

Determination of moisture diffusivity in porous materials using gamma-method

M.I. Nizovtsev, S.V. Stankus, A.N. Sterlyagov*, V.I. Terekhov, R.A. Khairulin

Institute of Thermophysics SB RAS, Novosibirsk 1, Lavrent'ev Avenue, Novosibirsk 630090, Russia

Received 11 December 2007; received in revised form 27 December 2007

Available online 17 March 2008

Abstract

In the present paper, results of an experimental gamma ray study of moisture transfer processes in porous material are reported. Evolution of moisture profiles in porous concrete samples during sorption and capillary moistening has been examined. Measured moisture profiles were used to determine, by the Boltzmann–Matano method, the coefficient of moisture diffusion versus moisture content of the material under various moistening conditions.

© 2008 Published by Elsevier Ltd.

Keywords: Moisture diffusivity; Porous material; Gamma-method

1. Introduction

Heat and moisture transfer in porous media form the basis for many industrial processes. This transfer permits no simple analytical description, and presently available calculation procedures lean upon many restrictions and assumptions. To calculate thermal and moisture conditions in porous materials, it is necessary to know transport characteristics of moisture, which govern moisture transfer processes in such materials. That is why experimental studies of moisture transfer in porous materials aimed at determination of moisture characteristics of such materials attract much attention.

In the experimental study of moisture transfer processes, advanced contactless techniques offer much promise. Unlike traditional methods [1–3], these techniques allow rapid profiling of moisture concentration in porous media without destruction of materials, i.e., without introduction of inaccuracies resulting from violation of material structure. The best known and most widely used contactless

techniques for determining the humidity of porous materials are the nuclear magnetic resonance (NMR) method [4–6] and scanning neutron radiography [7,8]. Each of these methods has its own advantages and disadvantages. Of NMR, good accuracy and high spatial resolution are typical. This method, however, shows weak sensitivity to bound moisture and allows testing of relatively small samples only. Neutron radiography permits high-sensitive measurements of moisture concentration in porous materials in a broad range moisture content of the material; yet, the experimental data obtained with this technique are significantly affected by experimental noise and by inhomogeneities in the porosity of the material under testing.

In the present study, moisture transfer processes were examined using the gamma ray method, which allows high-precision measurement of moisture concentration in a porous material in a broad range of moisture contents [9,10].

2. Gamma-method

2.1. Description of the method

The contactless gamma ray method can be used to determine thermophysical properties of various materials such

* Corresponding author. Tel.: +7 383 3391336; fax: +7 383 3308480.

E-mail addresses: nizovtsev@itp.nsc.ru (M.I. Nizovtsev), stankus@itp.nsc.ru (S.V. Stankus), sterlyagov@itp.nsc.ru (A.N. Sterlyagov), terekhov@itp.nsc.ru (V.I. Terekhov), khairulin@itp.nsc.ru (R.A. Khairulin).

Nomenclature

d	thickness of sample (m)
D_w	effective coefficient of moisture diffusion (m^2/s)
I_s	gamma intensity transmitted by dry material (s^{-1})
I_w	gamma intensity transmitted by moist material (s^{-1})
r	pore radius (m)
S	Boltzmann variable ($\text{m}/\text{s}^{0.5}$)
t	time (s)
T	temperature ($^{\circ}\text{C}$)
ΔV	volumetric fraction of pores in the material (%)
W	moisture content of the material (kg/kg)

W_m	relative moisture content of the material (%)
x	spatial coordinate (m)

Greek symbols

μ_w	gamma ray attenuation factor of water ($\epsilon_w = 0.00862 \text{ m}^2/\text{kg}$ [13])
ρ_s	density of dry material (kg/m^3)

Subscripts

s	dry material
w	water

as, for instance, metal alloys [11]. The method consists in raying a sample under testing with a narrow gamma beam. The use of penetrating radiation for studying physical properties of materials is underlined by the capability of material atoms to interact with gamma quanta, which leads to beam intensity attenuation in the sample as the gamma radiation penetrates through the material.

