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A neutron Radiography Study of the Movement of Thin Layers of Water in a Porous Building Brick

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Abstract: This study examined the isothermal water absorption within an internally produced gap in a brick sample. This investigation was conducted at the 14th beam line of the IBR-2 reactor, located at the Joint Institute of Nuclear Research in Dubna, Russia, utilizing a real-time neutron radiography station. The gap was created as a result of subjecting a brick sample to hydrophobic surface treatment. During the process of absorption, regular intervals were used to obtain images of neutron radiography. The water content, θ, and the water profiles θ(x,t) along the flow direction, x, were obtained from neutron radiography images. Additionally, the evolution of the waterfront position as a function of the absorption time, t, was determined. The second Fick's law of diffusion was used to discuss the results. It was demonstrated that the position of the waterfront followed approximately the $t^{1/2}$ -scaling and the θ (x,t) profiles in terms of the Boltzmann scaling ($\varphi = x/t^{1/2}$) converged to a universal one master curve at high absorption time. It was shown that the water absorption process showed anomalous behavior. Anomalous diffusion approaches based on the modification of Fick's law were used to model the results. It was demonstrated that the anomalous diffusion described the results better than the classical one. It was demonstrated that neutron radiography is an effective tool for probing the absorption of thin water layers.

Keywords: Neutron radiography, Diffusivity, Anomalous, Profiles.

1 Introduction

Moisture transport in construction building materials, such as masonry, is a crucial physical process for their service life and durability because water and biological and hazardous chemical substances transported by it can cause and induce a series of deterioration processes. Moisture transport depends on the pore size distribution and hydrophilicity of the porous matrix, in addition to the properties of the flowing liquid. As a result, capillary moisture absorption and spread in bricks and other building materials should be a top priority because they affect how long they last [1].

Water penetration and distribution in building materials is a topic that may be effectively studied with the use of neutron radiography. The basic concept of neutron radiography (NR) is the recording of a neutron beam through an object being studied using an appropriate detector, such as a CCD camera, following attenuation [2]. Water greatly attenuates thermal neutrons because hydrogen atoms have a large scattering cross-section. Since water attenuates neutrons ten times more than some building materials [3], neutron radiography (NR) offers an effective and useful technique for studying the distribution and transport of moisture in porous media, including building materials. NR

radiography's raw data are two-dimensional images. They include qualitative and quantitative information regarding the investigated object's composition.

The moisture absorption of three distinct types of bricks was studied [4] using NR. Brick sample drying profiles and moisture diffusivities were determined using NR [5]. The distribution of moisture in a few brick samples using NR and determined transfer coefficients was carried out [2]. The impregnation of some hydrophobic agents in clay bricks was determined using NR [6]. NR was employed to study moisture absorption in bricks [7-9]. Diffusivities were determined based on the results. Experiments using NR were carried out [10] to investigate H_2O diffusion in D2O-saturated clay brick at temperatures ranging from 30 to 50 °C. NR was used to study the drying of kaolin clay and quartz sand samples [11,12]. Other NR applications include studying the fundamental aspects of the durability of cement-based materials and hydrogen-rich fluids in geomaterials (rocks and soils) and engineered porous media (bricks and ceramics, concrete, fuel cells, heat pipes, and porous glass) [13,14]. A recent overview of neutron

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imaging applications in geomechanics was presented [15].

Water absorption reduction is a critical factor in extending the service life of numerous building materials. Surface treatment with hydrophobic agents is considered an important method among others. The addition of a surface hydrophobic modifier results in a remarkable reduction in water absorption [16,17]. The application of a hydrophobic coating to brick and geopolymer samples resulted in the formation of two distinct layers. It was demonstrated that when a hydrophobic coat was applied to the surface of brick and geopolymer samples, the coat created two layers, the first being an internal thin layer on the sample's exterior surface, and the second being a jacket around the thin film layer. Water is absorbed in the gap between the two layers by capillary action [18,19].

To the best of the author's knowledge, no research has been carried out on determining the diffusivity of a thin layer of water absorption and distribution in artificial gaps created on porous samples.

