

DETERMINATION OF MOISTURE DIFFUSIVITY AND SALT DIFFUSION
COEFFICIENT OF BUILDING MATERIALS*

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Abstract. Basic models of coupled water and salt transport in building materials are presented in the paper. The methods for determination of water and salt transport and storage parameters are discussed from the point of view of the applicability of the parameters in the models. Finally, an example of determination of moisture diffusivity and salt diffusion coefficient from the measured moisture and salt concentration profiles is given.

Key words: coupled water and salt transport, moisture diffusivity, salt diffusion coefficient

INTRODUCTION

If salts are present in porous system of building materials in the form of solution, they are mostly not dangerous. 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 building material. The pressure exerted on pore walls can then lead to material destruction. Therefore, water-soluble salts are considered to belong to the most frequent causes of damage of both historical and contemporary building materials. However, the problem with salt ingress in building materials is not

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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.

Mathematical modelling of salt transport in building materials is one of the effective tools which can help to deal with the salt induced damage. To give only a couple of examples, the main reasons for the salt presence in building structures can be identified using computer simulations. The progress of salt accumulation in walls and renders can be predicted and the time of necessary repair estimated. The effect of different desalination methods can be simulated in advance and the application of the particular techniques optimised.

The effectiveness of the application of mathematical modelling in dealing with salt induced damage critically depends on the type of the model used. Salts can be transported in the porous system only in the presence of water. Therefore, modelling the salt ingress into porous materials in the form of coupled water and salt transport is indispensable. In some cases, the heat transport may also become important. On the other hand, a sufficiently accurate determination of water and salt transport and storage parameters is not an easy task because these can depend not only on the water content, salt concentration and temperature, but also on the history of salt presence in a material. As the accuracy of computer simulated data obtained by any model is primarily determined by the accuracy of its input parameters, a choice of a particular model should always be done in a direct link to the available material parameters.

In this paper, basic models of coupled water and salt transport in building materials are discussed from the point of view of their suitability to the solution of practical problems of salt-induced damage to building materials. The methods for determination of the necessary water and salt transport and storage parameters are discussed from the point of view of the applicability of the parameters in the models. Finally, an example of determination of moisture diffusivity and salt diffusion coefficient from the measured moisture and salt concentration profiles is given.

BASIC MODELS OF SALT TRANSPORT IN BUILDING MATERIALS

Most models applied in current research practice for the description of salt transport in building 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 [13], Funahashi [8], Cady and Weyers [3], Weyers [14], Zemajtis *et al.* [15], Costa and Appleton [7], and many others.

The model (1)-(4), which can be considered as the simplest choice ever in modelling 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 building materials is the consideration of fully coupled heat, moisture and chemical compounds transport phenomena. Among the most advanced models of this type, the model by Grunewald [9,10] belongs among very promising ones (see *e.g.* the comments by Černý and Rovnaníková [5] for more details).

However, it should be pointed out that this model is not an ideal solution, either. The main problem of the model [9,10] 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 the application of a model of this type was introduced by Boddy *et al.* [12], where an attempt of including water transport in the salt transport equation was made. However, Boddy *et al.* [12] still assumed a linear chloride diffusion problem with the diffusion coefficient as a function of time and temperature and, in addition, the problem of coupling water transport and salt transport was not solved at all.

A more successful attempt in this direction was made for the transport of NaCl solution in calcium silicate brick by Pel *et al.* [11] who used the well known Bear and Bachmat diffusion-advection model [1] in the form:

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

$$\frac{\partial w}{\partial t} = \text{div}(\kappa \text{ grad}w), \quad (7)$$

where C_f is the concentration of free salt in water, kg m^{-3} , C_b the concentration of bonded salt in the whole porous body, kg m^{-3} , D the salt diffusion coefficient, $\text{m}^2 \text{s}^{-1}$, \bar{v} the Darcy velocity, m/s , w the volumetric moisture content, $\text{m}^3 \text{m}^{-3}$, and κ the moisture diffusivity, $\text{m}^2 \text{s}^{-1}$. Expressing Darcy's velocity in terms of moisture diffusivity,

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

they arrived at 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.* [11] 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.

DETERMINATION OF MATERIAL PARAMETERS IN THE DIFFUSION-
ADVECTION MODEL OF COUPLED WATER AND SALT TRANSPORT

As it follows from the analysis in the previous section, the model by Bear and Bachmat [1] can be considered as a reasonable compromise between the complexity of the model and the necessity to determine its input parameters. Therefore, we will choose this model as the basic point for discussion.