In studying moisture transfer processes in porous materials, consideration should be given to the fact that the probing gamma radiation is attenuated, first, by the material skeleton and, second, by the moisture contained in material pores. Then, the gamma ray attenuation in a moist porous material can be described with the Bouguer law [12]:

$$I_w = I_s \cdot e^{-\rho_s \cdot \mu_w \cdot W_m \cdot d}. \quad (1)$$

It follows from (1) that, for the gamma ray intensity transmitted by a moist material to be adequately predicted, one has to know the gamma ray intensity transmitted by dry material. The gamma radiation mass attenuation factor of water is a constant ($\mu_w = 0.00862 \text{ m}^2/\text{kg}$ [13]). Thus, if the material under study has uniform porosity and uniform thickness, then the transmitted gamma ray intensity varies only with material humidity. On rearrangement, from (1) we obtain the following formula using which one can determine the relative moisture content of the material under study:

$$W_m = \frac{1}{\epsilon_w \rho_s d} \ln \left[\frac{I_s}{I_w} \right]. \quad (2)$$

Thus, relation (2) can be used to determine the moisture content from gamma ray intensities transmitted by dry and moist materials.

2.2. Experimental set-up

The present experimental study of moisture transfer processes in cellular concrete samples was carried out on a custom-made experimental set-up (Fig. 1). A detailed description of the set-up is given elsewhere [9,10].

The set-up comprised a container with inserted material sample. The upper and lower parts of the sample were contained in sealed chambers. In the upper chamber, a reservoir filled with a sorbent was located. A vessel with water was placed in the lower chamber. Both chambers were provided with thermohydrometric probes to register the air temperature and relative air humidity during the experiments.

The container was rayed at a certain height with gamma quanta (beam diameter 4 mm). The radiation source was Cs^{137} , known to emit gamma quanta with energy 662 keV. The transmitted radiation intensity was measured by a scintillation counter and registered by a computerized data gathering and processing system.

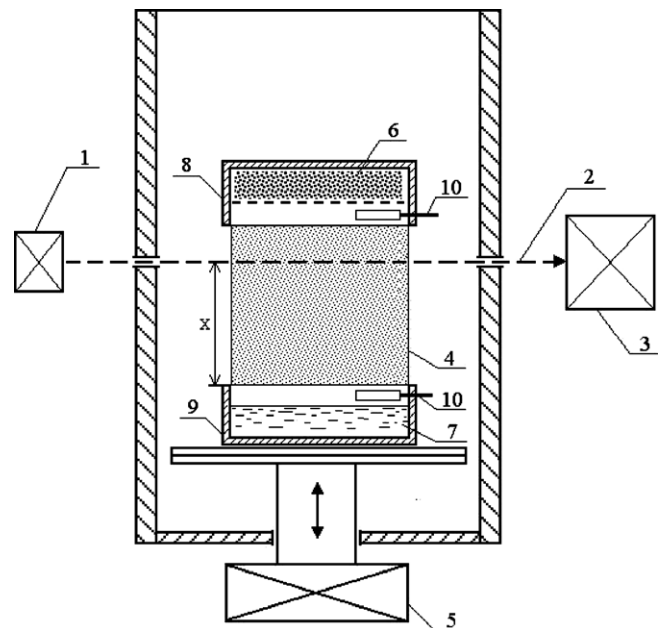


Fig. 1. Experimental set-up: (1) radiation source (Cs^{137}), (2) gamma ray, (3) scintillation gamma counter, (4) material sample under testing, (5) coordinate device, (6) sorbent, (7) water, (8) upper chamber, (9) lower chamber, (10) thermohydrometric probes.

The sample could be displaced relative to the gamma ray on a coordinate device, which allowed us to measure the attenuated gamma ray intensity at different (over height) sections of the sample.

The material under study was cellular concrete with density 400, 600, and 700 kg/m³. From this material, samples were prepared having dimensions 50 × 100 × 100 mm. Prior to experiments, the samples were given a vacuum-drying treatment at a temperature of 105 °C, following which their mass displayed no changes during subsequent drying. Then, a warm paraffin–colophony mixture was applied onto the side surfaces of the samples to make these surfaces impermeable to moisture. In this way, a one-dimensional process was created. Afterwards, the samples were fitted into the experimental set-up to measure the gamma ray attenuation coefficient for the perfectly dry material.

