

MODELING COUPLED MOISTURE AND SALT TRANSPORT IN POROUS MATERIALS

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Abstract:

Basic models of coupled moisture and salt transport in porous materials are discussed from the point of view of their suitability to the solution of practical problems of salt damage. A modified diffusion-advection model is identified as the most suitable for this purpose which is illustrated on an example of its practical application.

Keywords:

Moisture transport, salt transport, porous materials, modeling

INTRODUCTION

Practically all deterioration mechanisms in porous materials are related to transport processes of heat, momentum, various phases of water, and chemical compounds. For example, a chemical substance from the surroundings contacts first the surface of a porous material, and concentrates in the surface region. Then, it can be dissolved in water and transported in the form of a solution into the bulk. The rate of penetration of this substance into the bulk depends on a variety of physical and chemical quantities, for instance on temperature, moisture content in the material, the level of stress in the material element, on the structure of the porous matrix and the level of its deterioration, etc. Also, the external conditions are very important, for instance the supply of the particular substance in the surroundings, temperature and moisture variations in the surroundings.

An accurate description of all these effects requires an application of a proper model of transport processes, because the mechanisms governing the heat, momentum, moisture and chemical compounds transport are coupled. In many cases, these mechanisms cannot be completely separated from each other and treated individually without a significant loss of accuracy. Also, the mathematical models of heat, momentum, water and chemical compounds transport contain a variety of storage and transport material parameters, which have to be determined in a wide range of field quantities, such as temperature, moisture content and concentration of the particular chemical compound.

A choice of a mathematical model for describing a specific problem of material degradation is always a compromise between the required physical-chemical suitability and the accuracy on one side, and the availability of material data for performing the calculations on the other. The models should be simple enough to allow obtaining the required data with a minimum of necessary measured transport and storage material parameters. On the other hand, application of oversimplified models could lead to serious errors, which might harm the service life prediction process and results in unrealistic service life estimates.

Water-soluble salts in the porous system can present a serious threat to the material deterioration. The harmful effects of such salts consist in the fact that after possible water evaporation salt crystals and crystal-hydrates are formed that often have crystallization pressures

higher than the strength of the particular porous material. The pressure exerted on pore walls can then lead to material destruction. However, the problem with salt ingress in porous building materials is not restricted only to the materials of load bearing structures. Renders can be affected as well. Salts can crystallize either in the porous system of renders or on their surface. Crystallization on the surface does not lead to damage of render material but salt efflorescence appears on the surface, which is a serious aesthetic flaw.

In this paper, basic models of coupled moisture and salt transport in porous materials are discussed from the point of view of their suitability to the solution of practical problems of salt damage.

BASIC MODELS OF MOISTURE AND SALT TRANSPORT

Most models applied in current research practice for the description of salt transport in porous materials are very simple. These employ Fick's diffusion equation for the salt concentration with a constant diffusion coefficient and constant initial and boundary conditions:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (1)$$

$$C(0, t) = C_0 \quad (2)$$

$$C(\infty, t) = 0 \quad (3)$$

$$C(x, 0) = 0, \quad (4)$$

where D is the salt diffusion coefficient, C the salt concentration, C_0 the salt concentration at the exposed boundary, x the distance from the exposed boundary, t the time.

The problem (1) - (4) has a very simple mathematical solution (e.g., Carslaw and Jaeger [1]):

$$C(x, t) = C_0 \left[1 - \operatorname{erf} \left(\frac{x}{2\sqrt{Dt}} \right) \right], \quad (5)$$

and this is the main reason for its high application frequency. This model was employed for instance by Tuutti [2], Funahashi [3], Cady and Weyers [4], Weyers [5], Zemajtis et al. [6], Costa and Appleton [7] and many others.

The model (1)-(4), which can be considered as the simplest choice ever in modeling salt transport contains several serious flaws that have to be taken into account in any practical application of it. The main problems with the model consist in the assumption of constant diffusion coefficient and in the fact that it neglects the influence of water transport on the transport of chemical compounds. As a consequence, a single value of the diffusion coefficient can never be obtained from the measured concentration profiles, particularly if the measurements are performed over longer time periods. The diffusion coefficient then appears as a function of time. Nevertheless, this model proved to be useful in many practical applications because the calculated diffusion coefficients have at least a relative value, i.e., it is possible to compare diffusion coefficients in different types of materials and different environments.

