradation of material skeleton. At the same time the material is exposed to mechanical exertion which contributes to additional deterioration. This work attempts to analyze chemo-mechanical degradation of concrete from phenomenological point of view using numerical simulation. Diffusion of corrosive species was determined through local diffusion parameters and balance equations with sources and outflows prescribed by stoichiometry of chemical equations and reaction extent constant. Degradation of concrete was described with use of an additive scalar damage parameter adopting adequate evolution equations. The cellular automata method used is shown to be an effective calculation method. Contour lines of species concentration and partial damage parameter values were obtained using numerical simulation of the processes in 2D space. As expected, progressive degradation of concrete decreased bearing capacity of the RC section. Numerical simulation of residual cross-section strength was obtained by use of short-time destructive loading tests.

Key words: concrete, corrosion, continuum damage mechanics, transport equations, cellular automata

INTRODUCTION

It is known that ceramic materials are significantly affected by stress-assisted corrosion. The phenomena take place when the materials are exposed to environmental influence with the addition of stress state loading. Deterioration mechanisms involve aggressive substances penetrating concrete porous structure. Due to topochemical reactions material skeleton degrades facilitating subsequent diffusion of aggressive agents and becoming more vulnerable to applied loads. From the point of view of thermodynamics, mechanical and environmental loadings are both the driving forces for irreversible material degradation. Their action is of synergetic nature, which means that their joint activity

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increases and accelerates the deterioration and failure of concrete compared to simple sum of their effects. There is no universal model for this problem due to complexity of encountered processes as well as nature of the material itself. Instead, several general models were formulated within the theory of mixtures. Differences rely on various types of chemical corrosion analysed and on the choice of fundamental phenomena described.

The paper concentrates on coupled interaction of chemical and mechanical damage in concrete. A short survey of present-day modelling is given to provide a general and comprehensive view justifying the proposed model. Next, the boundary value problem (BVP) is formulated with the use of internal variables formalism. The BVP equations together with evolution equations distinctly determine the adopted model. Numerical simulations of mechanical state of material and running processes demand novel methods of numerical calculations. Discretization of constitutive integral equation with use of recursively defined internal variable of creep strains, two different types of cellular automata modelling transport processes and construction of interaction curves are briefly presented. The results of numerical calculations, limited to one type of cross-section for brevity, clearly show a decrease in structural bearing capacity.

PROCESSESS TAKING PLACE IN CEMENTITIOUS MATERIALS

Some of the substances are very corrosive to cementitious materials, causing various effects diminishing bearing capacity of the system: dissolution of material skeleton, decalcification of hardened cement paste, reduction of the pH-value and synergetic coupling between mechanical and chemical loading. The deterioration and damage to the material may be intensified and accelerated, when it undergoes a corrosive attack simultaneously with a mechanical loading. A study of chemical attack effects superimposed with mechanical loading was reported by Schneider and Chen [1998, 2005]. The results showed clearly that the strength of cementitious materials declined much more rapidly under stress-assisted corrosion than under pure chemical corrosion. This has been called a chemomechanical effect. The effect depends greatly on the stress level, but it has been observed even for elastic stress.

Two main approaches have been used to model transport of solute species through porous material. The first one is based on a thorough description of all phases and constituents involved in the processes. In this approach multiple mass conservation equations are needed to obtain detailed description of the components fields. Depending on a situation different types of equations are used. In the case of pressure prevailing phenomena (encountered in soil mechanics, consolidation theory or gas flow, important pore pressure conditions and hygromechanical coupling) the theory developed by Biot is used [Meschke and Grasberger 2003]. A similar approach using pressure gradient is given by the Darcy state law [Glasser et al. 2008].