The experimental procedure used in the present study to examine the moisture transfer was as follows. Certain humidity conditions were produced at the upper and lower surfaces of the sample; these conditions were then kept unchanged in time. The sample under study was moved, with the help of the coordinate device, to a certain position in the way of the gamma ray. From the measured gamma ray attenuation factor we were able to determine the moisture content of the material at certain cross-section in the sample. Thus, performing repeated measurements at different cross-sections, we were able to measure moisture profiles over the height of the sample at various times.

Our calculations and testing data showed that the relative systematic error in our experiments was 0.012 throughout the entire range moisture content of the material. This error was dominated by the inaccuracy in determining the gamma ray attenuation factor for water and from the geometric error for particular samples. The absolute random error in the experiments, caused by the statistical nature of emission and absorption processes for gamma radiation, was 0.1% of the moisture content of the material. Estimates obtained as described in [11] show that, in the case of interest, the absolute error in the determination moisture content of the material can be written as

$$\Delta W_m(\%) = a \cdot W_m + b = 0.012W_m + 0.1 \quad (3)$$

Here, ΔW_m is the absolute error, %; a is the relative systematic inaccuracy; b is the absolute random inaccuracy, %; and W_m is the relative moisture content of the material, %.

Thus, in the present study the absolute error in determining the moisture content of the material with the gamma ray method at low moisture contents (about 1%) was about 0.1%. With increasing moisture content (to 50%) this error increased linearly to 0.7%.

3. Material structure

The pattern and intensity of moisture transfer processes in porous materials is significantly affected by the porous

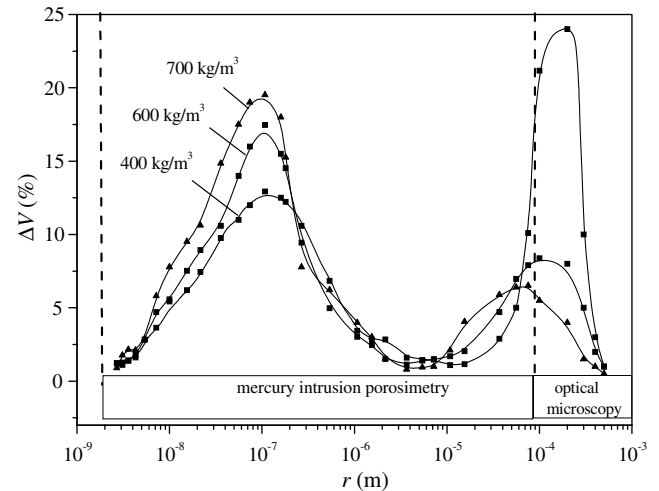


Fig. 2. Differential size distribution of pores.

structure of these materials. Several experimental methods were applied to characterize the particular concrete samples used in the present study in terms of their structural characteristics. The cellular concrete is a high-porosity heat-insulating constructional material which has gained, over the recent years, a wide utility in civil engineering.

Prior to conducting the moisture transfer study, differential distributions of pores over pore radius for cellular concrete samples of various porosities were obtained (Fig. 2). The distribution of large-sized pores was revealed by the optical method using optical microscopy. The distribution of small-sized pores was examined by mercury intrusion porosimetry.

The graphs in Fig. 2 shows that the differential distributions of pores in the concrete samples displayed, irrespective of the material density, two peaks. In other words, according to the presently adopted classification of porous materials [14], cellular concrete belongs to materials with a bimodal size distribution of pores. An analysis of data obtained showed that the porosity in cellular concrete mainly involved large cellular voids (10⁻⁴ m) and small-sized capillary pores (10⁻⁷ m) in inter-void porous walls. Thus, cellular concrete has an inter-related, conventionally closed porosity, with walls between large cellular voids being pierced by fine capillary pores.

4. Experimental results

The moisture transfer study was performed for cellular concrete samples under isothermal conditions ($T = 20$ °C) in two major damping conditions: capillary and sorption moistening.

