Studying of salt diffusion coefficient in brick—Analytical and numerical methods

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The salt diffusion in fully saturated brick at isothermal condition is investigated. A commonly used experiment methodology, diffusion cell method, is adopted to estimate the salt diffusion coefficient. The mathematical model is developed. An approximate analytical solution is given. The solution agrees with the numerical solution. The analytical solution has applications in a wide number of fields with diffusion cell method. Furthermore, the diffusion coefficients of different types of salt for different types of brick are discussed. © 2005 Springer Science + Business Media, Inc.

1. Introduction

Porous materials have been widely used in building construction. The application of the materials in an outdoor environment inevitably involves exposure to moisture and solution. Due to the increased maintenance and repair costs of the building materials, salt transport in porous materials has been a subject of scientific interest for many years, and has become the focus of much attention in decades. The mechanism of the damage is known to be a function of a number of parameters such as the material, the material porosity, salt, salt concentration, temperature and so on. Thus it is important to correctly predict the salt transport in porous materials in order to well understand the damage processes. This paper studies the salt transport in one of the commonly used building materials: brick. And the sodium chloride (NaCl) is chosen as the major salt type, which is one of the main salt components found in building materials.

Brick is used as exterior and interior wall for many types of structures. Research of salt attack on brick has been reported [1, 2]. However, a detailed study of salt transport on brick is lacking. Papers are more concerned with chloride transport in concrete. These papers have emphasized the fact that the chloride transport is a complex phenomenon including the transport of chloride ions into concrete and the diffusion of ions through concrete pore water [3, 4]. The chloride ion transport is influenced significantly by the physical and chemical binding of ions in concrete. Even the process of chloride ion diffusion depends on numerous interdependent parameters. For instance, it has been reported that the diffusion process is directly influenced by the microstructure of the material [5].

The study of chloride ion transport in concrete has mainly laid on the means of experiment assuming either the steady-state or the unsteady-state case. In the steady-state case, usually the sample disc is put in between two chambers, called diffusion cell, and Fick's first law is applied. The calculated diffusion coefficient is called effective diffusion coefficient to distinguish it from the one which is obtained in the unsteady-state case [6]. In the unsteady-state situation, the sample is maintained in contact with a solution of constant salt concentration and the salt profile of the sample is measured. Fick's second law is adopted. The calculated diffusion coefficient is called apparent diffusion coefficient [6]. In both experiments, the essential difficult part is time consuming due to the concrete pore structure. Therefore, electrical methods are normally applied to the sample to accelerate the diffusion process. The process is then modelled by Nernst-Planck equation [6, 7]. For porous material brick, the often used measurement methods are destructive by drilling or grinding the sample and chemically determining the salt concentration. Some non-destructive ways have been introduced also [8].

In this paper, the diffusion of salt in fully saturated brick is studied at isothermal condition by means of diffusion cell measurement and mathematical methods. When completely saturated, the convective transport can be neglected, hence a pure diffusion mechanism by concentration gradient is going to be studied in this paper. A fundamental parameter is the diffusion coefficient in pure salt diffusion process from which the transport rate and the depth of salt as a function of time can be predicted or calculated. The mathematical methods involve analytical and numerical calculations of the diffusion process in combination with measurement data.

The diffusion cell measurement involves the application of a constant solution gradient across the sample to produce steady-state conditions [9]. Literature has shown that no analytical solutions to the problems of diffusion cell methods are currently available. A recent published paper [10] has developed semianalytical solutions to general diffusion cell measurements. A set of programs was available for the final solutions. No explicit solutions were given. As a special case in [10], Lu *et al.* [11] developed an approximate analytical solution to the diffusion cell measurement for salt diffusion in brick. This paper extends the previous work of [11] to including more measurement data and studying the measurement parameters in a more systematic way.

2. Measurement equipment

The schematic picture of the experiment equipment is displayed in Fig. 1. The basic measurement principle has been used by many researchers. The method is based on the so called porous diaphragm technique and was developed in our laboratory to measure the rate of salt diffusion in ceramic brick materials [12].