The principal problem with the determination of material parameters within the framework of the model (6)-(7) consists in the fact that, in Eq. (6), there appear both D and κ . This leads to the necessity of solving together the inverse problems to both (6) and (7), in general. However, solving inverse problems of parabolic problems is not an easy task even in the case of one equation (see [5] for a more detailed analysis and a survey of methods). So, it is quite logical that most researchers try to avoid this problem.

One of the possibilities how to deal with the solution of the inverse problem for D and κ in coupled water and salt transport is to neglect D as it was done in [11]. However, this idea did not appear to be particularly useful even in the original paper.

A second possibility consists in using the concept of apparent transport parameters [12]. The main difference between the apparent parameters and the thermodynamically "pure" parameters of the coupled water and salt transport is that the apparent parameters do not express "pure" effects but combined effects. So, the apparent salt diffusion coefficients include not only the free salt diffusion in the porous space but also the effect of salts bonding on the pore walls and the effect of salt transport due to the water movement. The notion of apparent moisture diffusivity then means that it is related not to the water itself, but to the salt-in-water solution, *i.e.* the whole liquid phase.

Under these simplifying assumptions, the same parabolic differential equations and the same boundary and initial conditions for both water transport and salt transport are formally obtained, namely:

$$\frac{\partial C}{\partial t} = \text{div}(D(C) \text{grad } C) \quad (9)$$

$$C(0, t) = C_1 \quad (10)$$

$$C(\infty, t) = C_2 \quad (11)$$

$$C(x, 0) = C_2, \quad (12)$$

where C is either water concentration in kg of water per kg of the dry porous body or chloride concentration in kg of Cl^- per kg of the dry porous body, D is either the apparent moisture diffusivity or the apparent salt diffusion coefficient. Therefore, the calculation of concentration-dependent diffusion coefficients from the measured salt concentration profiles could be done using basically the same inverse

methods as those for the determination of moisture-dependent moisture diffusivity or temperature-dependent thermal conductivity (see [6]).

The main flaw of this approach is that the apparent parameters are in fact not any objectively defined physical quantities. So, the concept of apparent transport coefficients can be useful in the solution of a class of case studies only where the same initial and boundary conditions are valid. In other cases, it is useless.

In this paper, we propose a simple technique consisting in the identification of D and κ using two independent experiments.

A SIMPLE TECHNIQUE FOR DETERMINATION OF MOISTURE DIFFUSIVITY AND SALT DIFFUSION COEFFICIENT

We propose the following set of two simple experiments for the identification of moisture diffusivity and salt diffusion coefficient. In the first experiment, the moisture diffusivity κ is determined in the common way, using inverse analysis of moisture profiles measured during the penetration of distilled water into a dry sample. As a result, we obtain a $\kappa(w)$ function. In the second experiment, the salt solution of a chosen concentration penetrates into a water-saturated sample and the salt diffusion coefficient D is determined using inverse analysis of measured salt concentration profiles. In this way, we obtain a $D(C)$ function.

It should be noted that the proposed concept of D and κ identification does not present yet a general solution of the problem, and its application has some limitations. First, it can be applied for dilute solutions only because otherwise the moisture diffusivity would also be a function of salt concentration. Second, its application is limited to low-bonding salts for the particular material because the analysis does not include the ion-binding isotherm.

The first limitation is rather critical. For concentrated solutions it would not even be sufficient to measure moisture diffusivity as a function of the concentration of penetrating solution in a series of the above experiments. Such moisture diffusivities could not be successfully applied in any general coupled water and salt transport problems as the salt concentration is mainly a function of position.

On the other hand, the second limitation of the method would be relatively easy to cure. With the measured ion binding isotherm, $C_b = C_b(C_f)$, the salt diffusion coefficient in the second proposed experiment could be calculated using the solution of the inverse problem to Eq. (13),

$$\left(1 + \frac{1}{w_{sat}} \frac{\partial C_b}{\partial C_f}\right) \frac{\partial C_f}{\partial t} = \text{div}(D \text{ grad} C_f) \quad (13)$$

which is only slightly different from the solution of the problem (9)-(12).

EXAMPLE OF DETERMINATION OF MOISTURE DIFFUSIVITY
AND SALT DIFFUSION COEFFICIENT FROM MOISTURE
AND SALT CONCENTRATION PROFILES

In the experimental work, rod-shaped calcined gypsum samples with the dimensions of 20 x 40 x 290 mm were used for the determination of moisture profiles, and 40 x 40 x 190 mm for the determination of NaCl concentration profiles. Epoxy resin was employed for water and vapour proof insulation on the lateral sides to assure 1-D water and salt solution transport. In the case of NaCl solution penetration into water saturated gypsum samples, the outlying front sides of the samples were insulated as well to avoid evaporation.