4.1. Capillary moistening

In the capillary moistening regime the lower surface of the sample was in contact with water, whereas the upper surface of the sample during the experiment was in 30%

humid air, which value corresponded to an absolute air humidity of 3 g/m^3 . In the course of the experiment, distributions moisture content of the material over the height of the sample were measured following certain time intervals, to subsequently determine “spatial” moisture profiles (see Fig. 3). In this figure, plotted along the abscissa axis is the relative moisture content of the material, and along the ordinate axis, the coordinate of the cross-section at which the moisture content was determined. The spatial coordinate was reckoned from the lower surface of the sample. In several experiments, measurements moisture content of the material versus time at fixed cross-sections of the samples were carried out to obtain “temporal” moisture profiles (see Fig. 4); inspection of the latter profiles shows that the rate of material moistening varied over the height of the sample.

Thus, the experimental data show that the moisture transfer process during the capillary moistening proceeded rather actively. Following a 48 h period, the relative moisture content of the material reached values in excess of 50%. Probably, the latter can be attributed to the fact at high moisture content almost all pores in the material were filled with water so that the moisture transfer process could be intensified due to the action of capillary forces.

4.2. Sorption moistening

During the sorption moistening, the low surface of the sample was over the water surface, at 100% relative air humidity, which value at a temperature of $20 \text{ }^\circ\text{C}$ corresponded to an absolute air humidity of 10 g/m^3 . The upper surface of the sample was in the air chamber with almost zero relative air humidity. In the course of the experiments, profiles of moisture content of the material over the height of the sample were determined following certain time intervals (see Fig. 5) to subsequently determine the evolution

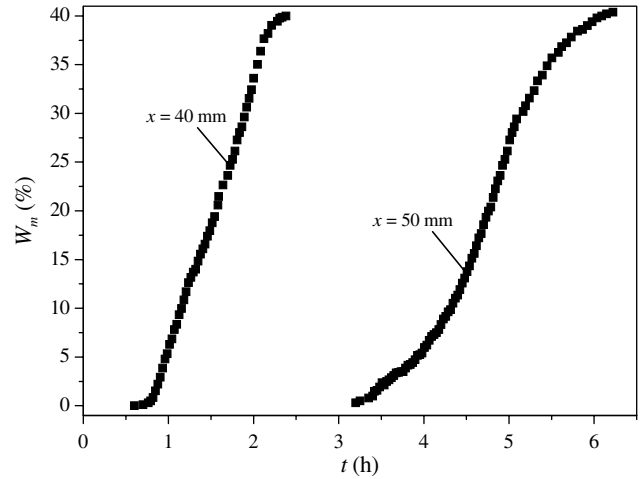


Fig. 4. Variation of moisture content of the porous material in time during capillary moistening.

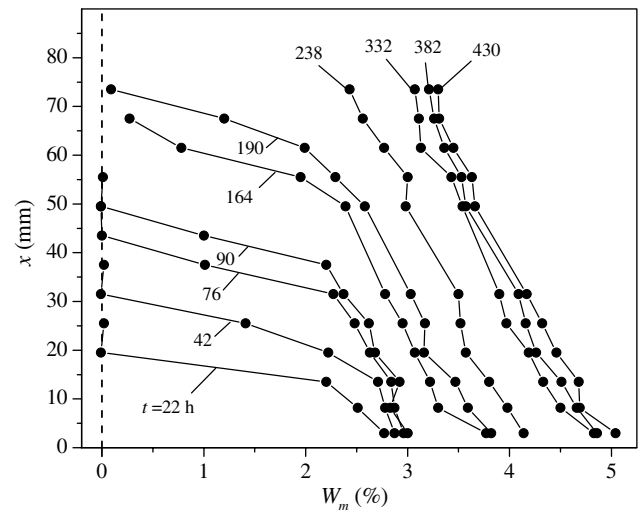


Fig. 5. Moisture profiles in the sample of porous material under sorption moistening.

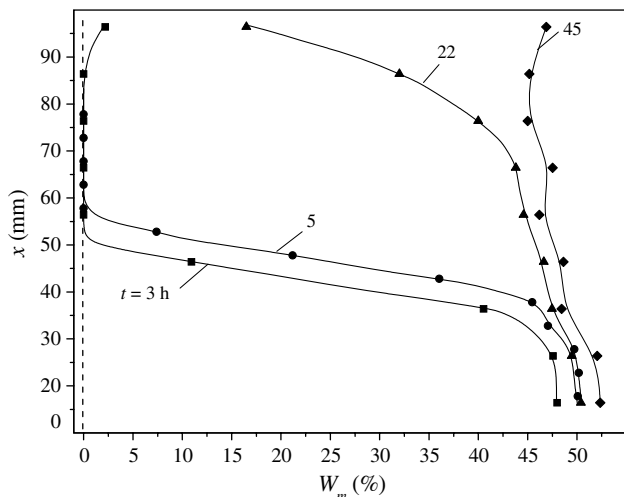


Fig. 3. Moisture profiles in the sample of porous material under capillary moistening.

