International Journal of Heat and Mass Transfer 56 (2013) 212–222

Contents lists available at SciVerse ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Moisture diffusivity in mortars of different water–cement ratios and in narrow ranges of air humidity changes

Halina Garbalińska [a], Stefan J. Kowalski [b,*], Maciej Staszak [b,1]

[a] West Pomeranian University of Technology, Faculty of Civil Engineering and Architecture, Aleja Piastów 50, 70-311 Szczecin, Poland
[b] Poznań University of Technology, Institute of Technology and Chemical Engineering, pl. Marii Skłodowskiej Curie 2, 60-965 Poznań, Poland

A R T I C L E I N F O

Article history:
Received 28 June 2012
Accepted 16 September 2012
Available online 27 October 2012

Keywords:
Cement-based mortar
Narrow range humidity
Experiments
Analytical solutions
Diffusivity

A B S T R A C T

The aim of this paper is to assess moisture diffusivity in a group of cement-based mortars of different water–cement ratios by different moisture contents. Five levels of moisture content equivalent to five narrow ranges of contiguous air humidity changes were considered. The effective (average) diffusion coefficients were estimated for these five moisture levels and the experiments covered mortars with three different water–cement ratios amounting to 0.50; 0.65; 0.80. The diffusion coefficients were determined by way of adjustment of the theoretical and experimental drying curves. The theoretical drying curves were determined from numerical solutions of linear diffusion equations with a constant (average) diffusion coefficient for a given narrow humidity range. The theoretical drying curves were correlated with those corresponding experimentally using the Levenberg–Marquardt modified iterative algorithm. The Gauss–Newton numerical method was used for a minimization problem by fitting the drying curves, and in this way the values of diffusion coefficients for the selected materials were determined.

1. Introduction

In recent years, research on moisture transport in cement-mortar materials has been conducted in a double-track manner. On the one hand, there have been many studies concerning modification of the internal structure with a view to controlling the transport of pure water or water which carries contaminants [1–6]. On the other hand, however, some attempts have been made towards obtaining a more and more precise description of moisture transport processes in porous materials. The latter has entailed the necessity for continuous development of mathematical models and the need to collect accurately-determined material parameters. Hence, models of moisture transport in building materials have been developed gradually [7–13]. At the same time, the techniques for measuring coefficients of moisture transport in porous materials have undergone significant improvements.

Many methods for determining transport coefficients are based on moisture profiles (see e.g. [14]). Most frequently, in such studies, rod samples are used as they make it possible to determine moisture for a number of measuring spots and on that basis to prepare the moisture profiles. In order to measure moisture, methods such as nuclear magnetic resonance (NMR) [15–18] microwave reflection, transmission [19,20] or beam suppression γ [21,22] are used.

According to [23], these are not particularly suitable for thin plaster materials. In [23], a specific method, the piecewise constant κ-PCK method, was proposed. Its results were confronted with two other methods: the Motano method (originally used for diffusion in metals [24], and then adapted for moisture diffusion, e.g. [14]) and the double integration method [14]. In paper [23] it was stated that the double integration method and the Motano method present high compliance in all moisture content ranges. Nevertheless, an analysis conducted previously by Černy et al. [25] showed that the Motano method was not very precise at both low and high moisture levels. Tested in [23], the PCK method proved its relatively good compliance with both the double integration method and the Motano method for high moisture values, whereas it was established that it was of no use for low moisture levels. According to [23], for the scope of sorption, it needs to be supplemented by another method as, for instance, that presented in [26].

In [5] an attempt was made to reconstruct the dependence of diffusivity on water content. Drawn in three different ways (ideal continuous wetting, ideal continuous drying, random access of pores by water, experiment), the diffusivity graphs showed high qualitative and quantitative differences. However, in each case, the non-linearity of the phenomenon manifested itself very clearly.

As assessed by the authors of [5], courses of diffusivity in function of relative water content that had been presented in the past

---

[*] Corresponding author. Tel.: +48 616653622, mobile: +48 605983460; fax: +48 616653649.

E-mail addresses: halina@zut.edu.pl (H. Garbalińska), stefan.j.kowalski@put.poznan.pl (S.J. Kowalski), maciej.staszak@put.poznan.pl (M. Staszak).

URL: http://www.zip.tce.put.poznan.pl/sjkowalski (S.J. Kowalski).

[1] Tel.: +48 61 665 3760; fax. +48 61 665 3649.

0017-9310/$ - see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ijheatmasstransfer.2012.09.026

for unsaturated soils, cement stone and other similar products, did not comply with one another, or it was impossible to indicate one specific variation course of the transport coefficient towards relative water content.

Although the literature delivers many more works presenting various methods for determining moisture transport coefficients [14,15,25,27–30], they provide results which are rather difficult to compare.

Undoubtedly, this can be attributed to the complexity of the process, because the flow of water in porous material may be subject to different mechanisms, dependent on the level of dampness. Within the range of hygroscopic dampness, the dominating diffusion is that which is accompanied by laminar flow, effusion and surface flow, whereas at high moisture levels there is also some capillary flow that dominates within the supra-hygroscopic range. The situation is additionally complicated by the fact that two porous objects tested at the same moisture can present entirely different values of moisture transport coefficients, just because they can differ widely from each other in their porosity structures.

There are a considerable number of works presenting attempts to link the structures to material parameters that are responsible for moisture transport, e.g. [5,10,31–36], whereas many researchers emphasize the importance of the w/c parameter for forming the microstructure, proving good opportunities to use it for controlling the diffusivity coefficient, e.g. [2–5,37–39].