The goal of this study is to show that Fick's second law of diffusion can be used to model the movement of a thin layer that is absorbed in gaps that are artificially made on porous samples. The real-time neutron radiography station at the 14th beam line of the IBR-2 reactor at the Joint Institute of Nuclear Research, Dubna, Russia was utilized to conduct an experiment on thin layer water absorption and distribution in gaps generated artificially on porous samples.

2 Theories

The macroscopic theory of flow in porous media has started in soil science by the empirical Darcy law, which was extended to the unsaturated flow. It was applied to describe water (moisture) flow in building materials. Moisture transport in unsaturated porous media including construction building materials (in one dimension) is described by Fick's law (Dracy's law) of diffusion [20], x (neglecting gravity) given by

$$q(\theta) = -D(\theta)\frac{\partial\theta}{\partial x}, \qquad 1$$

where θ is the volumetric moisture content (cm^3 /cm^3) , $q(\theta)$ is the flux of moisture (cm/s) and $D(\theta)$ is the moisture diffusivity (cm^2/s). The diffusivity is given by

$$D(\theta) = K(\theta)\frac{\partial\psi}{\partial\theta}, \qquad 2$$

where $K(\theta)$ (cm/s) and ψ (cm) are the hydraulic conductivity or permeability and the moisture potential, respectively. The diffusivity is a function of the porous geometry and the physical parameters characterizing flowing liquid (viscosity, surface tension, and contact angle) [20,21]. The continuity equation is given by

$$\frac{\partial\theta}{\partial t} = -\frac{\partial q}{\partial x}, \qquad 3$$

where t is the water absorption time. From both equations 1 and 2, the following differential diffusion is obtained:

$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial x}\left(D(\theta)\frac{\partial\theta}{\partial x}\right), \qquad 4$$

It usually describes the moisture transport in unsaturated porous media.
Assuming that θ depends on the single variable ϕ - the so-called Boltzmann variable

$$\phi = \frac{x}{\sqrt{t}}, \qquad 5$$

where x is ordinate and t is the absorption time. This assumption is reasonable at least when the waterfront position – let us name it x_m - is proportional to the square root of time. By substituting ϕ into equation 4 and using the boundary conditions:

$$\theta = \theta_i \qquad x > 0 \qquad t = 0$$
$$\theta = \theta_s \qquad x = 0 \qquad t \geq 0 \qquad 6$$

where θ_i is the initial water content of the porous material (for initially dry material $\theta_i \approx 0$) and θ_s is the saturation water content, the equation reduces to the ordinary differential equation

$$-\frac{\phi}{2}\frac{d\theta}{d\phi} = \frac{d}{d\phi}\left(D(\theta)\ \frac{d\theta}{d\phi}\right) \qquad 7$$

Integration of equation 7, with the conditions

$$\theta = \theta_s \qquad \phi = 0,$$
$$\theta = \theta_i \qquad \phi \to \infty \qquad 8$$

yields the diffusivity [9]

$$D(\theta) = -\frac{1}{2}\frac{d\phi}{d\theta}\int_0^{\theta}\phi\ d\theta \qquad 9$$

If the function $\theta(\phi)$ is known from the experiment, which is our case, one can determine the diffusivity function $D(\theta)$. The shape of the $D(\theta)$ -function depends strongly on the experimental function $\theta(\phi)$.

Some authors [9, 22.23] found it is necessary to go beyond the Darcy relation between the volumetric moisture flow q and the moisture gradient (1), assuming what follows

$$q = -D(\theta)\left(\left|\frac{\partial\theta}{\partial x}\right|\right)^{\alpha} \qquad 10$$

where α is a real number ($\alpha = 1$ corresponds to the Darcy or Fick law). when the waterfront does not scale with $t^{1/2}$, the water absorption process becomes anomalous. In such a case, the non-linear diffusion equation

$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial x}\left(D(\theta)\left(\left|\frac{\partial\theta}{\partial x}\right|\right)^{\alpha}\right) \qquad 11$$

can be used, where α is a real number ($\alpha = 1$ corresponds to Darcy or Fick's law). The so-called Generalized Boltzmann variable, γ

$$\gamma = \frac{x}{t^n}, \qquad 12$$

with the index n being a real number, replacing that given by equation 4. Again, treating θ as a quantity known from the experiment, we can solve Eq. 10 for the diffusivity. Assuming that θ depends on the single variable γ - (this assumption is reasonable at least when the waterfront position x_m is proportional to the imbibition time to the power n, with the index n being a real number), one finally obtains the following form of the diffusivity