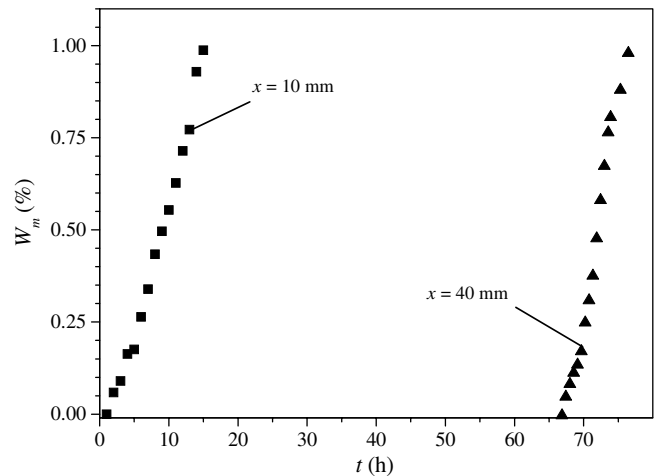


Fig. 6. Variation of moisture content of the porous material in time during sorption moistening.

moisture content of the material in time at fixed cross-sections of the sample (Fig. 6).

As it is seen from the graphs, during the sorption moistening gradual evolution of moisture content profiles in the sample was observed. A typical sorption moistening experiment lasted for more than two weeks, with the relative moisture content of the material reaching a value of 5% at the lower surface of the sample, which decreased towards at the upper surface to assume there a value of 4%.

Thus, the experimental data show that the moisture transfer processes in the cellular concrete samples under sorption moistening proceeded much less intensely compared to capillary moistening.

5. Determination of moisture diffusivity

In predicting humidity conditions in porous materials, the moisture diffusivity which defines the intensity of moisture transfer processes, can be conveniently used. Traditional procedures for experimental determination of the coefficient of moisture diffusion are complex and not accurate enough. In the present study, the moisture diffusivity was determined by processing experimental moisture profiles. To this end, the Boltzmann–Matano method was invoked based on the solution of time-dependent diffusion equation in one-dimensional statement [15]:

$$\frac{\partial W}{\partial t} = \frac{\partial}{\partial x} \left(D_w \cdot \frac{\partial W}{\partial x} \right) \tag{4}$$

Here, D_w is the effective coefficient of moisture diffusion in the material, m^2/s ; W is the moisture content of the material, kg/kg ; x is the spatial coordinate, m ; and t is time, s .

This method is based on using an automodel variable that allows relation (4) to be reduced to a differential equation in total derivatives. The application condition for this method is uniform initial moisture distribution in semi-bounded body. In addition, the sample must be sufficiently large so that the conditions of moisture transfer in a semi-bounded body are fulfilled. In the latter case, the initial and boundary conditions for Eq. (4) read:

$$\begin{aligned} &\text{for } 0 < x < \infty \text{ and } t = 0 : W = W_0 = 0, \\ &\text{for } x = 0 \text{ and } t \geq 0 : W = W_1, \partial W / \partial x = 0, \\ &\text{for } x \rightarrow \infty \text{ and } t \geq 0 : W = W_0, \partial W / \partial x = 0. \end{aligned} \tag{5}$$

To solve Eq. (4), we introduce a new automodel variable $S = x/t^{0.5}$, dependent on time and on the spatial coordinate. Substituting this variable into the diffusion equation and applying the Boltzmann transform, in Eq. (4) we pass over from partial to total derivatives, which procedure simplifies the solution of the equation:

$$-\frac{S}{2} \frac{dW}{dS} = \frac{d}{dS} \left(D_w \frac{dW}{dS} \right) \tag{6}$$

Here, the initial and boundary conditions assume the form

$$\begin{aligned} &\text{for } S = 0 : W = W_1; \\ &\text{for } S \rightarrow \infty : W = W_0, \quad \partial W / \partial S = 0 \end{aligned} \tag{7}$$