In the determination of moisture profiles, the specimens were put in contact with water in vertical direction and moisture profiles along the samples were determined in 10 chosen time intervals using the capacitance moisture meter [12]. After the measurements, the samples were cut into 15 pieces and the moisture content was determined in each piece by the gravimetric method for the sake of calibration of the capacitance moisture sensor.

The measurements of salt concentration profiles were also performed in the arrangement of vertical suction experiment. 3% NaCl solution was used as the penetrating solution. Duration of the experiment was 1 hour. After this time, the samples were cut into 8 pieces and in each piece chloride concentration was measured.

For the determination of chloride concentration, the particular pieces of the samples were first ground in a vibration mill so that grains smaller than 0.063 mm were obtained. Then, 10 g of the ground sample was leached for 30 minutes in 180 ml of 80°C warm distilled water, and a magnetic stirrer was used to speed up the leaching process. The content of chlorides in the leach was determined by a turbidity method, *i.e.* titration of the volumetric solution of mercuric nitrate on sodium nitroprusside indicator. The first turbidity indicated the equivalence point. On the basis of consumption of mercuric nitrate, the content of water-soluble chlorides in the sample was determined.

Both experiments were performed in air-conditioned laboratory at $23 \pm 1^\circ\text{C}$ and $30 \pm 2\%$ of relative humidity.

On the basis of measured water and NaCl concentration profiles, the determination of moisture diffusivity κ in dependence on moisture content and determination of salt diffusion coefficient $D(C)$ in dependence on salt concentration was made using the Matano method. This method is based on the application of Boltzmann transformation. Details on the method can be found in [10].

Figures 1 and 2 show salt concentration profiles and moisture profiles in the Boltzmann form. For the sake of illustration, the smoothed data obtained from the measured data by means of linear filtration method which were used in the inverse analysis of measured profiles are given as well.

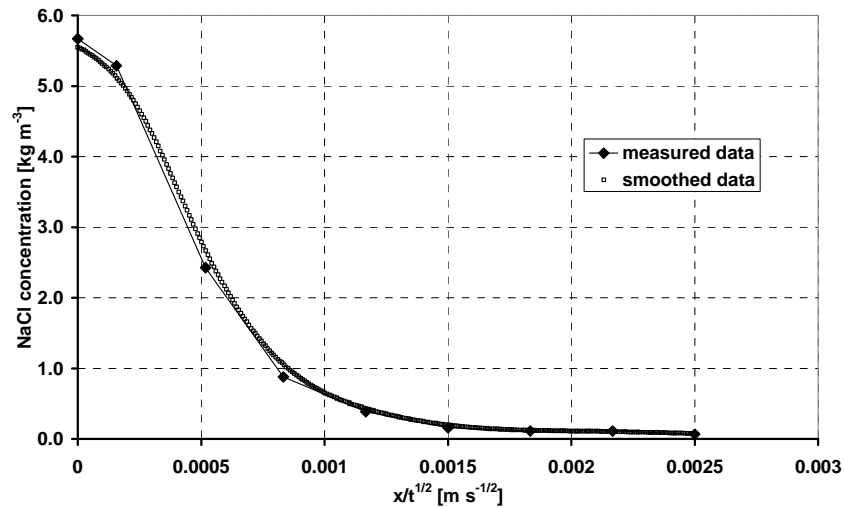


Fig. 1. Salt concentration profiles expressed in mass of NaCl per volume of the solution

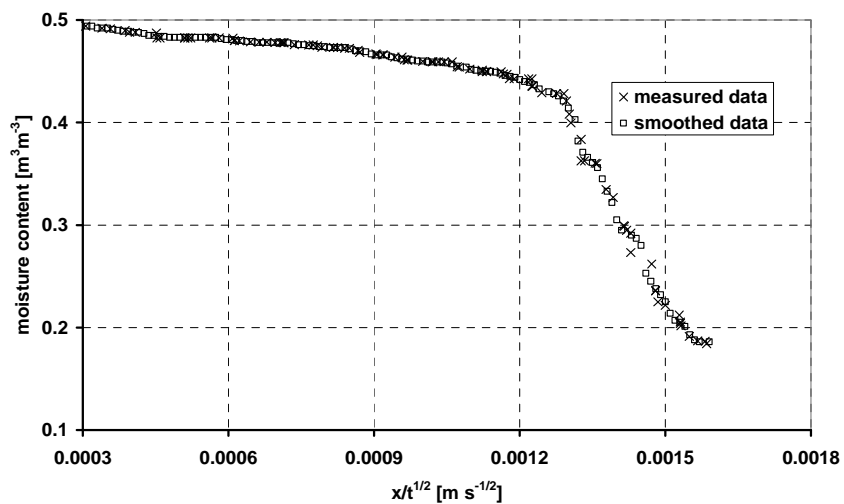


Fig. 2. Moisture profiles in water volume per volume of gypsum sample

The results of calculation of NaCl diffusion coefficient and moisture diffusivity in gypsum are shown in Figures 3 and 4.

