INFLUENCE OF HYSTERETIC MOISTURE TRANSFER ON CONCRETE DURABILITY

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Abstract: Concrete has a crucial role in protecting reinforcement steel from corrosion. The high alkalinity of cement promotes the formation of a passive layer on the steel, protecting it against corrosion. Destabilization of this layer can happen because of the presence of chlorides, or because of a pH reduction due to carbonation of the cement matrix due to ingress of CO_2 from the atmosphere. Corroded steel reinforcement leads to a reduction of the mechanical properties and can induce accelerated fracture formation. As chloride ions are transported via liquid water, the water permeability plays an important role. Moreover as carbonation is determined by the transport of CO_2 in the remaining gas phase, the gas permeability also plays a major role. The water and gas permeability are directly depending on the saturation degree or moisture content. Concrete shows a strong hysteresis in sorption, and consequently, the permeability and the ingress of e.g. chlorides and CO_2 , - and thus the durability of concrete - will be highly influenced by the hysteretic moisture behavior. In this paper, we present a sorption hysteresis model for concrete based on the independent domain theory.

1 INTRODUCTION

The durability of reinforced concrete is crucial for civil and building infrastructures. The high alkalinity of the cement matrix promotes the formation of a passive layer on steel, protecting it against corrosion. Destabilization of this layer can happen because of the presence of chlorides, or because of a pH drop due to atmospheric carbonation [1]-[2]. Steel corrosion may then initiate and lead to concrete degradation. As chloride ions are transported via liquid water, the water permeability plays an important role. Moreover, as carbonation is determined by the transport of CO_2 in the remaining gas phase, the gas permeability also plays a major role. The water and gas permeability are directly depending on the saturation degree or moisture content. Concrete shows a strong hysteresis in sorption, and consequently, the permeability and the ingress of e.g. chlorides and CO_2 , - and thus the durability of concrete - will be highly influenced by the hysteretic moisture behavior.

hysteresis Moisture refers to the phenomenon that, at the same relative humidity, the material experiences a different degree of moisture saturation or level of moisture content depending on its loading history. The difference in moisture content at the same relative humidity for cement or concrete during repeated cycles of ad- and desorption under isothermal conditions is illustrated in amongst others De Belie et al. [3], Anderberg and Wadsö [4], Baroghel-Bouny [5] and Maruyama and Igarashi [6]. These hysteresis measurements count a limited number of ad/desorption loops, as the sorption process of concrete is slow. However, when a concrete structure is exposed to environmental conditions, it undergoes a large number of sorption cycles. Thus the ability to predict the hysteretic response of a concrete structure would be valuable especially with respect to its durability.

In this paper, we present a sorption hysteresis model for concrete based on the independent domain theory. The history of filling and emptying of sorption sites is efficiently monitored using an integrated PM formulation. This model is implemented in a finite element code for coupled heat and mass transfer. It allows assessing the influence of sorption hysteresis on the moisture behavior of concrete when exposed to changing temperature and relative humidity conditions. We compare simulations using the moisture hysteresis model with commonly performed simplified simulations only using the main ador desorption curve. It is found that sorption hysteresis has a high influence on the moisture behavior of the outside layer of concrete and as such CO₂ or chloride ingress are highly underestimated when using only ad- or desorption isotherms.

2 MOISTURE HYSTERESIS MODEL

Following the independent domain approach [7]-[11], a porous material contains a number of domains or sorption sites, which behave independently. Figure 1 describes the typical behavior of a sorption site. With increasing relative humidity, an initially dry sorption site becomes filled at a critical relative humidity ϕ_a resulting in a moisture content m_{f} . When the relative humidity decreases again, the site unfills at a lower relative humidity ϕ_d , resulting in a jump back to the unfilled state. Since $\phi_d \leq \phi_a$ hysteresis occurs between adsorption and desorption, ϕ_a and ϕ_d represent respectively the relative humidity in the adsorption $(_a)$ or desorption $(_d)$ regime. Adsorption includes both multilayer adsorption of water molecules on the pore wall surfaces and capillary condensation. Current physical explanations for the existence of hysteresis for a single sorption site are, for example, capillary condensation hysteresis, contact angle hysteresis and the ink-bottle effect due to entry pores with small diameter.



Figure 1. Characteristic behavior of a hysteretic sorption site. During increasing relative humidity a jump in moisture content occurs to m_f at a relative humdity ϕ_a . During decreasing relative humidity, unfilling occurs at ϕ_d , jumping back from m_f to the dry state.