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$$D(\theta,t) = -n\ t^{(1+\alpha)n-1}\left|\frac{d\gamma}{d\theta}\right|^{\alpha}\int_0^{\theta}\gamma\ d\theta \qquad 13$$

When α = 1/n - 1 [9, 22], the anomalous diffusivity $D_a(\theta)$ which is only a function of the water content θ is obtained as

$$D_a(\theta) = -\frac{1}{1+\alpha}\left|\frac{d\gamma}{d\theta}\right|\int_0^{\theta}\gamma\ d\theta\ , \qquad 14$$

Additionally, another function of diffusivity can be obtained when α =1 [23]. In such cases, the diffusivity depends on both the moisture content θ and the absorption time:

$$D(\theta) = -nt^{2n-1}\left|\frac{d\gamma}{d\theta}\right|\int_0^{\theta}\gamma\ d\theta, \qquad 15$$

The classic time-independent form of the diffusivity (eq. 9) corresponds to $\alpha = 1$ and *n=1/2* in eq. 13.

3 Experimental details

The NR and tomography station installed at the 14th beam line of the high flux pulsed reactor IBR-2, Joint Institute for Nuclear Research (JINR), Dubna, Russa [24,25] was used to experiment. The neutron flux at the sample place is $5.5\text{x}10^6$ n/cm^2/s^{-1}. A neutron beam measuring 20 × 20 cm is generated through a system of collimators, with the characteristic parameter L/D ranging from 200 to 2000. The detector system consists of a ^{6}LiF/ZnS scintillation screen and a CCD camera. It is utilized for image recording, with an approximate average spatial image resolution of 300 µm. The process of obtaining images and conducting additional analysis is executed utilizing the Image J software [26]

The process of water infiltration into the gap formed by a brick sample was examined using NR. A polymer-based waterproofing slurry was used to coat the external surfaces of the sample. Following the application of distilled water to moisten the samples, a rigid brush was utilized to provide two layers of the coat perpendicularly. The samples are subsequently dried for several additional days. The polymeric waterproofing slurry was eliminated from both the upper and lower surfaces of the samples. The samples were positioned in a vertical orientation and their ends were submerged in a container of water. The water level in the container submerges approximately 3 millimeters of the sample ends. Many NR images were continuously recorded throughout the process of moisture absorption. From the obtained NR images recorded at the start and end of the water absorption process, it was noted that the water level remained constant. Furthermore, the quantity of water absorbed is insignificant when compared to the overall amount present in the container.

4 Results and Discussion

The NR images were acquired during the water absorption process. Fig. 1 shows some selected images for the brick samples investigated at different absorption times. The water-filled portion of the investigated samples is darker than the dry ones. The images shown in Fig.1 are obtained as follows: dark current images (background images) in the absence of the incident neutron beam, images for the incident neutron beam (without the sample), and images for the samples absorbing water during the incident neutron beam were recorded. Subtracting dark current images from the registered NR images was carried out to obtain images free from the background. These images were normalized concerning an image for the incident neutron beam.

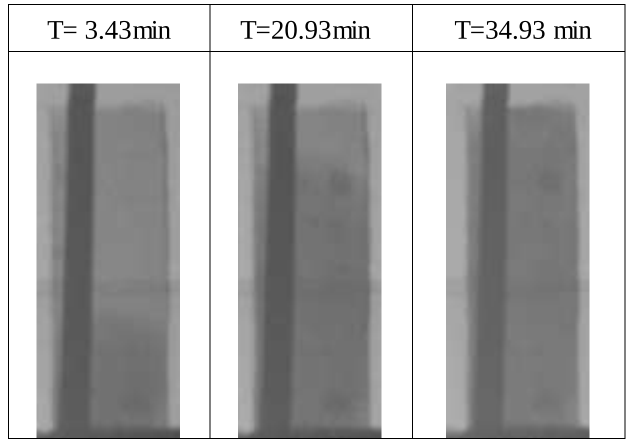

Fig. 1: Some selected neutron radiography images.

