Numerical Determination of Ion Diffusivity in Bricks Based on Non-Stationary Diffusion Experiments

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Abstract

Ion concentration profiles in sandwich-brick specimens have been determined experimentally at different degrees of water saturation. Based on the experimental data, ion diffusion coefficients were determined by inverse analysis. The diffusion equation served as a basis for an optimization technique. It has been shown that the ion diffusivity in bricks decreases markedly as the moisture content decreases. The interface between two adjacent bricks slows down the ion migration at low water content in particular. It is planed to investigate the interface between mortar and bricks in further experimental and numerical studies.

Keywords: Ion diffusivity, brick, interface, water content, inverse analysis

Rechnerische Bestimmung der Ionenbeweglichkeit in Ziegelstein unter Zuhilfenahme von Ergebnissen aus nichtstationären Diffusionsversuchen

Zusammenfassung

An zusammengesetzten Elementen aus Ziegelproben unterschiedlicher Wassersättigungsgrade wurden Profile der Ionenkonzentration experimentell bestimmt. Mit Hilfe der inversen Analyse wurden die Ionendiffusionskoeffizienten in feuchtem Ziegelstein unter Verwendung der experimentell bestimmten Daten ermittelt.

Der Auswertung wurde die nichtlineare Diffusionstheorie zugrunde gelegt. Es konnte nachgewiesen werden, daß der Diffusionskoeffizient empfindlich vom Wassersättigungsgrad abhängt. Er sinkt rasch mit abnehmendem Wassergehalt. Die Grenzfläche zwischen zwei aneinander grenzenden Steinen wirkt insbesondere bei niedrigem Feuchtigkeitsgehalt als deutliche Diffusionsbremse. Es ist vorgesehen, bei weiteren Untersuchungen die Grenzfläche zwischen Mörtel und Ziegelstein näher zu analysieren.

Stichwörter: *Ionendiffusion, Ziegelstein, Grenzfläche, Wassergehalt, inverse Analyse*

1   Introduction

Water and water movement are at the origin of numerous types of damage observed in masonry. Water movement in most porous building materials can be numerically simulated in a realistic way. A quantitative description of salt migration, however, is much more complex [1]. Salt migration can be subdivided at least into two processes based totally on different mechanisms. Dissolved ions can be transported with the migrating water. But in micro-porous materials such as concrete a strong filter effect has to be taken into consideration. The second transport mechanism is ion concentration gradient driven diffusion. This mechanism is used in practice to remove dissolved ions from salt containing masonry by application of a compress.

In this contribution the ion diffusivity in brick masonry shall be determined. Based on experimental results, a numerical model is validated. The ion diffusion coefficient can then be calculated by inverse analysis. The influence of water content on the observed diffusivity has been studied in particular.

Results provide us with a solid basis for the prediction of time-dependent redistribution of ions in moist masonry. Results may also help to optimize desalination by the compress method.

2   Preparation of samples

In order to measure the diffusivity of ions such as Na^+ in bricks at different moisture levels, sandwich-brick specimens have been prepared. Usual masonry brick with a pore volume of 34% determined by mercury high pressure porosimetry were used. Cylindrical specimens were drilled out parallel to the extruding direction and cut into discs. These layered specimens consist of three brick plates composed as shown in Figure 1. The central brick layer was impregnated by a salt solution (Na_2SO_4). The three brick plates are equilibrated with the same moisture con-
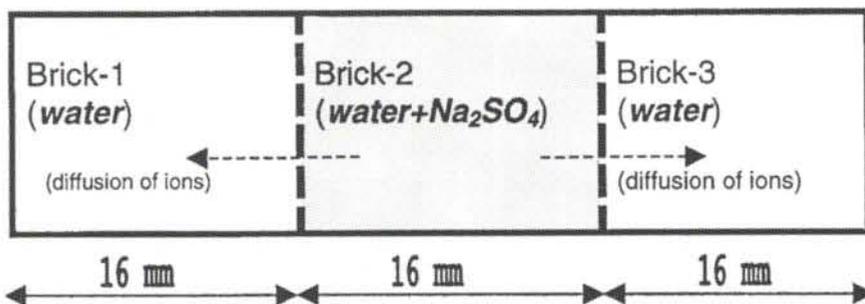

Figure 1:   Schematic representation of the three layers of the test block

tent. Four different moisture contents have been chosen, i.e. 25, 55, 78 and 100% of the maximum moisture capacity of the brick, W_{max}. Paper filters have been inserted between two adjacent plates in order to ensure a better contact at the interfacial zones. This sandwich construction is mechanically compressed in order to enhance the contact between the three layers. Immediately all the faces of the sandwich specimens are sealed in order to prevent any moisture exchange between specimens and the surrounding atmosphere.