A porous material consists of a number of sorption sites. Thus changes in moisture content are the outcome of the behavior of an assemblage of independent sorption sites. An efficient way to represent sorption sites is to map a site characterized by (ϕ_a, ϕ_d) , on the half ϕ_a - ϕ_d space, also called the PM space (Preisach-Mayergoyz [11],[12]). The PM space is represented in Figure 2. On the PM space, a frequency density distribution $\rho(\phi_a, \phi_d)$, called the PM distribution, represents the number of sorption sites. The PM space is

triangular and filling occurs in its lower surface, since $\phi_d \le \phi_a$.



Figure 2. The triangular PM space on which a density distribution function $\rho(\phi_a, \phi_d)$ is defined. The gray triangle represents the area over which integration is needed to obtain the function value $H(\text{RH}_F, \text{RH}_E)$ in the integrated PM space (see Figure 4).

Suppose the specimen is taken through a relative humidity (RH) history RH=0, RH_A, RH_B , RH_C (= RH_A), $RH_D \approx 1$, RH_E (= RH_A), RH_F (= RH_B), 0 (Figure 3a). The series of Figure 3b show the states of the sorption elements in the PM space for each RH (filled elements are gray). The resulting sorption curve is shown in Figure 3c. Initially the porous material is at RH=0, all sorption sites are unfilled and the moisture content is 0. When RH is increased towards RH_B, the sorption sites become progressively filled from left to right in the PM space, and the main adsorption curve is followed. As RH decreases from RH_B to RH_C the elements empty from top to bottom and a primary desorption curve is formed. We observe a different moisture content at RH_A and RH_C (=RH_A) resulting from the difference in filling $(A \rightarrow B)$ and unfilling of elements $(B\rightarrow C)$. Comparing the gray areas representing the filling in the PM space for state A and C, we see that the material is at a different filling state. When RH is further increased from RH_C, a loop is formed, which closes at RH_B. From then on, the main adsorption isotherm is again followed towards RH_D. When decreasing RH, the main desorption isotherm is followed, emptying elements from top to bottom. While increasing and decreasing again RH, a second loop is formed $(E \rightarrow F \rightarrow E)$, which is congruent with the first loop $(B \rightarrow C \rightarrow B)$ formed during the adsorption.



Figure 3. Typical snapshots during (a) a relative humidity loading history of (b) the PM space and (c) the moisture content M. ϕ_a is the relative humidity in adsorption, ϕ_d the relative humidity in desorption and Ω is the region of domains completely filled with water.

The region of filled sites is denoted Ω . After a certain RH loading history, Ω is typically bordered at the right side by a staircase line. Performing integration over the Ω domain gives the moisture content *M*:

$$M(\phi) = \iint_{\Omega} \rho(\phi_a, \phi_d) m(\phi_a, \phi_d) d\phi_a d\phi_d$$
(1)

To determine the integrals, the PM density $\rho(\phi_a, \phi_d)$ and the moisture function $m(\phi_a, \phi_d)$ have to be known. Instead of trying to directly determine these functions, the integrated PM (IPM) approach is adopted. In the IPM approach, an integral function *H* is defined as:

$$H(\phi_a, \phi_d) = \int_{\phi_d}^{\phi_a} \int_{y}^{\phi_a} \rho(x, y) m(x, y) \,\mathrm{d} x \mathrm{d} y \tag{2}$$

where for example $H(RH_E, RH_F)$ equals the integral over the triangle (RH_E, RH_E) , (RH_F, RH_E) , (RH_F, RH_F) (shown in Figure 2). By the introduction of the IPM function *H*, the integral *M* in equation (1) can be calculated as: $M(\phi) = H(\phi_{a1}, \phi_{d1}) - H(\phi_{a1}, \phi_{d2}) +$

$$H(\phi_{a2}, \phi_{d2}) - H(\phi_{a2}, \phi_{d3}) + \dots$$
(3)
= $\sum_{i=1}^{m} H(\phi_{ai}, \phi_{di}) - \sum_{i=1}^{n} H(\phi_{ai}, \phi_{di+1})$

with m and n the number of vertical respectively horizontal boundary segments characterizing the staircase border. Thus the moisture content M can be determined by summing and subtracting values of H. The IPM function H covers the triangle of the PM space, attains its maximum at the bottom and right borders, decreases towards the diagonal and becomes zero on the diagonal (Figure 4).



Figure 4. Representation of the function $H(\phi_a, \phi_d)$ in the IPM space. The IPM function is zero on the diagonal and attains maximal values at the boundaries.