The water content (volumetric fraction of water, cm^3/cm^3) distributions along the flow direction (x) as a function of the absorption time and corrected for neutron scattering were extracted from the acquired NR images (simply water profiles, $\theta(x,t)$) according to

$$\theta(x,t) = \frac{1}{L\Sigma_{water}} ln\left(\frac{I_{dry}-I_{sca1}}{I_{dry+wetted}-I_{sca2}}\right), \qquad 16$$

where $\sum_{water}$ is the effective macroscopic cross section for water (0.33 mm^{-1}), L is the sample thickness (cm), I_{dry} and $I_{dry+wetted}$ are total transmitted neutron fluxes (intensities or brightness) (n.cm^{-2}.$^{-1}$s) for the dry and wetted sample, respectively, and (n.cm^{-2}.$^{-1}$s) are scattered neutron components for the dry and wetted sample, respectively. For determination I_{sca2}, a neutron-absorbing shield (Cd strip) is placed perpendicularly to the neutron beam axis in front of the object under investigation (brick samples). The strip absorbs almost all of the incident thermal neutrons and the response of the imaging device behind the strip corresponds to contributions of the scattered neutrons only. The scattered neutron component can be determined and subtracted from the total intensity of neutrons outgoing from the sample, to give the intensity of directly transmitted neutrons. These procedures were used in this work to determine both I_{sca1} and hence to correct for the neutron scattering effect during moisture absorption in the sample investigated. The Cd strips are shown in black color in the NR images obtained (Fig. 1). The Image J software [26] was used to draw two rectangular areas along the flow direction for every image obtained. One area is beside and

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the other is on the Cd strip. The neutron intensities extracted from the dry and wetted sample images altogether with Eq.16 were used to determine the water profiles, $\theta(x,t)$. The profiles determined for the sample investigated are shown in Figs. 2a and b.

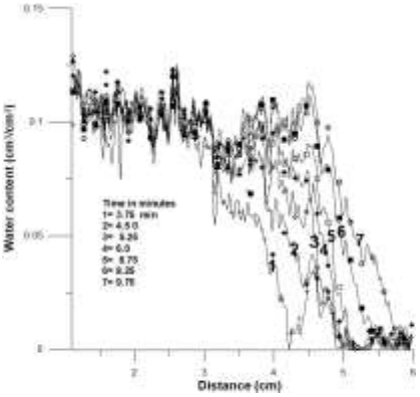

Fig. 2a: $\theta(x,t)$ for the sample investigated.

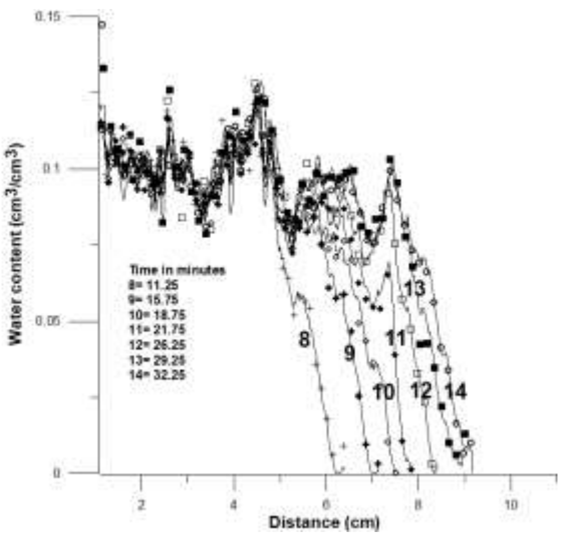

Fig 2b: $\theta(x,t)$ for the sample investigated.

The water front positions for the water absorption processes in the sample investigated in terms of the absorption times, t were determined from the water profiles and the results are shown in Fig.3. As one can see, the water front positions follow approximately the $t^{1/2}$ scaling. The results were fitted with straight line equation and the slope (capillary penetration coefficient) of the fit line was found to be 1.27 $cm.min^{-0.5}$.