Another extreme in describing salt transport in porous materials is a consideration of fully coupled heat, moisture and chemical compounds transport phenomena. Among the most advanced models of this type, the model by Grunewald [8], [9] belongs to very promising (see e.g. the comments by Černý and Rovnaníková [10] for more details).

However, it should be pointed out that nor this model is an ideal solution. The main problem of the model [8], [9] consists in its complexity and particularly in the fact that it requires too many parameters to be determined in advance. The measurements of some of them are very time consuming so that for instance determination of liquid convection coefficients can take several months if static methods are used. In addition, all coefficients should be measured as functions of all state variables, which means at least a dependence on temperature, moisture content and salt concentration. Therefore, this model can be reasonably used only in the case that a database of necessary material parameters is already available.

A possible way out from the problems arising due to either oversimplification or too high complexity is to choose a reasonable compromise, i.e. a model that is somewhere in between these two extremes. Unfortunately, such models were only rarely used in the research practice until now. An example of application of a model of this type was introduced by Boddy et al. [11] where an attempt of including moisture transport in the salt transport equation was done. However, Boddy et al. [11] still assumed a linear salt diffusion problem with the diffusion coefficient as a function of time and temperature, and in addition, the problem of coupling moisture transport and salt transport was not solved at all.

A more successful attempt in this direction was done for the transport of NaCl solution in calcium silicate brick by Pel et al. [12] who used the Bear and Bachmat diffusion-advection model [13] in the form:

$$\frac{\partial(wC_f)}{\partial t} = \text{div}(wD \text{grad}C_f) - \text{div}(C_f \vec{v}) - \frac{\partial C_b}{\partial t} \quad (6)$$

$$\frac{\partial w}{\partial t} = -\text{div} \vec{v}, \quad (7)$$

where C_f is the concentration of free salt in water (in kg/m^3 of the solution), C_b the concentration of bonded salt in the whole porous body (in kg/m^3 of the sample), D the salt diffusion coefficient (in m^2/s), \vec{v} the Darcy's velocity (in m/s), w the volumetric moisture content (in m^3/m^3), and κ the moisture diffusivity (in m^2/s). Expressing Darcy's velocity in terms of moisture diffusivity,

$$\vec{v} = -\kappa \text{grad} w, \quad (8)$$

they arrived to a system of two parabolic partially coupled partial differential equations with three principal material parameters, namely D , κ and the ion binding isotherm $C_b = C_b(C_f)$. In a practical application of the model, Pel et al. [12] neglected the dispersion term in Eq. (6) so that they faced just the determination of κ and $C_b = C_b(C_f)$. This was done using NMR measurements of moisture content and Na^+ ions concentration. However, the agreement between the experimental and computational Na^+ ions concentration profiles was not very good. Therefore, either the model was too simple or possibly the choice or the way of determination of the necessary material parameters was not ideal.

DIFFUSION-ADVECTION MODEL OF MOISTURE AND SALT TRANSPORT INCLUDING SALT BONDING AND CRYSTALLIZATION

In this paper, the moisture and salt transport modeling treatment chosen by Pel et al. [12] is modified and extended in order to avoid the apparent mistakes in the original model. The basic idea that the mechanism of salt solution transport is described by Bear and Bachmat diffusion-advection model [13] taking into account not only the influence of moisture flow on salt transport but also the effect of salt bonding on pore walls is preserved. In addition to the effects taken into account in the

original model from [12], salt crystallization and water vapor transport are introduced into the model in order to facilitate modeling of drying experiments. Also, contrary to the model from [12], the dispersion effects in the liquid phase are considered as significant and included into the parameter identification process.