Both free sodium and sulfate ions will then gradually migrate towards the adjacent untreated layers, untill the entire composite system reaches its final ionic equilibrium. This ionic transfer is predominantly governed by diffusion. Diffusion indu-ced by ionic concentration gradients in the porewater. As all layers have the same moisture content, no moisture motion may take place. That means the ions cannot be transfered by an advection process. Initially some water movement may take place in the opposite direction of ion diffusion due to osmosis. It must be also outlined that, at the chosen moisture contents (25, 55, 78 and 100% of the maximum moisture capacity of the brick, W_{max}), water held in pores is mainly in liquid phase. A typical desorption isotherm of a clean brick is given in Figure 2 [2]. The equilibrium moisture content in salt containing bricks is much more higher.

At selected durations of diffusion, the three plates were disconnected and milled down step by step up to defined depths. The brick powder was used to analyse the water-soluble sodium content.
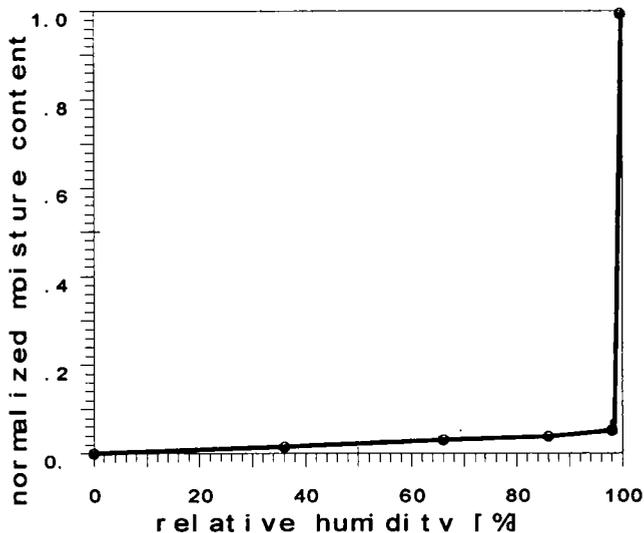

Figure 2: Typical desorption isotherm of an untreated brick at 20 °C [2]

3   Transport model

By assuming that no advection phenomenon and no chemical reaction can take place during the test, the transient transfer of the ions held in porewater of the porous system can be described by a diffusion equation given by the second Fick's law which is mathematically expressed by the following partial differential equation :

$$\frac{\partial C}{\partial t} = \text{div}(D(C) \cdot \text{grad } C) \tag{1}$$

in which t is the diffusion time and C is the ion concentration

$D(C)$ is the ion diffusion coefficient. The diffusion coefficient $D(C)$ may depend on the actual ion concentration C. As it will be shown in the following numerical analyses, however, a constant ion diffusion coefficient can describe the diffusion process with sufficient accuracy for a given moisture content.

In accordance with the experiments, the following initial conditions are assumed:

$$C_{(x,\,t)\,=\,0} = C_0 \qquad \text{for x within the impregnated middle brick} \tag{2a}$$

$$C_{(x,\,t)\,=\,0} = 0 \qquad \text{for x within the left or right brick} \tag{2b}$$

No boundary conditions are fixed because the system is closed (no interaction with its surrounding).