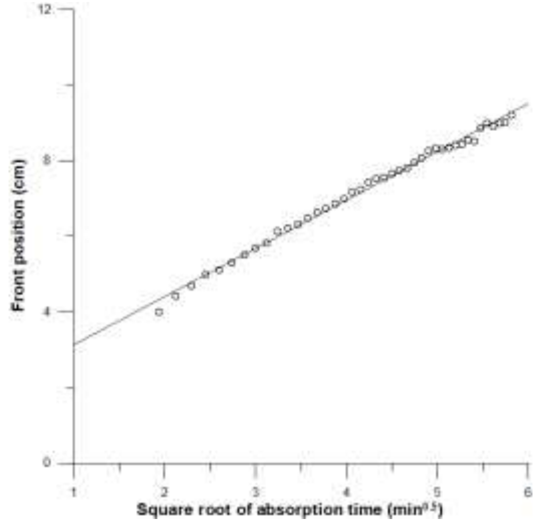

Fig. 3 : Waterfront position versus $t^{1/2}$.

Diffusion-based approaches using Fick's second law were used to model the unsaturated water flow processes in the investigated sample. A motivation for applying such a theory is the waterfront positions followed $\sqrt{t}$-scaling (Fig.3) . The results of the water profiles were replotted against the Boltzmann variable, $\phi =$, and shown in Figs. 4. As one can see, the profiles θ - ϕ partially collapse to single master curves at all absorption times (Figs. 4a and b), however, at high absorption times there is better collapse (Figs. 4b). This means that Fick's second law of diffusion can not be used to model the whole water absorption process in the artificially created gap. However, it can be applied at the highest water absorption times.

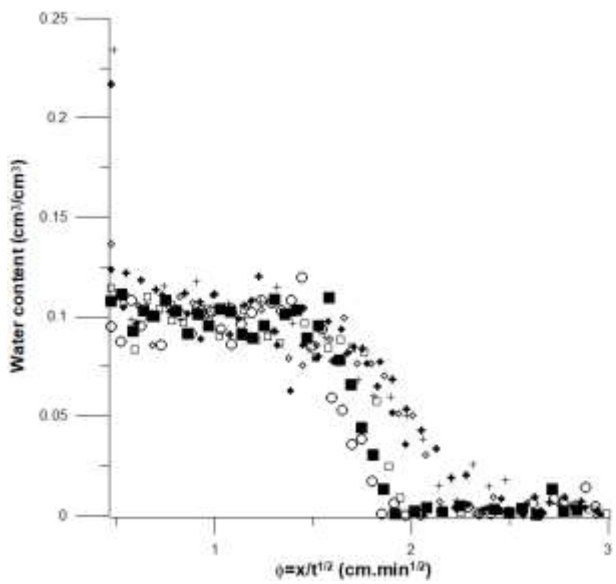

Fig.4a: Master curves of the θ - ϕ profiles (Fig.2a).

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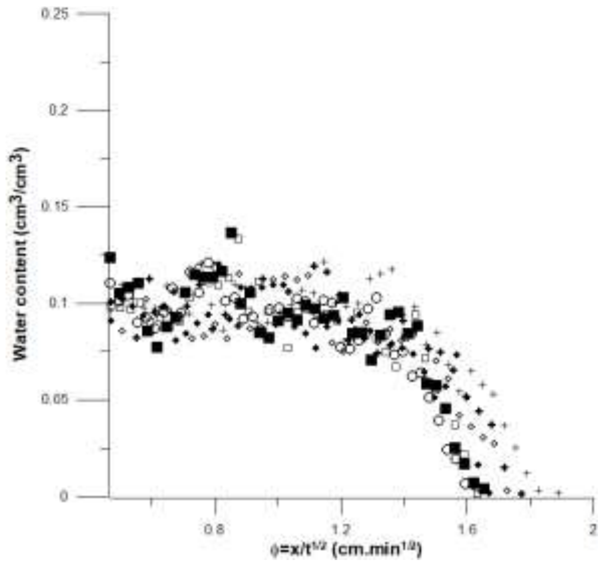

Fig.4b: Master curves of the θ - ϕ profiles (Fig.2b).