The main equipment contains two diffusion cells or chambers and the brick sample is mounted in between them. The ratio of the volume of chamber β is much bigger than that of chamber α in such way that when system reaches equilibrium stage, the change of salt concentrations in chamber β can be ignored. Also, the volume of chamber α is much bigger than that of the brick sample considering the approximation error of measuring the salt concentrations in chamber α instead of the real brick sample. Throughout all the processes both chambers are stirred well enough so that the solution can be considered as homogeneous. In addition, solution levels in two chambers are set at the same to avoid the generation of hydrostatic pressure. In fact, due to the small density difference in both chambers, hydrostatic pressure does exist. However, the negligible pressure has no effect on the measurements. More explanations in this aspect are given further in the conclusion section. Moreover, the whole experiment is kept at the isothermal condition at the temperature $25 \pm 0.05^{\circ}$ C. The change of concentration in chamber α was measured by monitoring the increases in electrical conductance as a function of time.

The control of mechanical vibration, temperature, polarization effects on the electrodes and the leakage



Figure 1 Schematic picture of the measurement equipment (not in scale).

of the electrolyte over the samples has been considered mainly in building the measurement equipment. A more detailed description of the measurement equipment and techniques are given at [12].

Different series of tests are cast. Ordinary cylindrical shaped brick samples are used with different diameters and thicknesses. Chamber β contains constant salt solution. The salt concentration in chamber α is much lower. Normally at the initial time its concentration is near 0 mol 1⁻¹.

3. Model development

One-dimensional salt diffusion in a homogeneous isotropic brick is modelled by Fick's law as [13]:

$$\frac{\partial C}{\partial t} = \frac{\partial C}{\partial x} \left(D \frac{\partial C}{\partial x} \right) \tag{1}$$

where C [mol m⁻³] is the salt concentration in brick and D [m² s⁻¹] is the salt diffusion coefficient in brick.

The following initial and boundary conditions are applied in the diffusion cell measurement for two chambers and the sample:

$$C = C_{\alpha 0}, \quad C = C_0 \quad L > x > 0 \quad t = 0$$
 (2)

$$C = C_{\alpha}, \quad A\varepsilon D \frac{\partial C}{\partial x} = V_{\alpha} \frac{\mathrm{d}C_{\alpha}}{\mathrm{d}t} \quad x = 0 \quad t > 0 \quad (3)$$

$$C = C_{\beta}, \quad A\varepsilon D \frac{\partial C}{\partial x} = -V\beta \frac{\mathrm{d}C_{\beta}}{\mathrm{d}t} \quad x = L \quad t > 0$$

(4)

where α , chamber α ; β , chamber β ; L, length of the brick sample (m); A, area of the brick sample which is exposed to chambers (m^2) ; ε , porosity of the brick sample (m³ m⁻³), V_{α} , volume of chamber α (m³); V_{β} , volume of chamber β (m³); $C_{\alpha 0}$, initial salt concentration in chamber α (mol m⁻³); $C_{\alpha\beta}$, initial salt concentration in brick (mol m⁻³); C_{α} , salt concentration in chamber α (mol m⁻³); C_{β} , salt concentration in chamber β (mol m⁻³); Equations 3 and 4 assume that the rates of the changes of salt contents in chambers are equal to the rates of flow of salt from the brick into the chambers. If necessary a resistance factor can be introduced to present the rates of salt concentration change with respect to that of the brick. If the volume ratio of chamber β and chamber α is big in the sense that when equilibrium reaches the concentration in one of the chambers, say chamber β , changes small, then the boundary condition (4) at x = L can be simplified as the following:

$$C_{\beta} = \text{const}, \quad C = C_{\beta} \quad x = L \quad t > 0$$
 (5)

In our tests we assume Equation 5 is satisfied throughout the whole test $(\frac{V_{\beta}}{V_{\alpha}} \ge 93.3)$. As the diffusion starts, salt in chamber β diffuses

As the diffusion starts, salt in chamber β diffuses through the brick sample to the chamber α . The salt concentrations in chamber α are measured by monitoring the conductance with AC as a function of time. The salt diffusion coefficient *D* in brick is then calculated from the measured data. A simple procedure of evaluating D from the tests is as following:

Assuming at time $t > t_0$, steady-state is reached, integrating Equation 3 and collecting term gives the following formula which is used to estimate *D*:

$$\varepsilon D = \ln \left(1 + \frac{C_{\alpha} - C_{\alpha o}}{C_{\beta} - C_{\alpha}} \right) \frac{V_{\alpha} L}{A(t - t_{o})} \tag{6}$$

As $C_{\alpha} \ll C_{\beta}$, Equation 6 can be simplified as

$$\varepsilon D = \frac{C_{\alpha} - C_{\alpha o}}{C_{\beta}} \frac{V_{\alpha} L}{A(t - t_{o})} \tag{7}$$

By linear regression method (e.g. [14]), *D* can be estimated from the measurements.

4. Analytical solutions

The analytical solution of Equation 1 in semi-infinite material with the following initial and boundary condition is widely used by researchers (e.g. [15]):

$$C = 0, \quad x > 0 \quad t = 0$$

 $C = C_0, \quad x = 0 \quad t > 0$
(8)

The solution can be expressed as

$$C = C_{\rm o} {\rm erfc} \frac{x}{2(Dt)^{1/2}} \tag{9}$$

Integrating Equation 9 by x gives the total amount of salt diffuses to the material with respect to time:

$$M = 2C_{\rm o} \left(\frac{Dt}{\pi}\right)^{1/2} \tag{10}$$

The equation can be used to determine the diffusion coefficient with the help of linear regression methods [15].

In this paper an approximate analytical solution of Equations 1 to 3 and 5 with finite sized sample is developed. Apply Laplace transformation on Equation 1, we find the solution to be

$$\bar{C}(s,x) = a\cosh(qx) + b\sinh(qx) + \frac{C_0}{S} \quad (11)$$

where $q = \sqrt{\frac{s}{D}}$ and the coefficients *a*, *b* are determined by the boundary conditions (3) and (5). A bar over a function designates its Laplace transform on *t*. For instance,

$$\bar{C}(s,x) = \mathcal{L}(C(t,x)) = \int_0^\infty \exp(-s\tau)C(\tau,x)\,\mathrm{d}\tau$$
(12)

Apply Laplace transform in boundary condition (3), we get the left side

$$A\varepsilon D\mathcal{L}\left(\frac{\partial C}{\partial x}\right)|_{x=0} = A\varepsilon D(a \operatorname{qsin}h(\operatorname{qx}) + bq \operatorname{cos}h(qx))|_{x=0} = A\varepsilon Dbq$$
(13)

and the right side

$$V_{\alpha} \mathcal{L}\left(\frac{\mathrm{d}C_{\alpha}}{\mathrm{d}t}\right) = V_{\alpha}(s \ \bar{C}_{\alpha} - C_{\alpha}|_{t=0})$$

$$= V_{\alpha}(s \ \bar{C}|_{x=0} - C_{\alpha o})$$

$$= V_{\alpha}\left(s\left(a \cosh(qx) + b \sinh(qx) + \frac{C_{o}}{s}\right)\Big|_{x=0} - C_{\alpha o}\right)$$

$$= V_{\alpha}sa + V_{\alpha}(C_{o} - C_{\alpha o}) \qquad (14)$$

Similar action can be done to Equation 5. We get the following boundary conditions in its Laplace form:

$$asV_{\alpha} - b\,A\varepsilon Dq = -V_{\alpha}(C_{\rm o} - C_{\alpha \rm o}) \qquad (15)$$

$$a\cosh(qL) + b\sinh(qL) = \frac{C_{\beta} - C_{o}}{s}$$
(16)

The coefficient a is then found to be

$$a = \frac{-V_{\alpha}(C_{o} - C_{\alpha o})\sinh h(qL) + \frac{(C_{\beta} - C_{o})\varepsilon DAq}{s}}{sV_{\alpha}\sinh(qL) + \varepsilon DAq\cosh(qL)} \quad (17)$$