4   Numerical analysis

In the numerical analysis, equation (1) is used in its integral formulation [3]. We can integrate equation (1) in space over a conveniently small finite volume V of the diffusion region and write:

$$\frac{\partial}{\partial t} \int_V C \cdot dv = \int_V div(D \cdot gradC) \cdot dv \tag{3}$$

By means of the divergence theorem, the right-hand term of equation (3) is converted to a surface integral, and assuming an average value C over the volume V, equation (3) can be written as follows:

$$V \cdot \frac{\partial C}{\partial t} = \int_\Gamma D \cdot gradC \cdot dA \tag{4}$$

In our case, the diffusion problem is simplified to a one-dimensional. The integrated finite difference method [3] is used to solve the problem. The diffusion domain is discretized into n small subdomains. The discretization is refined in the two

interfacial zones, this allows us to take into account eventually the presence of the filters, as it will be shown later.

For the *i*-th element, the exchange of ion species with its neighbouring elements (*i*-1) and (*i*+1), can be approximated by the mass balance as follows:

$$V_i \cdot \frac{\Delta C_i}{\Delta t} = D_{i-1,i} \cdot \frac{C_i - C_{i-1}}{x_i - x_{i-1}} \cdot A_{i-1,i} + D_{i,i+1} \cdot \frac{C_i - C_{i+1}}{x_i - x_{i+1}} A_{i,i+1} \tag{5}$$

In equation (5) $A_{l,k}$ is the area between adjacent elements l and k, and $D_{l,k}$ is the harmonic mean diffusion coefficient of the elements, this later term is constant in our case. In inhomogeneous materials or if the diffusivity depends on the concentration this assumption is not valid any more.

The set of equations (5) can then be solved numerically by an adequate method for the unknown C_i.

5   Determination of the ion diffusivity

Ion diffusivity is a priori the unknown material parameter. The measured ion concentration profiles will be used in order to find the best ion diffusion coefficient. This can be realized by minimizing the functional quadratic error $E(D_b, D_f)$:

$$E(D_b, D_f) = \sum_i \left[C_{cal.}(x_i, \tau, D_b, D_f) - C_{exp.}(x_i, \tau) \right]^2 \tag{6}$$

In this functional, $C_{cal.}$ and $C_{exp.}$ are the calculated and the measured ion concentration respectively at position x_i and at diffusion time τ. D_b and D_f are the ion diffusivities of the brick and the interface respectively. As it will be seen in the next section, the interfacial zone has not to be modelled specifically if the bricks are moisture saturated because the contact between adjacent brick layers is ideal with regard to the ion transfer. On the contrary, at lower moisture content, the interface with the inserted paper filter slows down the ion diffusion process.

6   Results

6.1   Ion concentration profiles

Figure 3 shows the spatial distribution of the sodium ion concentration in the sandwich brick specimen, represented as relative ion content per dry mass of brick. Because of complete symmetry of the system one half is shown only. In this case, the specimen is moisture saturated ($W = W_{max}$). The dashed line represents the ion concentration at the beginning of the diffusion process. The measured ion concentration at different positions, as indicated as distance from the symmetry axis of the
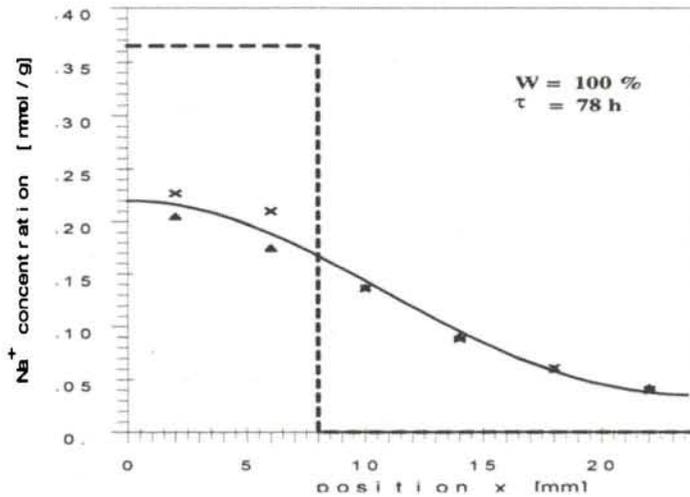
Figure 3: Measured (symbols) and simulated (solid line) Na^+-concentration profiles after a diffusion time of 78 h if the system is moisture saturated ($W = W_{max}$). The dashed line represents the ion concentration profile at the beginning.

specimen after a diffusion time of 78 h is given in Figure 3 by two symbols (x stands for one side of the specimen and Δ for the other). The continuous line represents the simulated profile for the fitted ion coefficient of diffusion as obtained by the above described optimization technique (equations 4 and 6).