On some transformations, we obtain the following equation from which the coefficient of moisture diffusion in the material can be determined:

$$D_w(W) = -\frac{1}{2} \frac{1}{\frac{dW}{dS}} \int_{W_0}^W S dW. \tag{8}$$

As it is seen from (8), for determine the moisture diffusivity versus moisture content of the material, necessary to have moisture profiles plotted in automodel coordinates. Here, in accordance with the applicability conditions of the method, among all available moisture profiles only those profiles can be used which were taken at not too long times, with zero moisture concentration at the upper surface of the sample, so that the condition of semi-infinite body be fulfilled.

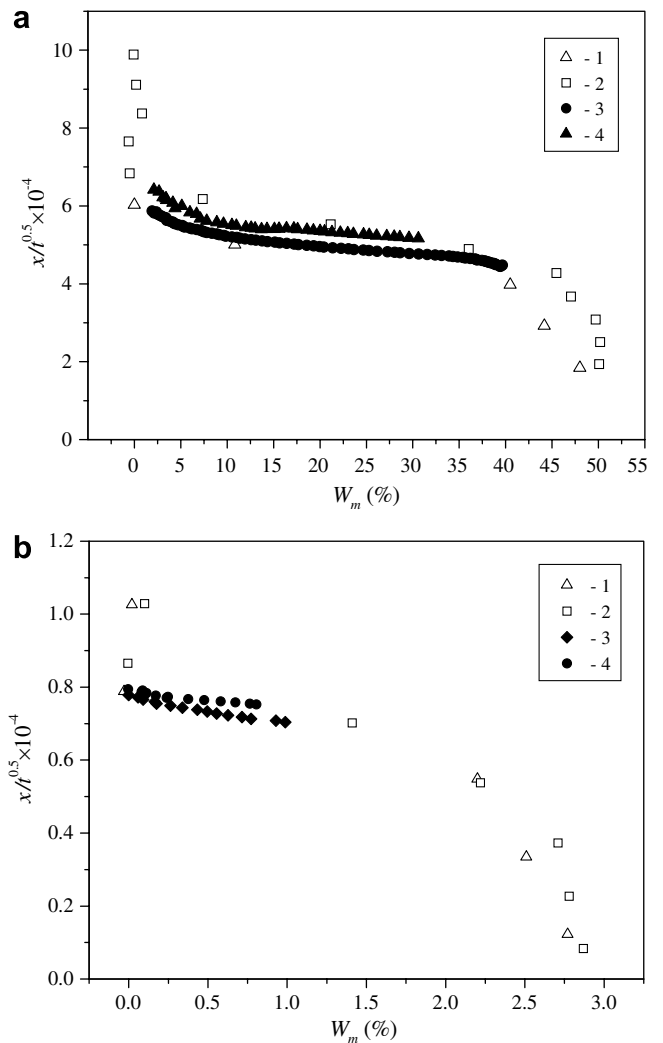


Fig. 7. Moisture profiles in cellular concrete (a) capillary moistening: 1, 2 – “spatial” profiles taken after 3 and 5 h; 3, 4 – “temporal” profiles taken at $x = 40$ and 50 mm (b) sorption moistening: 1, 2 – “spatial” profiles taken after 22 and 42 h; 3, 4 – “temporal” profiles taken at $x = 10$ and 40 mm.

Fig. 7 shows moisture profiles obtained in capillary and sorption moistening tests and plotted in automodel coordinates.

As it is seen from the graphs, in each moistening regime the “spatial” and “temporal” moisture profiles well correlate with each other if plotted in automodel coordinates. The latter proves that the adopted calculation procedure was adequate and could be used to process experimental data. The profiles plotted in automodel coordinates were processed by formula (8) to yield the moisture diffusivity as a function of moisture content of the material for cellular concrete samples with density 600 kg/m^3 under various moistening conditions at 20°C (see Fig. 8).