Since, the water front positions slightly deviate from the $\sqrt{t}$-scaling (the fit line- Fig. 3 did not pass through the origin), the anomalous diffusion approach describing the water absorption process inside the gap was checked in this work. The water front positions were fitted using the power law equation (Eq. 12) and the results are shown in Fig. 5. The exponent n obtained from the fit line was found to be 0.36. It means the diffusion process in the gap is sub-diffusive (n<0.5). In terms of the generalized Boltzmann variable, γ and following the same procedures for determining the θ - ϕ profiles, the $\theta - \gamma$ profiles were obtained. The results are shown in Figs.6a and b. The convergence of the $\theta - \gamma$ profiles is better than that for the θ - ϕ profiles. It means that the abnormal diffusion approaches described successfully the present results better than the normal diffusion. Water diffusivities can be determined from either the θ - ϕ or $\theta - \gamma$ profiles - it is beyond the scope of the present work

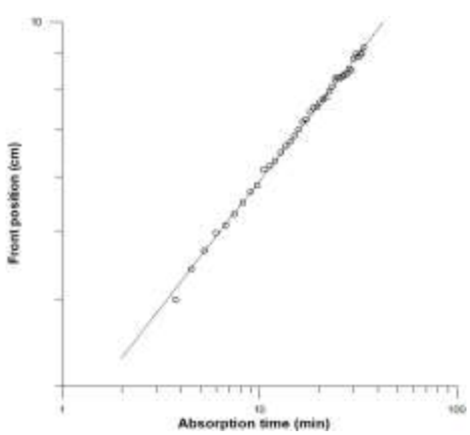

Fig. 5: Water front position versus absorption time along with a power law fit line.

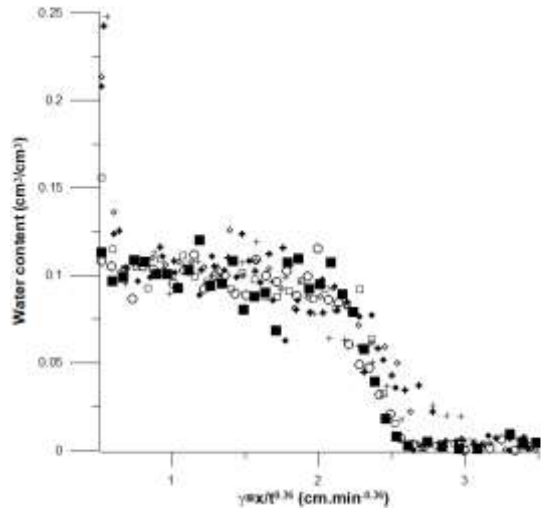

Fig.6a: Master curves of the $\theta - \gamma$ profiles (Fig.2a).

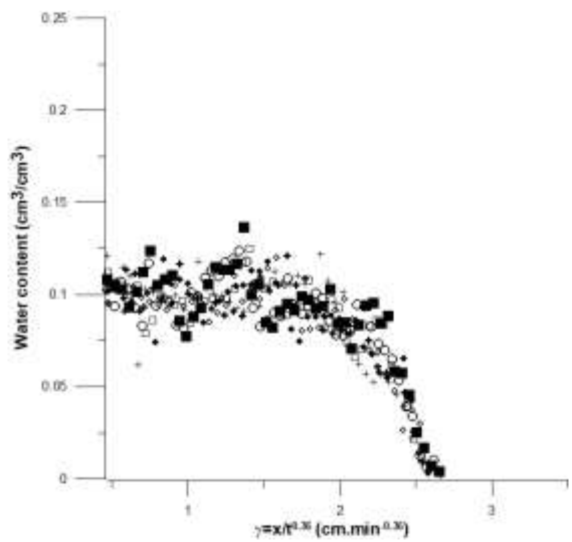

Fig.6b: Master curves of the $\theta - \gamma$ profiles (Fig.2b).

4 Conclusions

Neutron radiography was used to record water absorption in a gap created as a result of subjecting a brick sample to hydrophobic surface treatment. The profiles θ(x,t) along the flow direction, x, were obtained from neutron radiography images. The progress of the waterfront position as a function of the absorption time, t, was determined. The second Fick's law of diffusion was used to discuss the results. It was shown that the position of the waterfront followed approximately the $t^{1/2}$ -scaling and the θ (x,t) profiles in terms of the Boltzmann scaling ($\varphi = x/t^{1/2}$) converged to a universal one master curve at high absorption time. The water absorption process showed anomalous behaviour. Anomalous diffusion approaches based on the modification of Fick's law were used to model the results. It was demonstrated that the anomalous

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