The modified mathematical model can then be written as follows:

$$\frac{\partial(wC_f)}{\partial t} H(C_{f,sat} - C_f) = \frac{\partial}{\partial x} (wD \frac{\partial C_f}{\partial x}) + \frac{\partial}{\partial x} (C_f \kappa \frac{\partial w}{\partial x}) - \frac{\partial C_b}{\partial t} - \frac{\partial C_c}{\partial t} \quad (9)$$

$$\frac{\partial w}{\partial t} = \frac{\partial}{\partial x} \left(\kappa \frac{\partial w}{\partial x} \right) + \frac{\partial}{\partial x} \left(\frac{\delta}{\rho_w} \frac{\partial p_v}{\partial x} \right) \quad (10)$$

$$\frac{\partial C_c}{\partial t} = \frac{\partial [w(C_f - C_{f,sat})]}{\partial t} H(C_f - C_{f,sat}) \quad (11)$$

where C_c is the amount of crystallized salt (in kg/m^3 of the sample), $C_{f,sat}$ is the saturated free salt concentration (in kg/m^3 of the solution), H is the Heaviside step unit function, $H(x \geq 0) = 1$, $H(x < 0) = 0$, δ the water vapor diffusion permeability (in s), p_v the partial pressure of water vapor (Pa), and ρ_w the density of water (in kg/m^3).

PARAMETER IDENTIFICATION OF THE DIFFUSION-ADVECTION MODEL OF MOISTURE AND SALT TRANSPORT

For the determination of moisture diffusivity as a function of moisture content and salt diffusion coefficient as a function of salt concentration, a salt solution suction experiment from a free salt solution level into a dry specimen is proposed. This makes it possible to neglect salt crystallization and water vapor transport so that the model is simplified as

$$\frac{\partial(wC_f)}{\partial t} = \frac{\partial}{\partial x} (wD \frac{\partial C_f}{\partial x}) + \frac{\partial}{\partial x} (C_f \kappa \frac{\partial w}{\partial x}) - \frac{\partial C_b}{\partial t} \quad (12)$$

$$\frac{\partial w}{\partial t} = \frac{\partial}{\partial x} \left(\kappa \frac{\partial w}{\partial x} \right). \quad (13)$$

The arrangement of the experiment is analogous to standard water suction experiments. The samples are first dried at 80°C and 0.1 mbar and water and vapor-proof insulated by epoxy resin on all lateral sides. Then, they are exposed by one of their faces to the penetrating salt solution. Duration of the experiment is chosen different for three different groups of samples. After this time, the samples are cut into several pieces and in each piece water content and salt concentration are measured. Moisture content is determined by the gravimetric method using weighing the moist and dried specimens so that the moisture profiles are obtained. In the determination of salt concentration, the particular samples are after drying first ground by a vibration mill so that grains smaller than 0.063 mm are obtained. Then the ground samples are overflowed by 80°C warm distilled water and leached. The salt contents in particular leaches are measured using the ion selective electrode. On the basis of measured ion binding isotherm, $C_b = C_b(C_f)$, the profiles of bound and free salts are determined.

The measured moisture and salt concentration profiles is then subject of inverse analysis, using Eqs. (12) and (13). The conditions of the experimental setup described above allow for a

significant simplification of the generally quite complicated inverse problem. The constant initial conditions and Dirichlet boundary conditions on both ends of the specimen for both moisture content and salt concentration where one of the Dirichlet boundary conditions is equal to the initial condition make it possible to consider the inverse problem as a semi-infinite one, and an extension of the standard Boltzmann-Matano treatment can be applied.

After applying Boltzmann transformation to equations (12), (13) and performing some straightforward algebraic operations in a similar way as with the solution of the inverse problem to one parabolic equation (see [10] for details), the following formulas for the determination of κ and D are obtained:

$$\kappa(z_0) = \frac{1}{2t_0 \left(\frac{dw}{dz} \right)_{z_0}} \int_{z_0}^{\infty} z \frac{dw}{dz} dz, \quad (14)$$

$$D(z_0) = - \frac{C_f(z_0) \kappa(z_0) \left(\frac{dw}{dz} \right)_{z_0}}{w(z_0) \left(\frac{dC_f}{dz} \right)_{z_0}} + \frac{1}{2t_0 \cdot w(z_0) \cdot \left(\frac{dC_f}{dz} \right)_{z_0}} \int_{z_0}^{\infty} z \left(\frac{d(wC_f)}{dz} + \frac{dC_b}{dC_f} \frac{dC_f}{dz} \right) dz, \quad (15)$$

where z is the space variable, t_0 is the time corresponding to the chosen moisture and salt concentration profiles $w = w(z, t_0)$, $C_f = C_f(z, t_0)$, the chosen values of moisture and salt concentration are $w_0 = w(z_0, t_0)$, $C_{f0} = C_f(z_0, t_0)$, and the corresponding moisture diffusivity and salt diffusion coefficient $\kappa(z_0) = \kappa(w_0, C_{f0})$, $D(z_0) = D(w_0, C_{f0})$.