Expressing sinh(qL) and cosh(qL) in terms of q, for t > d, the inverse Laplace transform of the second term of Equation 17 is [11]:

$$\mathcal{L}^{-1}\left(\frac{\frac{(C_{\beta}-C_{o})\varepsilon DAq}{s}}{sV_{\alpha}\sinh(qL)+\varepsilon DAq\cosh(qL)}\right)$$
$$\cong (C_{\beta}-C_{o})\left(1-\exp\left(-\frac{t}{d}\right)\right)$$
(18)

where d is defined as

$$d = \frac{V_{\alpha}L}{\varepsilon AD} + \frac{L^2}{2D}$$
(19)

Similarly, the inverse Laplace transform of the first term of Equation 17 is obtained as

$$\mathcal{L}^{-1}\left(\frac{-V_{\alpha}(C_{o} - C_{\alpha o})\sinh h(qL)}{sV_{\alpha}\sinh(qL) + \varepsilon DAq\cosh(qL)}\right)$$
$$\cong \frac{V_{\alpha}(C_{o} - C_{\alpha o})}{V_{\alpha} + \frac{\varepsilon LA}{2}}\exp\left(-\frac{t}{d}\right)$$
(20)

As the volume ration between chamber α and the specimen is big, $AL \ll V_{\alpha}$, therefore Equation 20 can be approximated as

$$\frac{V_{\alpha}(C_{\rm o} - C_{\alpha \rm o})}{V_{\alpha} + \frac{\varepsilon LA}{2}} = \frac{C_{\rm o} - C_{\alpha \rm o}}{1 + \frac{\varepsilon LA}{2V_{\alpha}}} \cong C_{\rm o} - C_{\alpha \rm o} \quad (21)$$

Hence Equation 17 gives

$$\mathcal{L}^{-1}(a) = -(C_{o} - C_{\alpha o})\exp\left(-\frac{t}{d}\right) + (C_{\beta} - C_{o})\left(1 - \exp\left(-\frac{t}{d}\right)\right) \quad (22)$$

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We get from Equations 3 and 11 as

$$C_{\alpha} = C|_{x=o} = \mathcal{L}^{-1} \left(a \cosh(qx) + b \sin(qx) + \frac{C_{o}}{s} \right)|_{x=o} = \mathcal{L}^{-1}(a) + C_{o}$$
$$= (C_{\beta} - C_{o}) - (C_{\alpha o} - C_{\beta}) \exp\left(-\frac{t}{d}\right) + C_{o} \quad (23)$$

Note that the characteristic number d is of the fundamental importance in determining the salt transport process in brick and thus in chambers. It has a time unit.

The temporal change of the concentration in chamber α is explicitly expressed in Equation 23. In [11], it was assumed that the initial salt concentration in chamber α and the brick sample are the same. This restriction is eliminated in this paper. Thus the result can be seen as an extension from the precious result in [11].

5. Model predictions and discussions

The measurements were divided into three groups in order to see the effects on the evaluated D of different chamber and sample parameters. Note that the diffusion coefficient D is determined from the measurement by assuming the steady-state condition is achieved. Ideally, the steady-state condition is achieved when salt concentration changes in both chambers α and β are neglected. However, the diffusion cell measurement requires a concentration change in chamber α . Hence only quasi-steady-state condition is set on the sample. The validation of the analytical and the numerical results are applied when $t > t_{\alpha 0}$, where t_0 presents the time required for detectable salt quantities diffuse through the specimen. The time t_0 also represents the lower bound of the time required to achieve quasi-steady-state condition.

Table I sumarizes briefly the sample properties in group I. The salt was chosen as NaCl. All the brick samples are new red brick. Corresponding calculated salt diffusion coefficients of NaCl from the experiment are also presented in the same table. Initial salt concentrations in chamber α and brick samples were varied. The simulation results are showed in Fig. 2. The lower bounds of the required time to reach the quasi-steady-state conditions were taken as 15, 20 and

10 h and the measurement duration periods were 145, 163 and 100 h. Thus the time required for reaching the quasi-steady-state conditions was less than 13% compared to the total measurement duration time in group I. This number is smaller than 20% which was suggested in [9] for guaranteeing the quasi-steady-state condition.