In the case of Figure 4, the measured and simulated ion concentration profiles are plotted for a diffusion duration of 77 h and if the moisture content of the specimen is 78 % of the maximum moisture capacity of the brick ($W = W_{max}$). As it can be seen from Figure 4, the ion concentration gradient at the interface between the initially Na_2SO_4-impregnated brick (the centre) and its adjacent brick plates (initially free from Na_2SO_4) is steeper as in the rest of the specimen. This effect can be explained by the action of the interface which plays a role of an additional resistance with respect to ion migration. Consequently the ion transfer in the interfacial zones is slowed down. As has been mentioned above already, this effect is taken into account in the numerical model by introducing a thin layer at the interfacial regions. This layer is characterized by its thickness and its own ion diffusion coefficient (D_f). The simulated ion concentration profile shown in Figure 4 by a solid line represents the best fit of diffusion equation (4) and equation (6) for the diffusivity of the brick (D_b) and the diffusivity of the interface (D_f).

Figure 5 shows the experimental and simulated ion concentration profiles after a duration of diffusion of 51 h and if the moisture content is 55% of W_{max}. Here again, the interface is taken into consideration in the modelling. In this case the resi-
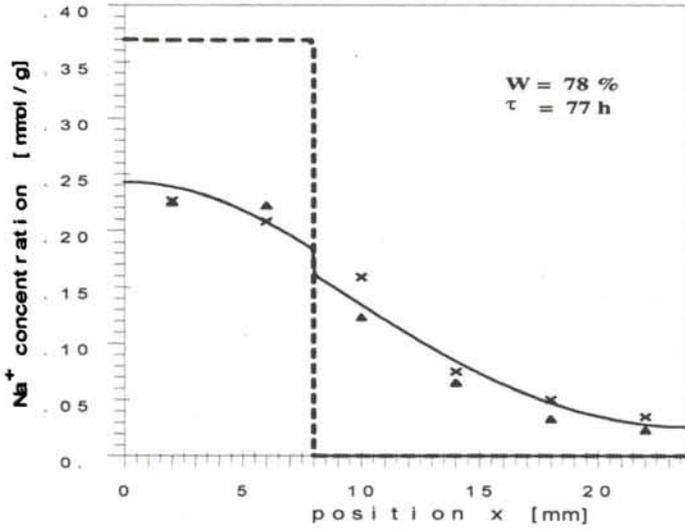
Figure 4: Measured (symbols) and simulated (solid line) Na^+-concentration profiles after a diffusion time of 77 h and if the moisture content is $W = 78\% \cdot W_{max}$. The dashed line represents the ion concentration profile at the beginning.
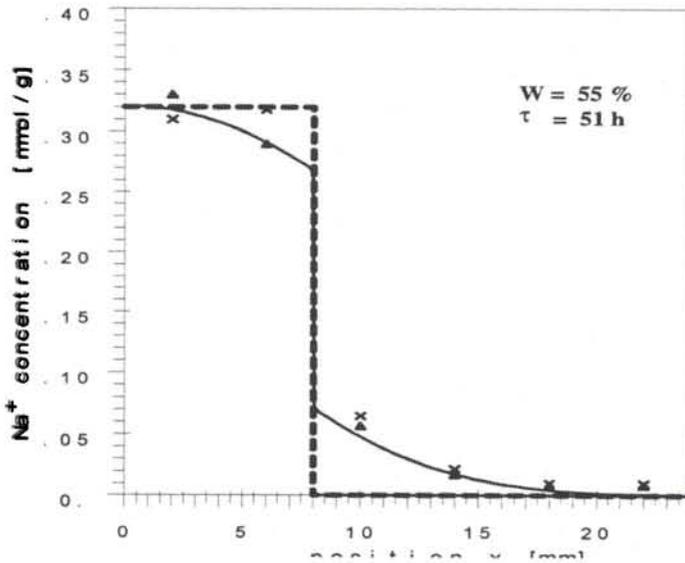

Figure 5: Experimental and simulated ion concentration profiles after a diffusion time of 51 h and if the moisture content is 55% of W_{max}.

stance of the interface with respect to the ion diffusion is much more pronounced as in the case shown in Figure 4.