In a practical calculation of κ and D , a set of z_0 values is chosen, $0 \leq z_0 \leq \infty$, the corresponding $[w_{0i}, \kappa(w_{0i}, C_{f0i})]$, $[C_{f0i}, D(w_{0i}, C_{f0i})]$ points calculated, and $\kappa(w)$ and $D(C_f)$ functions constructed.

VERIFICATION OF THE COMPUTATIONAL MODEL IN A WETTING EXPERIMENT

For the verification of the diffusion-advection model of coupled moisture and salt transport, 1M-NaCl solution suction into sandstone samples was chosen. The chloride binding isotherm of sandstone was measured first. The samples of sandstone with the dimensions of 20 x 40 x 160 mm were then subjected to 1-D chloride solution penetration and moisture profiles and chloride concentration profiles were determined for the suction times of 30, 60 and 90 minutes using the method described in the previous Section. Moisture diffusivity and chloride diffusion coefficient were then calculated using Eqs. (14), (15).

Fig. 1 shows the moisture diffusivity of sandstone as function of moisture content. We can see that obtained results correspond to the values typical for highly capillary active materials.

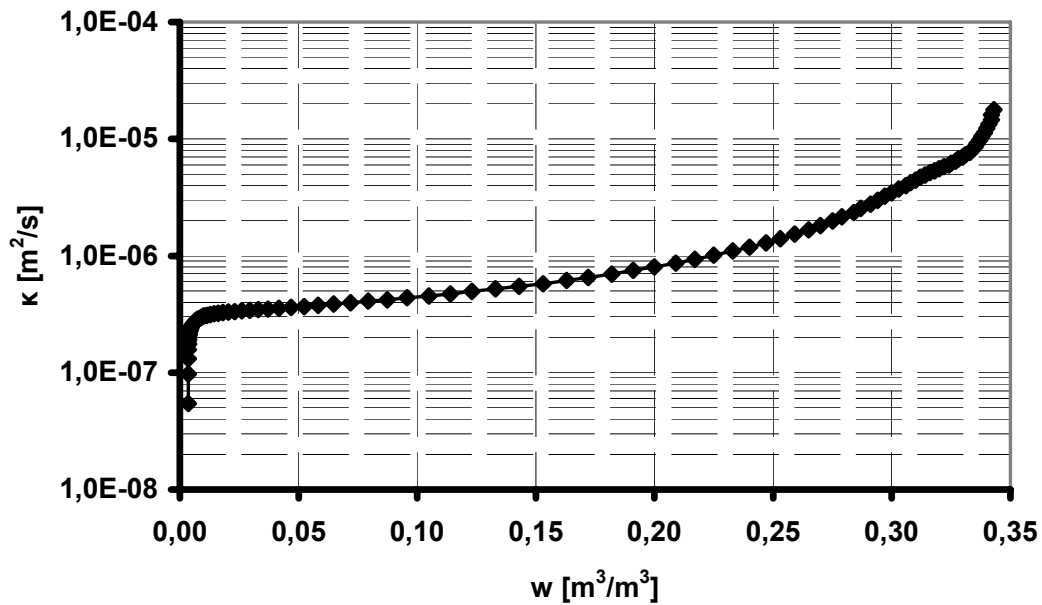


Fig. 1 Moisture diffusivity of sandstone

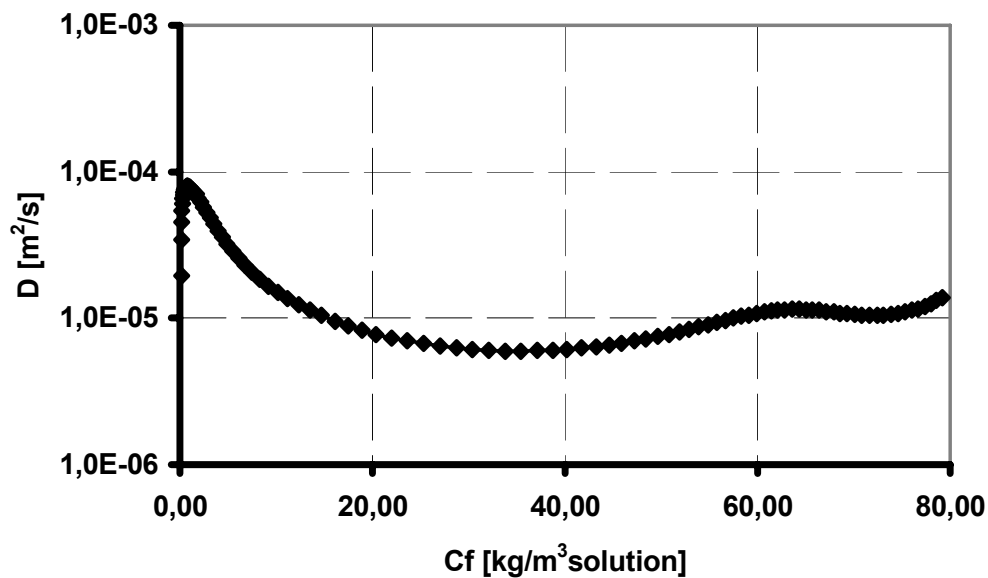


Fig. 2 Chloride diffusion coefficient of sandstone

Fig. 2 shows the chloride diffusion coefficient of sandstone in dependence on free ion concentration calculated using the $\kappa(w)$ function in Fig. 1. We can see that from the quantitative point of view, the calculated diffusion coefficient is quite high, about four orders of magnitude higher than the diffusion coefficients of most ions in free water. Therefore, the common diffusion mechanism was probably not the only driving force for the chloride transport within the liquid phase and some other driving forces were taking place here. The acceleration of chloride transport can be attributed most easily to dispersion effects in the liquid phase or to surface diffusion on pore walls.