All these models are useful in investigating the fundamental mechanisms involved, but their use is rather complex and not realistic for structural computing and practical considerations [Le Bellégo et al. 2000]. Due to uncertainties involved in calibration of such complex models, structural lifetime can be more conveniently assessed by using a macroscopic approach which exploits the power and generality of the basic Fick's laws

to predict the quantitative response of systems undergoing diffusion. Early models were based on an analytical solution of the equation, resting on a series of simplifying assumptions of semi-infinite domain, constant diffusion coefficient and constant exposure conditions [Glasser et al. 2008].

In electric neutral mixture the concentration gradients are the main driving forces of molecular diffusion. The diffusion coefficient at the macroscopic scale is equal to the diffusion coefficient at microscopic scale modified by a purely geometrical factor of tortuosity [Samson et al. 1999], accounting for the complexity of the porous system. It is a function of the water content because it is related to the volume of liquid in the pore space [Samson et al. 2005], reflecting the connectedness of the moisture phase. Above a given saturation level, however, the diffusion coefficient might be independent of the water content. The influence of an externally applied stress on the concrete permeability remains poorly understood. Banthia et al. [2005], found that the presence of a compressive stress, below a certain threshold value, decreases the permeability. When the stress applied exceeds this threshold, a significant increase in the permeability occurs. This can be explained by joint effect of volumetric strains as well as micro and macro cracking induced by mechanical load. The same volumetric strains effect was observed by Werner et al. [2000] and Choinska et al. [2007].

The influence of various process parameters on the diffusivity of aggressive solutions can be taken into account by the use of a multi-factor law [Saetta et al. 1998, Saetta and Vitaliani 2004]. The effective diffusivity of a complex material can be evaluated combining diffusivities of inclusions (aggregate) and matrix (cement paste) in Christensen composite model [Xi et al. 2000]. Matrix permeability also depends on mechanical deterioration of the material [Pouya et al. 2013]. The fluid filled crack tends to increase permeability, whereas an empty (unsaturated) crack constitutes an obstacle to the flow and decreases the effective permeability in the direction orthogonal to its plane. The relation between porosity and permeability involves an intrinsic permeability tensor describing plane Poiseuille flow through discrete fracture zones [Bangert et al. 2003].

The equations modelling the chemical reactions can be solved simultaneously with the transport relationships. In Samson and Marchand [2007] opinion, the current trend in reactive transport modelling is to separate the transport part of the process from the chemical reactions. In the sequential iterative approach (SIA), the transport and chemical reaction equations are iterated with every step until convergence is reached. Another class of algorithms consists in solving the equations set without iterations between transport and chemical modules and is called the sequential non-iterative approach (SNIA).

If transport processes are slow compared to chemical reactions, the equilibrium relationships can be used instead of kinetic expressions. The local equilibrium assumption can be checked basing on the dimensionless Damköhler number, relating characteristic times for diffusion and chemical reactions: $a^D = \frac{t_D}{t_r}$. If $Da >> 1$, the time scales of the reactions are much shorter than the time scales associated with diffusion. Samson's [2007] calculations show that for a typical concrete composition the Damköhler number is 800. Due to relatively low diffusion coefficients, high reaction rate and high surface area of cementitious materials, the criterion of local equilibrium state is respected.

In Saetta et al. [1999] as well as in Cervera et al. [1999], the effective stress tensor is split into two components, d_m^+ and d_m^-, describing different independent mechanisms of concrete degradation under tensile or compressive loading conditions. The decomposition is performed over the free energy potential (i.e., the Helmholtz free energy). The two damage parameters grow independently, and the damage due to tension that has no effect on the response in compression and vice versa. This allows for describing both the crack closure effect as well as unloading and cycling loading consequences.