Table II presents the sample and chamber properties in group II. Salt presents again NaCl. Different types of brick were selected such as old light brick, new red brick and old dark brick. Results are displayed in Fig. 3. In this group, the time required for reaching the quasisteady-state condition was 10, 10 and 10 h and the measurement duration was 113, 110 and 132 h respectively. Thus the time required for achieving the quasi-steadystate condition was less than 10% compared to the total measurement time.

Table III shows the sample and chamber properties in group III. The big difference in this group from the previous groups was that different types of salt were used such as sodium carbonate (Na₂CO₃) and calcium chloride (CaCl₂). Results are demonstrated in Fig. 4. In this group, the time required for reaching the quasisteady-state condition was 20, 10 and 20 h. respectively and the corresponding measurement duration time was 2325, 233 and 140 h. Thus the time required for achieving the quasi-steady-state condition was less than 15% of the total measurement time.

Group II and group III demonstrate that the theories developed for sodium chloride diffusion in new red brick in this paper are applicable for other types of salt and brick also.

These figures can serve as the tests of the accuracy of the proposal analytical solution. It can be seen the close agreement between the analytical, numerical solutions and measured data.

Fig. 5 also shows the simulated distribution of NaCl concentrations for different time periods for sample 1 in group I. Linear profile was approaching for the sample after 10-h diffusion.

Another property of the brick which affecting the salt transport is its binding capability. The salt binding effect isn't studied in this paper. It was claimed in [9] that the binding increases the time required to achieve a quasi-steady-state profile. Fig. 5 demonstrates that salt concentration profiles in the brick sample quickly follow a linear property as time increases. The binding effect ceases after ten hours. Hence it is believed that the binding effect is so small that it can be ignored.

TABLE I Specimen properties, group I

Group I, temperature (25°C)										
	$A (\mathrm{cm}^2)$	L (cm)	$\varepsilon ~({\rm cm}^3/{\rm cm}^3)$	$V_{\alpha}(\mathrm{cm}^3)$	$V_{\beta}(\text{cm}^3)$	$D \times 10^{-5} (\mathrm{cm}^2 \mathrm{s}^{-1})$	$C_{\alpha 0} \pmod{1^{-1}}$	$C_{\beta} \pmod{1^{-1}}$	$C_o mmol \ 1^{-1}$	
Sample 1	4.83	1.04	0.223	23.51	2200.0	0.502	0.0	100.0	0.0	
New red brick						NaCl				
Sample 2	4.83	1.04	0.223	23.51	2200.0	0.501	0.22	100.0	50.0	
New red brick						NaCl				
Sample 3	4.83	10.04	0.223	23.51	2200.0	0.484	11.56	100.0	50.0	
New red brick						NaCl				

Group II, temperature $(25^{\circ}C)$										
	$A (\mathrm{cm}^2)$	L (cm)	$\varepsilon (\text{cm}^3/\text{cm}^3)$	$V_{\alpha}(\text{cm}^3)$	$V_{\beta}(\text{cm}^3)$	$D \times 10^{-5} (\mathrm{cm}^2 \mathrm{s}^{-1})$	$C_{\alpha 0} \pmod{1^{-1}}$	$C_{\beta} \pmod{1^{-1}}$	$C_o \ mmol \ 1^{-1}$	
Sample 4 Old light brick	4.85	1.04	0.284	23.53	2200.0	0.469 NaCl	0.39	100.0	100.0	
Sample 5 New red brick	4.82	1.02	0.248	23.59	2200.0	0.510 NaCl	0.44	100.0	100.0	
Sample 6 Old dark brick	4.83	1.12	0.205	23.13	2200.0	0.348 NaCl	0.36	100.0	100.0	















Figure 2 (a) Calculation of salt concentrations in chamber α for sample 1, group I. (b) Calculation of salt concentrations in chamber α for sample 2, group I. (c) Calculation of salt concentrations in chamber α for sample 3, group I.