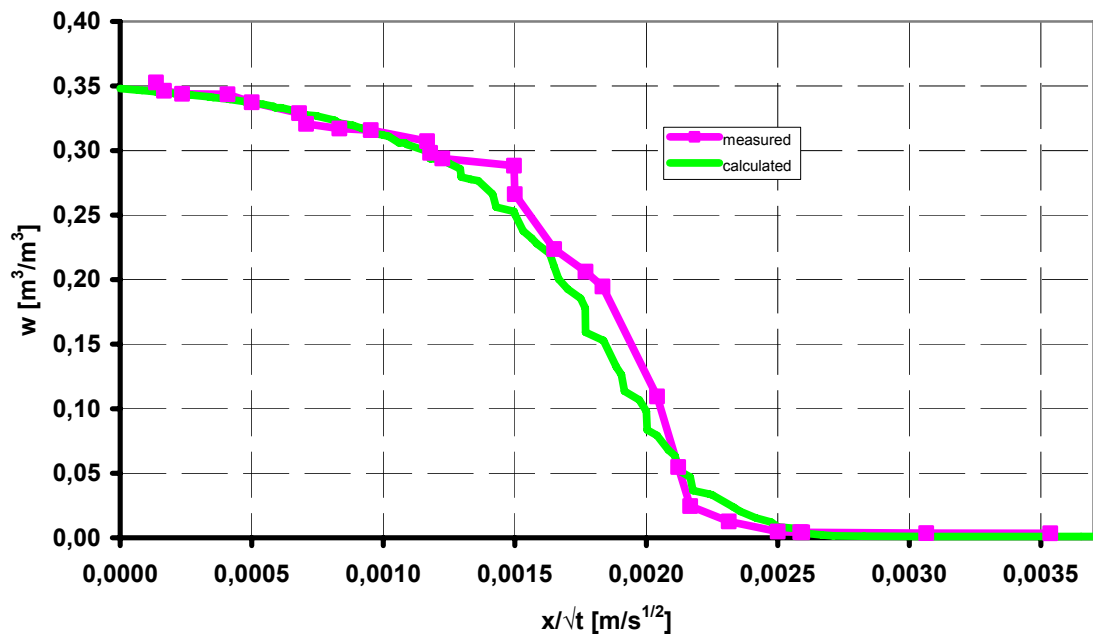


Fig. 3 Comparison of measured and calculated moisture profiles

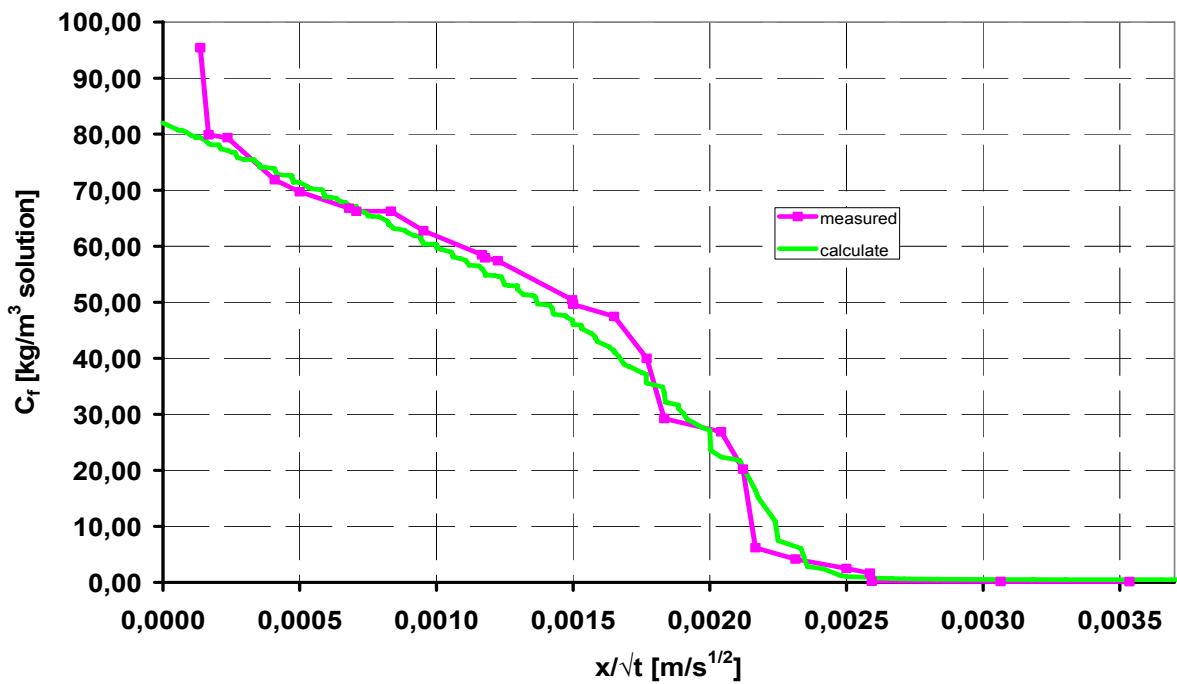


Fig. 4 Comparison of measured and calculated chloride concentration profiles

For the verification of the numerical stability of the parameter identification process of the applied coupled moisture and salt transport model, a forward analysis was performed. The moisture diffusivity and chloride diffusion coefficient given in Figs. 1, 2 were substituted into the computational implementation of the model (12), (13) and moisture and free chloride concentration profiles were calculated for the suction times of 30, 60 and 90 minutes exactly as in the experiments. The results in Figs. 3, 4 show that the calculated moisture and chloride concentration

profiles agreed reasonably well with the measured profiles, the differences were well within the error range of the measuring methods.

CONCLUSIONS

A modified diffusion-advection model was identified as the most suitable option for modeling coupled moisture and salt transport in porous materials at present. It can avoid both the oversimplifications such as the assumption of linear salt diffusion problem without water flow effect which application could result in significant distortion of physical reality and the too high complexities connected with an application of fully coupled heat, moisture, salt and momentum transport models which could face almost irresolvable problems with the accuracy of necessary input parameters.

The basic condition for the applicability of the diffusion-advection model in this paper in scientific and technical practice which is the stability of the parameter identification process was met very well. The computational implementation of the model was also found to be sufficiently accurate, which makes good prerequisites for the successful future practical applications of the model.

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