CONSTITUTIVE EQUATIONS

The constitutive equations for viscoelastic material of a concrete matrix skeleton are in the form of Volterra's integral equation of second kind:

$$\varepsilon(t) = \frac{\sigma(t)}{E(t)} - \int_{\tau_0}^{t} \sigma(\tau) K(t,\tau) d\tau, \quad \varepsilon(t_n) = \sigma(t_0) J(t_n,t_0) + \sum_{i=1}^{n-1} \Delta\sigma(t_i) J(t_n,t_i) \tag{1}$$

where the basic creep is modelled by the integral kernel in the form of a Dirichlet series, Bazant [1988]. This corresponds to Kelvin-Voigt elements placed in a series and can be easily coupled with the material deterioration parameter; see for ex. Torrenti et al. [2008]. The time discretization is performed using the implicit Euler scheme:

$$\begin{aligned} \varepsilon(t_{i+1}) &= \varepsilon(t_i) + \frac{\sigma(t_{i+1})}{E(t_{i+1})} - \frac{\sigma(t_i)}{E(t_i)} + \\ &+ \sum_{k=1}^{r} \left\{ [f_k(t_{i+1}) - f_k(t_i)] \sum_{j=0}^{i-1} \int_{t_j}^{t_{j+1}} \sigma(\tau) g_k(\tau) d\tau + f_k(t_{i+1}) \int_{t_i}^{t_{i+1}} \sigma(\tau) g_k(\tau) d\tau \right\} \end{aligned} \tag{2}$$

where $t_{i+1} - t_i$ stands for the actual time step. Denoting $\mathbf{F} = \{f_1, ..., f_r\}$ $\mathbf{G} = \{g_1, ..., g_r\}$, $\mathbf{Y} = \{Y^{(1)}, ..., Y^{(r)}\}$, $Y^{(i)} \equiv Y^{(t_i)}$ we introduce an internal variable of state in the form of recursive definition: $Y_0^{(k)} = 0$, $Y_i^{(k)} = Y_{i-1}^{(k)} + \int_{t_i}^{t_{i+1}} \sigma(\tau) g_k(\tau) d\tau$.

Such discretization enables us to change the integral equation into a set of algebraic equations and all that is therefore required is storing the value of the internal variable in each point in each previous calculation step:

$$\varepsilon(t_{i+1}) = \varepsilon(t_i) + \frac{\sigma(t_{i+1})}{E(t_{i+1})} - \frac{\sigma(t_i)}{E(t_i)} + [\mathbf{F}(t_{i+1}) - \mathbf{F}(t_i)] \mathbf{Y}_{i-1}^T + \mathbf{F}(t_{i+1}) \int_{t_i}^{t_{i+1}} \sigma(\tau) \mathbf{G}^T(\tau) d\tau \tag{3}$$

Assuming electrical neutrality, the transport equations based on assumption of a Fickian process of diffusion within liquid phase in isothermal conditions, can be written in the form [Glasser et al. 2008]:

$$\frac{\partial c_i}{\partial t} - \mathrm{div}(D_i \mathbf{grad}(c_i) - c_i \mathbf{v}) + r_i = 0, \quad i = 1, 2, ... \tag{4}$$

where successive terms in this set of equations stand for concentration rate, concentration diffusion, convection flux and sources, respectively. The last terms represent mass change due to chemical reactions. It can be proved from Onsager's relations [Zaborski 2010], that the form expressing velocities in the convection terms by:

$$\mathbf{v} = -D_w \mathbf{grad}(w) \tag{5}$$

is equivalent to cross-effects of flows included. The diffusion coefficients, D_i, depend on saturation, temperature, concentration, chemical reaction constant as well as strain and it is usually expressed in a "curve-fitting" form [Saetta et al. 1998]:

$$D_i = D_{i0} \cdot F_1(h) \cdot F_2(T) \cdot F_3(c_i) \cdot F_4(K) \cdot F_5(\varepsilon). \tag{6}$$

The terms representing temperature and chemical reaction constant are neglected, the former because of isothermal states assumed and the latter due to lack of precise experimental data. The impact of stress and strain state is limited to dilatational strains, $F_5(\varepsilon) = 1 + k_{rc}\mathrm{tr}(\varepsilon)$, basing on experimental results of Schneider and Chen [2005].