In Figure 6 the diffusion after 146 h at a water saturation of $W = 25\% \cdot W_{max}$ is shown. By comparing this case with the case shown in Figure 5, the resistance of the interface with respect to the ion migration seems to be less pronounced. This can probably be explained by the influence of the adaptation of moisture between brick and interfacial paper. In the case shown in Figure 6, the exposure time is nearly three times longer than in the case shown in Figure 5, the interface may gradually lose its resistance when exposed to ions for a long time.

6.2   Ion diffusion coefficient

The optimal ion diffusivities obtained by combining the measured ion concentration profiles with the diffusion equation by means of an inverse analysis are given in the Table 1.

As can be expected the ion diffusivity decreases significantly with the degree of water saturation. If the brick is moisture saturated the ion diffusivity is roughly 7.5 times lower than the diffusivity in pure water. It should be mentioned that the calculated values of diffusivity of the moisture saturated brick are in agreement with results obtained with different experimental arrangement [5]. These values reflect
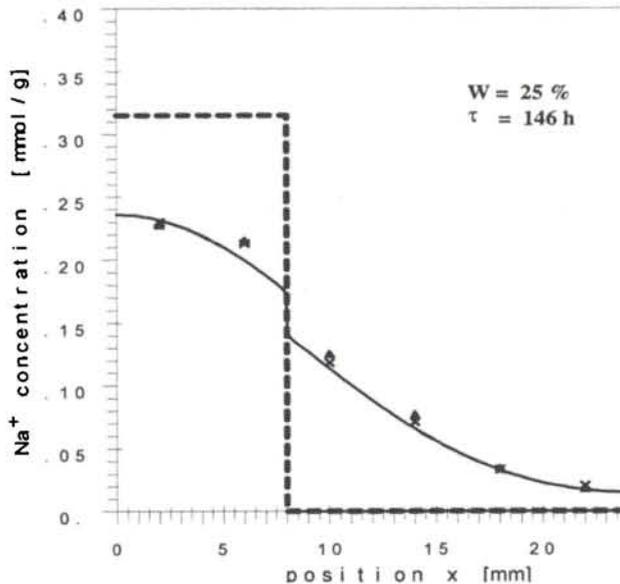

Figure 6:   Measured (symbols) and simulated (solid line) Na+ concentration profiles after a diffusion time of 146 h and if the moisture content is   . The dashed line represents the ion concentration profile at the beginning.

Table 1: Ion diffusivity of the brick and the interface between brick plates at different Ide-grees of water saturation and at different durations of diffusion. For comparison the value in bulk water for infinite concentration [4] is given

Degree of water saturation [%]S	Duration of diffusion [h]	Ion diffusivity [10^{-6} cm^2/s]	
		Brick	Interface
100	78	1,67	1,67
78	77	1,54	0,07
55	51	1,02	0,00
25	146	0,70	0,02
Pure water	-	12,5	

the influence of ion interaction in the pore solution and of the tortuous pore system of the brick material. The role of the interface between the brick samples can be neglected in the moisture saturated state but it becomes important at lower water content.

In further experimental studies, the ion concentration must be measured more accurately in the vinicity of the transitional zone in order to get more detailed information on the shape of the concentration gradient in this critical region. In addition, the interface between mortar and brick shall be investigated in the future.

7   Conclusions

By means of a special sandwich-brick specimen, ion concentration profiles can be measured for different degrees of water saturation.

Based on the experimentally determined ion concentration profiles and by applying an appropriate inverse analysis the ionic diffusivity in bricks can be determined.

The interface between two adjacent bricks has been modelled in detail. This transitional region is characterized by its thickness and its own ion diffusivity. At low moisture content, the interfacial zone has a high resistance with respect to ion migration.

The diffusivity of Na$^+$ ions in brick increases with the water content.

Acknowledgement

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