With the above problem in mind – the moisture transport nonlinearity and the dependence of its course on not only the moisture-related conditions, but on material-specific conditions, as the pore structure geometry – an attempt was made within the framework of this work to propose what could be a simple and efficient method for determining the diffusivity coefficient D. Here it was treated as an effective coefficient, describing jointly all transport process components that contribute to the global migration of water in a variable manner, depending on the dampness level and the specific geometry of the pores.

This paper is a continuation of previous works on the transport of water in porous materials, where the research and calculation methods were carried out in cement mortars. Initially, the tests concerned sorptive and desorptive measurements of the diffusion coefficient in relation to broad moisture intervals [40–42]. Unfortunately, this made the assessment of non-linearity of the moisture transport phenomena in porous materials more difficult. For this reason, the studies conducted in subsequent years focused only on a few, narrow moisture ranges [43–45], permitting an evaluation of the moisture-related variability of the diffusion coefficient.

In the studies presented herein, the authors decided to divide the entire range of air relative humidity $\Delta\varphi$ to five narrow, neighboring sections $\Delta\varphi_i$. For each of them, drying kinetics was measured, where the cement mortar samples went from a higher balance level to a lower balance level. Taking into consideration the important role of the water–cement ratios for forming the microstructure and influencing the diffusion coefficient, as signaled in the literature, those mortars that differed from one another with just this indicator were tested. The results collected in subsequent desorption cycles helped determine diffusion coefficients in three mortars for five moisture ranges.

In order to establish the dependence of the diffusion coefficient on the moisture content in cement-based mortar an attempt was made to determine the drying kinetics for the following narrow ranges of the air humidity changes: $12\% \leq \varphi \leq 30\%$, $30\% \leq \varphi \leq 50\%$, $50\% \leq \varphi \leq 75\%$, $75\% \leq \varphi \leq 85\%$, $85\% \leq \varphi \leq 97\%$.

The starting point for the calculations was an assumption of the constant diffusion coefficient for each narrow range of the air humidity $\Delta\varphi_i$. We anticipated that in the narrow ranges of the air humidity the magnitude of diffusion coefficient could not undergo excessively large changes and presupposed that the drying kinetics in the specified above ranges $\Delta\varphi_i$ was described by the linear diffusion equation.

The solution of this equation was obtained analytically through the application two methods and presented in the form of a probability integral and in the form of a series with respect to the system of eigenfunctions. The drying kinetics obtained in this way were correlated with those obtained experimentally. The Gauss–Newton method was used for a minimization problem by fitting the drying kinetics based on the probability integral, and the Levenberg–Marquardt optimization algorithm was used for the second solution. In this way the values of the diffusion coefficients for the selected materials and five ranges $\Delta\varphi_i$ were determined.

2. Experimental

2.1. Preparation of samples

The experiment concerned three mature cement mortars with different water–cement ratios. The ratios took up the following values: $w/c = 0.50, 0.65, 0.80$. To produce all the mortars, the same cement, aggregate and potable city water were used.

Pure Portland cement, class 32.5, with the following chemical composition was used:

roasting loss: 1.38%,
particles insoluble in HCl: 0.38%,
sulphuric anhydride SO_3: 2.30%,
silica SiO_2: 19.99%,
ferric oxide Fe_2O_3: 4.03%,
aluminium oxide: Al_2O_3: 4.51%,
calcium oxide CaO: 64.86%,
magnesium oxide MgO: 2.49%,
other: 0.06%.

The mineralogical composition of the cement applied was as follows: C_3S – 67.44%, C_2S – 6.45%, C_3A – 5.14%, C_4AF – 12.25%. This data was used for a later controlling measurement of the degree of hydration in the cement at each stage of curing in the individual mortars.

All the mortars were made based on quartz sand, grain size ≤2 mm. Initially, it was intended that natural sand collected from natural deposits should be used. Detailed tests showed, however, that its grain size content was highly diversified, containing oversize particles (6%). For this reason, the sand served as initial material only. After drying up to solid mass, it was screened with a vibrating woven screen (#0.125, #0.25, #0.5, #1.0, #2.0 mm), with a simultaneous elimination of oversize particles (over 2.0 mm). Individual fractions were stored separately, in tight containers. The target aggregate was composed of individual fractions in the following proportions: $0 \div 0.125$ mm – 2.1%, $0.125 \div 0.25$ mm – 13.6%, $0.25 \div 0.5$ mm – 24.3%, $0.5 \div 1.0$ mm – 30.0%, $1.0 \div 2.0$ mm – 30.0%.

The sand, cement and water thus prepared occurred in the mortars in the following weight proportions:

Mortar 1: ratio $w/c = 0.50$, cement-sand ratio → 1: 3.00,
Mortar 2: ratio $w/c = 0.65$, cement-sand ratio → 1: 3.52,
Mortar 3: ratio $w/c = 0.80$, cement-sand ratio → 1: 4.05.

The equation $(V_c + V_w)/V_k = \text{const.}$ was the starting point for defining the recipe. Adherence to it guaranteed that in each mortar there was the same volumetric content of matter participating in moisture transport (cement stone) in relation to matter that is practically inactive in this process (the aggregate).

From each mix a set of cylindrical samples was produced (diameter $d = 8$ cm, height $h = 16$ cm) in order to proceed with testing the moisture transport coefficients. After removal of the forms, the