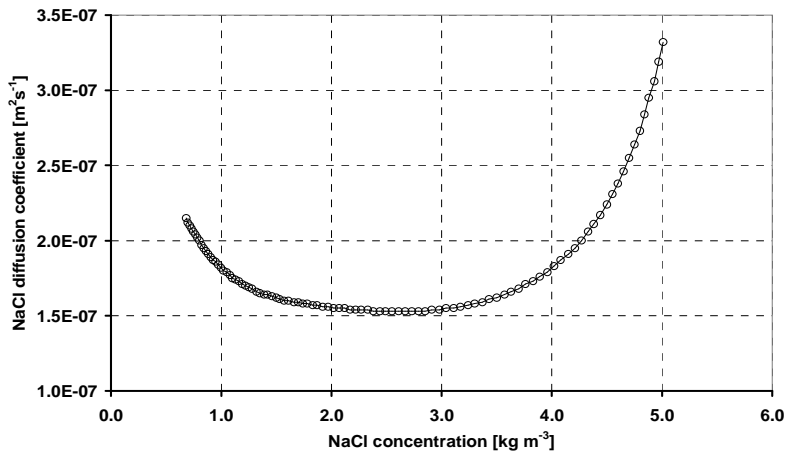


Fig. 3. NaCl diffusion coefficient in gypsum

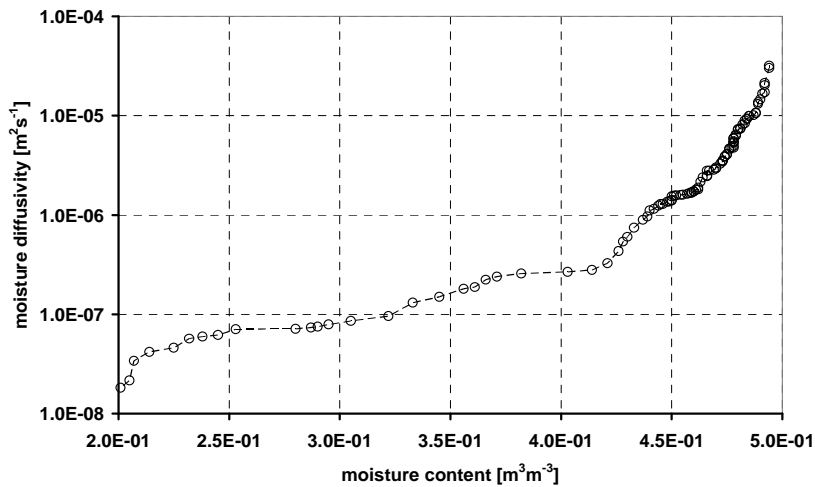


Fig. 4. Moisture diffusivity of gypsum

While the moisture diffusivity of gypsum is within the expected range for capillary active materials, the values of NaCl diffusion coefficient in gypsum are rather high, about two orders of magnitude higher than the corresponding NaCl diffusion coefficient in water. However, the forward analysis has clearly shown that the calculated coefficient can reproduce the original data fairly well. The explanation

may be possibly found in ion bonding on the pore walls because the measured salt concentration is basically the total salt content, not the free ion content. However, for a verification of this guess it would be necessary to know the chloride binding isotherm in gypsum. As no data for chloride bonding in gypsum have been published up to now, it is necessary to determine the chloride binding isotherm in future experimental work.

CONCLUSION

The experiments and calculations performed in this paper can be considered as a further step towards serious application of Bear and Bachmat diffusion-advection model for the solution of coupled moisture and salt transport in building materials in a wide range of problems appearing in practice. In future work, it is necessary to complete the obtained basic data set with the data for ion-binding isotherms and to perform a verification of the model in more complicated cases, such as salt solution penetration into a dry specimen.

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OKREŚLENIE DYFUZYJNOŚCI WILGOTNOŚCIOWEJ
ORAZ WSPÓŁCZYNNIKA DYFUZJI SOLI
W MATERIAŁACH BUDOWLANYCH

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Streszczenie. W pracy przedstawiono podstawowe modele połączonego transportu wody i soli w materiałach budowlanych. Metody określania parametrów transportu i gromadzenia wody i soli omówiono z punktu widzenia przydatności tych parametrów do budowania modeli. Na koniec podano przykład określania dyfuzyjności wodnej oraz współczynnika dyfuzji soli na podstawie zmierzonych profili wilgotności oraz stężenia soli.

Słowa kluczowe: połączonego transportu wody i soli, dyfuzyjność wilgotnościowa, współczynnik dyfuzji soli