3 IMPLEMENTATION

The IPM function $H(\phi_a, \phi_d)$ is determined based on experimental data of the moisture content M (equation (3)). An analytical expression is used where the parameter values are found by a best fitting procedure on the experimental data [13]. As the parameters are based on real ad- and desorption data, phenomena such as film forming and capillary condensation are implicitly incorporated.

The heat and mass transport equations are solved using the finite element method with capillary pressure and temperature as primary variables [13]. In order to obtain a mass and energy conservative system of equations, a mixed form of the capacitive terms is used, as described in Janssen et al. [14]. A Newton-Raphson iterative scheme is employed and an adaptive time stepping algorithm is embedded to optimize the efficiency of the numerical solver.

The material property dataset used for the simulations is composed for a common concrete based on material properties from different literature sources [13].

4 **RESULTS**

We present simulation results to exemplify the proposed model assuming a 30 cm thick concrete structure exposed on one side to (1) an environment of constant temperature, 23°C, and a sinusoidally varying relative humidity (between 90 and 30% RH) with a period of 1 year and (2) a real climate, i.e. the outdoor climate of Essen, Germany. An initial wet state of the concrete is considered.

Figures 5 shows the calculated response at different positions in the 30 cm thick concrete after being subjected for 40 days to the sinusoidal relative humidity fluctuations with a 1-year period, starting from an initial 'wet state' (90% RH, main desorption curve). The loops represent the response during the final calculated year (40th year). The main adsorption and main desorption curves are depicted in the figures as well. The moisture content, the corresponding vapor resistance factor and moisture capacity are shown as function of the relative humidity. Comparing the moisture content variations as determined by the hysteresis model with the main ad/desorption curves (Figure 5a), we observe an important difference for the outer concrete layers; deeper inside the concrete, the material approaches the main desorption curve. Fluctuations in the material properties occur over a depth of about 15 cm into the structure. Consequently the vapor resistance factor, thus moisture and gas permeability, which highly depend on the moisture saturation, differs significantly from the main ad/desorption values.

Figure 6 shows the moisture content, vapor resistance factor and moisture capacity at different positions in the 30 cm thick concrete structure after being exposed for 40 years to the outdoor climate in Essen, Germany. As concrete is initially in a wet state if sufficiently well cured on site, an initial 'wet state', equal to the initial outdoor conditions in Essen (91% RH, 0.5°C), is considered. When using the real climatic conditions of Essen, the climatic



Figure 5. Moisture content, vapor resistance factor and moisture capacity in function of relative humidity in a 30 cm thick concrete sample during the last year of 40 years exposure. The sample is initially in a 'wet state' (90% RH) and subjected to relative humidity fluctuations between 90 and 30% RH with a 1-year period.

variations penetrate less deep than the isothermal relative humidity fluctuations with a 1-year period as a climate comprises a superposition of daily and yearly variations.



Figure 6. Moisture content, vapor resistance factor and moisture capacity in function of relative humidity in a 30 cm thick concrete sample during the last year of 40 years exposure. The sample is initially in a 'wet state' (91% RH) and subjected to the outdoor climate of Essen, Germany.

The penetration depth is around 10 cm. Again a significant difference is observed between simulating with the main sorption curves or the hysteresis model. The moisture behavior is characterized by a cloud of values and do not form a single loop as in the isothermal simulations. The deeper inside the concrete material, the thinner the cloud gets. Remark that the simulation results using the main ador desorption curve also form a thin cloud and not a single line: due to the temperature fluctuations, different moisture contents can be found at the same relative humidity.

5 CONCLUSIONS

Degradation of concrete occurs in the outer concrete layer. It is in this same outer layer that hysteresis plays its major role and influences the permeability of CO₂ and chlorides. The moisture content, or saturation degree, is a good parameter to evaluate the risk for carbonation or chloride ingress, as the water permeability is directly related to the moisture content. With increasing saturation degree, the water permeability will increase and consequently gas permeability will decrease since more pores are filled with water. Thus the diffusion of CO₂ will be underestimated due to an overestimation of the saturation degree and consequently the risk for carbonation will not be predicted correctly. This means that if one would use the main desorption curve to simulate the behavior of an initial 'wet' concrete, the diffusion of CO₂ is underestimated compared to using the hysteresis model. On the other hand, the liquid permeability will be underestimated when the saturation degree is underestimated and consequently the risk for ingress of chlorides is underestimated. Therefore if one would use the main adsorption curve instead of the hysteresis model to describe the behavior of an initial 'dry' concrete. the moisture permeability is underestimated and so is the corrosion risk due to chloride infiltration.

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