To complete the formulation of the problem the evolution equations of internal variables, for the reaction extent constant and the damage parameter, have to be specified. In the Clausius-Duhem inequality for viscoelastic continua, $\boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}} - \dot{\psi}(\boldsymbol{\varepsilon}, \mathbf{q}_{\boldsymbol{\alpha}}) \geq 0$, the second term represents the derivative of Helmholtz free energy and $\mathbf{q}_{\boldsymbol{\alpha}}$ is a set of internal variables. Supposing linearity between the thermodynamic forces and variables from the principle of dissipation energy maximum one can formulate a nonlinear optimization problem with Kuhn-Tucker constraints in the form of evolution equations. Considering a first order reaction kinetics, the following form for the reaction rate is proposed [Comi et al. 2012]:

$$\dot{\xi} = \frac{(\xi_c - \xi)}{\tilde{t}}, \quad \xi_c = \frac{1 + b_1 \exp(-b_2)}{1 + b_1 \exp(-b_2 c_l)} \tag{7}$$

where b_1 and b_2 are the material parameters to be calibrated on the base of experimental data and c_l stands for the saturation degree.

The effective stress, due to material degradation is [Kattan and Voyidjis 2001]:

$$\overline{\sigma} = \frac{\sigma}{(1 - d_{ch})(1 - d_m)} \tag{8}$$

where scalar damage parameter was split into a chemical and a mechanical part, d_{ch} and d_m, respectively.

The chemical damage parameter is simply related to the chemical reaction constant by:

$$d_{ch} = \zeta K, \quad \zeta \in (0,1) \tag{9}$$

where ζ is an empirically determined parameter. The hypothesis allows one to obtain a good fitting with the existing experimental data and may be considered satisfactory until additional experimental results are available. The evolution equation for mechanical damage parameter is assumed in the form proposed by Kuhl et al. [2004]:

$$d_m = 1 - \frac{\eta_0}{\eta}[1 - \alpha_m + \alpha_m \exp[\beta_m(\eta_0 - \eta)]], \quad \eta(\varepsilon) = \sqrt{\frac{1}{E_0}\boldsymbol{\varepsilon} : \mathbf{C}_0 : \boldsymbol{\varepsilon}} \tag{10}$$

where the parameters α_m and β_m are determined from experiments and $\eta(\varepsilon)$ is an equivalent strain measure with a threshold value η_0.

NUMERICAL DISCRETIZATION

Numerical solution to the transport equations by Alternating-Direction Implicit (ADI) method consists of splitting differential operators for each direction of differentiation, obtaining tridiagonal matrix requiring significantly less computational effort Zaborski [1995]. The splitting however leads to expanded implicit/explicit schemes and their implementation in computer program is rather cumbersome. Moreover the method has no direct generalization for 3D problems.

These drawbacks may be avoided using somewhat simpler type of modeling like cellular automata. Introduced by von Neumann and Ulam for self-replication problems on the Turing's machine, cellular automata were applied to diffusion problems from the beginning. In the mechanics of continua however, the method is still considered as novel approach. Main idea of cellular automata consists of regular uniform grid of cells. Internal state of each cell is given by so-called automaton rule exploiting the state of the cell itself and its neighborhood. The automaton rule is identical for all cells on each time step, except the boundary cells treated differently. Biondini et al. [2004] successfully adapted cellular automata approach to diffusive attack from external aggressive agents on concrete structures through a proper selection of von Neumann neighborhood with radius $r = 1$:

$$c_i^{k+1} = \Phi_0 c_i^k + \sum_{j=1}^{d}\left(\Phi_j^- c_{i-1,j}^k + \Phi_j^+ c_{i+1,j}^k\right) \tag{11}$$

where c_i^k represents the concentration of component in the cell i at time t_k and Φ_s are evolutionary coefficients. The solution proposed by the authors, though generalized to stochastic processes and material anisotropy, has an important drawback. For adopted grid dimension Δx and time step Δt the diffusion coefficient D is constant in the whole area:

$$D = \Phi_1 \frac{\Delta x^2}{\Delta t} \tag{12}$$

that may be considered as rough approximation only. This drawback can be eliminated by proper time scaling [Zaborski 2010]; the generalized formulae handle the case of variable diffusion coefficients:

$$\frac{4D\Delta t}{\Delta x^2} \equiv \alpha \;\rightarrow\; \bar{c}_{m,n} = c_{m,n}(1-\alpha) + \frac{\alpha}{4}\left(c_{m+1,n} + c_{m,n+1} + c_{m-1,n} + c_{m,n-1}\right) \tag{13}$$