It is necessary to note that fact, that graphic dependence submitted in Fig. 8 reflects various moisture transport mechanisms, so that the moisture diffusivity must be considered as an effective characteristic. As it is seen from the graph, during the sorption moistening at low moisture content of the material, when the moisture transfer in the material is dominated by vapor transport, the coefficient of moisture diffusion has values of order $10^{-10} \text{ m}^2/\text{s}$. As the material becomes more and more moistened, a smaller amount of vapor becomes absorbed by pore walls, letting more vapor participate in moisture transfer processes. As a result, the coefficient of moisture diffusion increases to values of order $10^{-9} \text{ m}^2/\text{s}$ at a 3% relative moisture content of the material. As the moistening process proceeds further, the moisture diffusivity gradually increases since, although the role of vapor in the moisture transfer process diminishes, the contribution due to liquid moisture increases. During the capillary moistening, as the moisture content of the material increases to values in excess of 40% the moisture transport becomes controlled by liquid transfer. Here, the moisture transfer process gets intensified substantially due to the action of capillary forces. As a result, an appreciable growth of coefficient of moisture diffusion to values of $10^{-6} \text{ m}^2/\text{s}$ takes place.

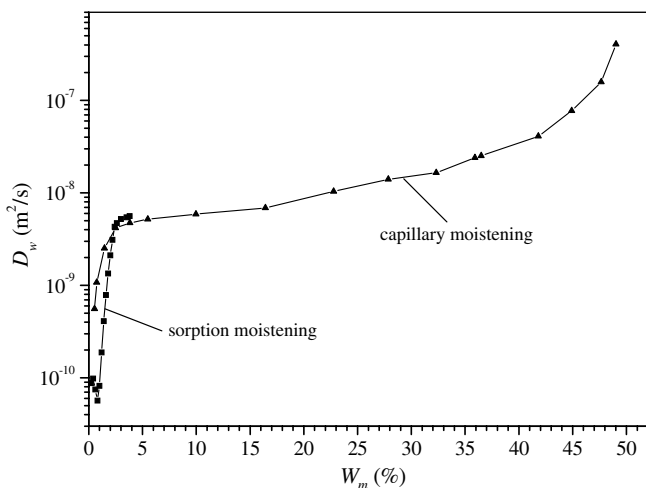


Fig. 8. Moisture diffusivity as a function of moisture content for cellular concrete under various moistening conditions.

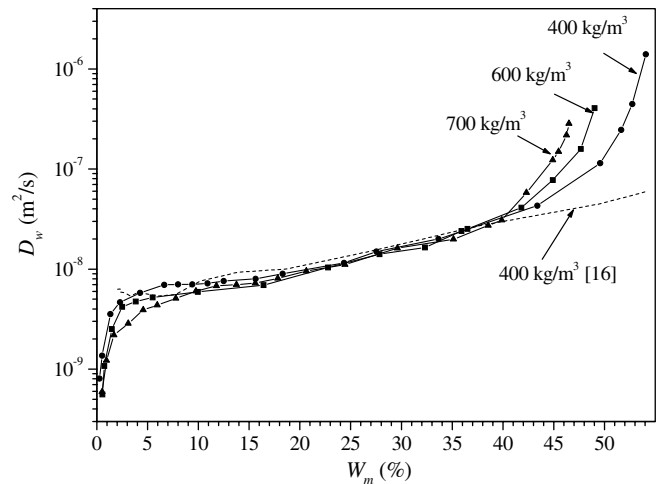


Fig. 9. Effective coefficient of moisture diffusion versus moisture content for concrete samples of various densities.

Using the gamma ray method, we experimentally examined moisture transfer processes in concrete samples of various densities. The data obtained were processed by the procedure described above. As a result, the effective coefficient of moisture diffusion was obtained as a function of moisture content of the concrete for samples with density 400, 600, and 700 kg/m^3 (see Fig. 9).

The obtained data prove that for cellular concrete samples of various densities in the moisture content range of 0–40% the curves of moisture diffusivity differ little from each other; yet, at higher moisture content the moisture diffusivity in dense concrete samples is higher. As noted above (see Fig. 2), the latter can be due to larger fraction of small-sized pores in the structure of dense samples compared to less dense samples. In Fig. 9, simulated data of [16] for cellular concrete samples with density 400 kg/m^3 are also shown, which fairly well agree with the present data obtained at moisture content of the material below 30%; at higher moisture content, however, our experimental data show a steeper rise of moisture diffusivity with moisture content of the material compared to the curve predicted in [16].