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Group III, temperature $(25^{\circ}C)$										
	$A (\mathrm{cm}^2)$	L (cm)	$\varepsilon (cm^3/cm^3)$	$V_{\alpha}(\text{cm}^3)$	$V_{\beta}(\text{cm}^3)$	$D \times 10^{-5} (\mathrm{cm}^2 \mathrm{s}^{-1})$	$C_{\alpha 0} \pmod{1^{-1}}$	$C_{\beta} \pmod{1^{-1}}$	$C_o \ mmol \ 1^{-1}$	
Sample 7 New red brick	4.83	1.04	0.231	23.51	2200.0	0.270 Na ₂ CO ₃	0.0	100.0	50.0	
Sample 8 New red brick	4.83	1.04	0.231	23.51	2200.0	0.254 Na ₂ CO ₃	0.0	100.0	0.0	
Sample 9 New red brick	4.83	1.04	0.231	23.51	2200.0	0.319 CaCl ₂	0.03	100.0	50.0	













Figure 3 (a) Calculation of salt concentrations in chamber α for sample 4, group II. (b) Calculation of salt concentrations in chamber α for sample 5, group II. (c) Calculation of salt concentrations in chamber α for sample 6, group II.



Figure 4 (a) Calculation of salt concentrations in chamber α for sample 7, group III. (b) Calculation of salt concentrations in chamber α for sample 8, group III. (c) Calculation of salt concentrations in chamber α for sample 9, group III.



Figure 5 Salt concentration profiles for sample 1, group I.

6. Conclusions

This paper presents a study of salt diffusion, mainly sodium chloride diffusion, in brick with analytical and numerical methods combining the experiment. Sodium carbonate and calcium chloride diffusions are also considered. Different kinds of brick samples are used in the experiment. Mathematical model is constructed and its analytical solution is obtained.

The analytical and numerical data are compared with the measurement data. Good agreement is achieved. The most important conclusion to be drawn is the explicit expression by analytical solution of the under measured salt concentration in the chamber which provides the easiest way in determining salt diffusion coefficient and studying the diffusion cell method. The solution mainly contains one character number of the sample and experimental properties, which characterizes the salt diffusion procedure.

Once concern in the measurement is the hydrostatic pressure generated by different solution densities when equal heights are set for the both chambers at the starting of the experiments. The pressures in chamber β were estimated to be 0.079 mmH₂O higher compared to those in chamber α which was very small. Furthermore, it was observed that the hydrostatic equilibrium was quickly reached in about 10 min when the solution level in chamber α was about 0.079 mm higher than that in the chamber β . This time period is much shorter than that required achieving quasi-steady-state condition, i.e., t_0 presenting the time for detectable salt quantities diffuse through the specimen. As discussed in the previous section, experimental, numerical and analytical data are applicable for only $t > t_0$. Therefore, it can be concluded that the small hydrostatic pressure generated in the beginning has no any effect in the measured diffusion coefficients.

Further concern is the osmotic effect on the measured data, as brick is a porous material. Osmotic pressure exists only in a semipermeable membrane which allows certain chemical species to pass through it. The ceramic brick used in the experiments are all engineering bricks manufactured commercially for buildings. These bricks are permeable for both water and salt and osmotic pressure can not develop over it. This has been tested in our laboratory. Above all, the commercially used membrane materials for osmotic studies or hyperfiltration (reverse osmosis) are cellulose triacetate and aromatic polyamides whose pore sizes are at most 2 nanometers [16]. In ceramic bricks the pore sizes are of order of micrometers and no impermeability against salts has been reported. Hence there is no osmotic pressure during the experiments.

As we mentioned in introduction, salt diffusion is a complex phenomenon. The diffusion coefficient D can vary with concentration. However, in practical experi-

ment, D is often defined as constant. Above all, there are different kinds of diffusion coefficients depending on the diffusing substance, system and conditions, In this paper, binary diffusion coefficients of salt at constant temperature 25°C are considered. Data generated for brick sample in our experiment show the average diffusion coefficient is $D = (0.499 \pm 0.004) \times 10^{-5}$ $cm^2 s^{-1}$ for NaCl in the concentration range of 0.05 mol dm^{-3} for fully saturated red new brick. This value has been verified by many experiments which haven't been shown in this paper due to the space limitation. This value can be used as the salt diffusion coefficient in fully saturated new red brick. More experimental values for NaCl diffusions in other types of brick materials are given in [12]. Furthermore, the diffusion coefficients for NaCl and other types of salt as functions of temperatures and salt concentrations have also been measured and simulated, the data are prepared under publication.

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