Substituting the above formulae into the transport equations one can derive the automaton rule for diffusion with cross effects for von Neumann's neighborhood:

$$\begin{aligned} c_{m,n}^{\alpha(1)} &= c_{m,n}^{\alpha(0)}\left(1-a^{\alpha}\right) + \frac{a^{\alpha}}{4}\left(c_{m+1,n}^{\alpha(0)} + c_{m,n+1}^{\alpha(0)} + c_{m-1,n}^{\alpha(0)} + c_{m,n-1}^{\alpha(0)}\right) - \\ &- \sum_{\beta\neq\alpha} a^{\alpha\beta} c_{m,n}^{\beta} + \sum_{\beta\neq\alpha} \frac{a^{\alpha\beta}}{4}\left(c_{m+1,n}^{\beta(0)} + c_{m,n+1}^{\beta(0)} + c_{m-1,n}^{\beta(0)} + c_{m,n-1}^{\beta(0)}\right) \\ a^{\alpha\beta} &= \frac{4D_{\alpha\beta}\Delta t}{h^2} \end{aligned} \tag{14}$$

as well as for Moore's neighborhood:

$$\begin{aligned} c_{m,n}^{\alpha(1)} &= c_{m,n}^{\alpha(0)}\left(1-a^{\alpha}\right) + \frac{a^{\alpha}}{5}\left[\left(c_{m+1,n}^{\alpha(0)} + c_{m,n+1}^{\alpha(0)} + c_{m-1,n}^{\alpha(0)} + c_{m,n-1}^{\alpha(0)}\right) + \right. \\ &\left. + \frac{1}{4}\left(c_{m+1,n+1}^{\alpha(0)} + c_{m+1,n-1}^{\alpha(0)} + c_{m-1,n+1}^{\alpha(0)} + c_{m-1,n-1}^{\alpha(0)}\right)\right] - \sum_{\beta\neq\alpha} a^{\alpha\beta} c_{m,n}^{\beta} + \\ &+ \sum_{\beta\neq\alpha} \frac{a^{\alpha\beta}}{5}\left[\left(c_{m+1,n}^{\beta(0)} + c_{m,n+1}^{\beta(0)} + c_{m-1,n}^{\beta(0)} + c_{m,n-1}^{\beta(0)}\right) + \frac{1}{4}\left(c_{m+1,n+1}^{\beta(0)} + c_{m+1,n-1}^{\beta(0)} + c_{m-1,n+1}^{\beta(0)} + c_{m-1,n-1}^{\beta(0)}\right)\right] \\ a^{\alpha\beta} &= \frac{20D_{\alpha\beta}\Delta t}{h^2} \end{aligned} \tag{15}$$

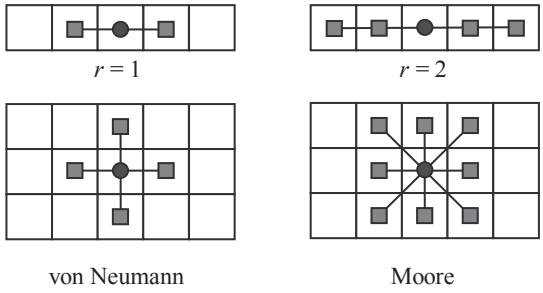

Fig. 1. Neighborhoods of von Neumann (left) and Moore (right)
Rys. 1. Sąsiedztwo Neumanna (z lewej) i Moore'a (z prawej)

Numerical simulations performed by Zaborski [2012] showed that the differences between calculation results for both types of neighborhood are less than 0.07% at each time step but have a tendency to accumulate with time.