6. Conclusions

In the present study, the gamma ray method was used to examine moisture transfer processes in porous material. The data show that, with this method, the moisture content of the material can be profiled with high accuracy in a non-destructive manner. The advantageous feature is that the method permits high measurement sensitivity and short measurement time.

Using this method, we examined moisture diffusivity versus moisture content of the material in cellular concrete samples under sorption and capillary moistening. The coefficient of moisture diffusion was found to increase appreciably with increasing moisture content in the ranges of

low and high moisture content of the material, whereas in the range of moisture content from 5% to 30% only a slight increase of coefficient of moisture diffusion was observed.

Acknowledgement

This work was supported by the Russian Foundation for Basic Research (Grant No. 06-08-81003_Bel-a).

References

- [1] E.I. Tertchnik, Determination of moisture characteristics by the method of a sectional column, *J. Eng. Phys. Thermophys.* 8 (2) (1965) 247–250.
- [2] V.D. Ermolenko, A new method for determining the moisture diffusion coefficient of moist materials, *J. Eng. Phys. Thermophys.* 5 (10) (1962) 70–72.
- [3] H. Glaser, Simplified calculation of vapor diffusion through layered walls involving the formation of water and ice, *Kältetechnik* 10, H. 11 (1958) 358–364.
- [4] R.J. Gummerson, C. Hall, W.D. Hoff, Unsaturated water flow within porous materials observed by NMR imaging, *Nature* 281 (1979) 56–58.
- [5] L. Pel, H. Brocken, K. Kopinga, Determination of moisture diffusivity in porous media using moisture concentration profiles, *Int. J. Heat Mass Transfer* 39 (6) (1996) 1273–1280.
- [6] I.V. Koptug, L. Yu. Khitrina, Yu. I. Aristov, An ^1H NMR microimaging study of water vapor sorption by individual porous pellets, *J. Phys. Chem. B* 104 (2000) 1695–1700.
- [7] L. Pel, A.A.J. Ketelaars, O.C.G. Adan, Determination of moisture diffusivity in porous media using scanning neutron radiography, *Int. J. Heat Mass Transfer* 36 (1993) 1261–1267.
- [8] B.-X. Wang, Z.-H. Fang, Water absorption and measurement of the mass diffusivity in porous media, *Int. J. Heat Mass Transfer* 31 (1988) 251–257.
- [9] M.I. Nizovtsev, S.V. Stankus, A.N. Sterlyagov, V.I. Terekhov, R.A. Khairulin, Experimental determination of the diffusivities of moisture in porous materials in capillary and sorption moistening, *J. Eng. Phys. Thermophys.* 78 (1) (2005) 67–73.
- [10] M.I. Nizovtsev, S.V. Stankus, A.N. Sterlyagov, V.I. Terekhov, R.A. Khairulin, Experimental determination of moisture diffusivity of aerated concrete using gamma scanning methods, *Izvestia Vuzov. Stroitelstvo.* 4 (2002) 123–127 (in Rus).
- [11] A.S. Basin, Investigation of thermal properties of substances using the gamma ray method, in: *Thermophysical Properties Substances of Materials*, Izd. Standartov, Moscow, 1991, pp. 100–131 (in Rus).
- [12] M.A. Berliner, *Electrical Measurements, Automatic Monitoring, and Humidity Control*, Energiya, Moscow, 1965, pp. 173–178 (in Rus).
- [13] S.V. Stankus, R.A. Khairulin, A.V. Baginsky, Thermodynamic and transfer properties of liquid perfluorobenzene and perfluorotriethyl amine, *Thermophys. Aeromech.* 8 (2) (2001) 293–302.
- [14] V.B. Fenelonov, *Introduction to Physical Chemistry of Supramolecular Structure of Adsorbents and Catalysts*, Russ. Acad. Sci., Sib. Div., Novosibirsk, 2002, pp. 120–154 (in Rus).
- [15] B.S. Bokshstein, *Diffusion in Metals*, Metallurgiya, Moscow, 1978, pp. 27–31 (in Rus).
- [16] A.G. Perekhozhentsev, *Problems in the Theory and Prediction of Moisture State of Non-Uniform Enclosure Structures*, Volgograd, 1997, pp. 114–116 (in Rus).