NUMERICAL RESULTS AND CONCLUSIONS

The effectiveness of proposed methodology is demonstrated for specific applications. A reinforced concrete cross section is considered exposed to environmental ammonium nitrate solution and flexure stress. By solving transport equations the agent concentration contour lines were obtained. The contour lines calculated with the source term are present for comparison in the same figure below. The reaction products are calcium nitrate and ammonia that indicate deterioration of material. The contour lines of accumulative damage parameter, presented in the figure, demonstrate lack of horizontal symmetry due to the mechanical load. From structural point of view, the question of paramount importance is how the damage of the cross-section influences its bearing capacity. The answer to this question can be assessed by numerical simulation of instantaneous overloading of the cross section up to the ultimate limit state. For instantaneous short-term loading, the mechanical model can be defined assuming a fixed value of the aging degree.

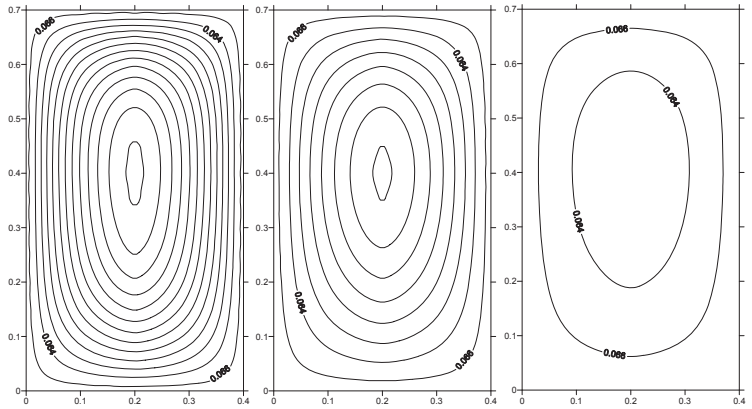

Fig. 2. Concentration contour lines without (left) and with (center) reaction included as well as damage contour lines (right)

Rys. 2. Koncentracja bez reakcji (z lewej) i z reakcją (w środku) oraz parametr uszkodzenia (z prawej)

Following Eurocode 2 recommendation of strain repartition the effective stresses were calculated from the theoretical curve of elastic-plastic state, proposed by Kuhl et al. [2004]. By integrating the stress over the cross section one can obtain the cross-sectional forces. The task is not as simple as it looks because strain softening of material under compression as well as under tension occurs. For this reason, extreme strain repartition does not always induce the extreme value of cross-section forces. This fact is often overlooked, resulting in non-concave interaction curves; see for ex. Papanikolaou [2012]. The remedy for this is to generate "sufficiently dense" set of cross section strains repar-

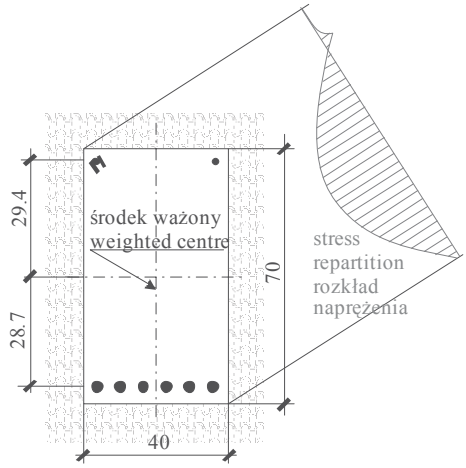

Fig. 3. Construction of interaction curves
Rys. 3. Konstrukcja krzywych interakcji

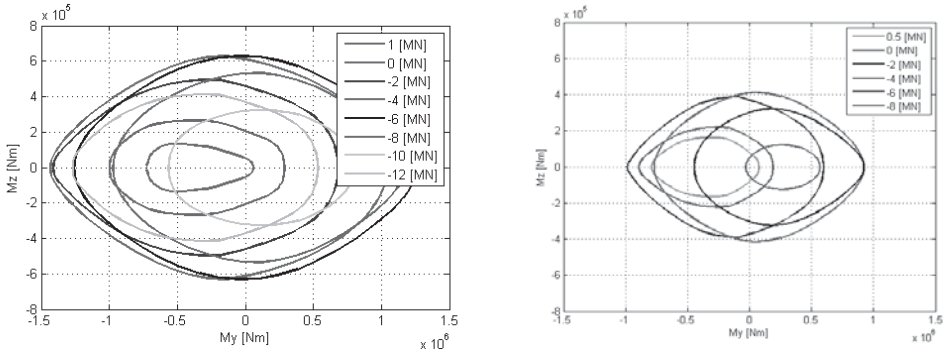

Fig. 4. Interaction curves: undamaged (left) and damaged (right)
Rys. 4. Krzywe interakcji dla przekroju nieuszkodzonego (z lewej) i uszkodzonego (z prawej)

titions and to construct a convex hull of obtained interaction points. For this purpose a Graham-Andrew algorithm from Computational Geometry Algorithms Library (Open Source Project) may be implemented. The section of interaction surfaces by plane zero axial force (biaxial bending) required use of iterative solution by bisection method, not the quickest, but stable for appropriate start points.

It is clearly visible from the presented interaction curves that the bearing capacity of the cross section considered decreases with time, leading to undesirable reduction of structure service life. The loading point, safely lying inside an interaction curve at the beginning, with time may end up at an unsafe position on the limit curve. The greatest decrease in bearing capacity is observed in the compressed zone what is the consequence of unilateral behavior of concrete. Due to lack of symmetry in two-sided reinforcement, the weighted centroid of the cross-section is displaced and the extreme values of bending moment are reached at non-zero values of axial force and vice versa.

The phenomenological model presented is a good representation of basic features of corrosive processes progressing in the RC cross section. Some material constants and process parameters need further, and more detailed, investigation.

The interaction curves presented demonstrate characteristics adequate to considered problem: solution symmetry and important contribution of compressed zone to the bearing capacity of cross section. The effect of scale is also noticeable, meaning that the degradation processes develop quicker in smaller elements. With time this is manifested by accelerated material deterioration despite decreasing concentration gradients. This proves the thesis of synergetic action of mechanical and chemical loading. Numerical simulations of these processes complement experimental research, extrapolating experimental data of laboratory samples to full-scale structural members.

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NUMERYCZNA SYMULACJA CHEMOMECHANICZNEJ DEGRADACJI BETONU I JEJ WPŁYWU NA NOŚNOŚĆ KONSTRUKCJI

Streszczenie. W niniejszej pracy analizuje się sprzężenie chemiczno-mechanicznego uszkodzenia materiału poprzez symulacje numeryczne procesów transportu i degradacji materiału. Transport substancji agresywnych opisany został równaniami dyfuzji z członem źródłowym określonym równaniem stechiometrycznym i stałą szybkości reakcji. Degradację betonu określa skalarny parametr uszkodzenia z równaniami ewolucji uszkodzenia chemicznego i mechanicznego. Do rozwiązania równań dyfuzji zastosowano automaty komórkowe, otrzymując warstwice koncentracji substancji i parametru zniszczenia. Nośność rezydualna przekroju została oszacowana na podstawie numerycznej symulacji testów niszczących, uzyskując obraz zmieniających się krzywych interakcji sił przekrojowych. Numeryczna symulacja potwierdziła wpływ synergicznego oddziaływania obciążenia mechanicznego i środowiskowego na degradację betonu oraz spadek nośności przekroju żelbetowego.

Słowa kluczowe: beton, korozja, kontynualna mechanika uszkodzeń, równania transportu, automaty komórkowe

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