Determination of moisture diffusivity in porous media using scanning neutron radiography

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Abstract—A technique for measuring moisture concentration profiles based on scanning neutron radiography is presented. With this technique moisture concentration profiles have been measured during drying of brick and kaolin clay. The isothermal moisture diffusivity as a function of moisture content could be determined directly from these profiles. An error analysis shows that the minimum spatial resolution for scanning should be 0.5 mm. If this condition is not satisfied, it is not possible, for example, to detect the minimum in the moisture diffusivity in the case of a receding drying front.

1. INTRODUCTION

MODELLING heat and mass transfer in porous media is an area of continuing research in various disciplines, e.g. chemical engineering, civil engineering and soil science. For describing heat and mass transport in such media a model at the macroscopic scale is necessary, allowing the porous material to be looked upon as a continuum. The basic equations for heat and mass transfer at this level were first established by Philip and de Vries [1]. Various authors, e.g. refs. [2, 3] have tried to derive this description at the macroscopic scale by means of applying the volume averaging technique to the microscopic equations. Although insight into the problem improved as a result of this, it is not yet possible to derive the equations of Philip and de Vries in a completely satisfactory way. Pragmatically, however, these equations are most frequently used, even though from a theoretical point of view some doubts still remain.

Once one is willing to accept the description of Philip and de Vries, a major problem is the determination of the various coefficients, since these coefficients depend strongly on moisture content and temperature. Very often overall experimental techniques are used which only give the average moisture content as a function of time (e.g. drying curves) [4]. This has the major disadvantage that some relation between the coefficients and moisture content has to be assumed *in advance* in order to derive these coefficients from the experiments. However, which relation is appropriate (e.g. exponential, power law, polynomial) is not known beforehand. As a matter of fact, obtaining such a relation was the main objective of the experiments. For determining the moisture diffusivity as a function of moisture content without having to assume such a relation, the experimental determination of the moisture concentration profile as a function of time would be advantageous. If such data are available with sufficient accuracy, the moisture diffusivity can be determined as a function of moisture content directly.

The classical way for obtaining moisture concentration profiles is the gravimetric method. Although this is the most direct method for determining an overall moisture content, it has two major disadvantages regarding moisture concentration profiles. First of all it is a destructive method and therefore every profile has to be determined using different samples. The fact that one is not always able to exactly duplicate sample preparation and experimental history often interferes [5]. On top of this, the method is laborious and time consuming. Secondly, the method is not accurate in its spatial resolution. Slices which can be cut are of the order of 0.5–1 cm, depending on the type of material. The moisture content which is determined is the average over such a slice. Especially when steep gradients occur, as for example with a receding drying front, this method will fail to determine this.

Apparently, an experimental method for determining transient moisture concentration profiles

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NOMENCLATURE

d	thickness of sample [m]	
d_i	equivalent thickness for component i [m]	
D_{θ}	isothermal moisture diffusivity [m^2 s^{-1}]	
I	intensity of neutron beam [m^{-2} s^{-1}]	
I_0	initial intensity of neutron beam [m^{-2} s^{-1}]	
l	height of the sample [m]	
t	time [s]	
T	temperature [°C]	
x	spatial coordinate [m].	

Greek symbols

θ moisture content [m^3 m^{-3}]

μ_i macroscopic attenuation coefficient of component i [m^{-1}].

Subscripts

mat   dry material

w   water.

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should be non-destructive and give a high spatial resolution. In the present study moisture concentration profiles were measured using scanning neutron radiography in drying experiments on bricks and clays. From these profiles moisture diffusivity was determined directly.

2. NEUTRON RADIOGRAPHY

2.1. General characteristics

When a beam of neutrons passes through a material, the neutrons will interact with the nuclei of this material (unlike γ-rays, which interact with the electrons). The attenuation of the neutron beam is determined by the cross-section for scattering and absorption [6] of the nuclei present in the sample. Because of the relatively large scattering cross-section of hydrogen, this transmission method is very sensitive for water. Unlike NMRI, no distinction can be made between water that is chemically bound or physically bound.

The intensity I of a neutron beam after passing a sample is:

$$I = I_0 \exp\left(-\sum_i d_i\mu_i\right), \qquad (1)$$

in which μ_i is a macroscopic attenuation coefficient of component i and d_i is the equivalent thickness for this component. For a rigid non-shrinking material with a volumetric moisture content θ and thickness d, equation (1) reduces to:

$$I = I_0 \exp\left[-d(\mu_{mat} + \theta\mu_w)\right]. \qquad (2)$$

μ_w and μ_{mat} are determined independently by meas-

Table 1. Macroscopic attenuation coefficients for various materials as measured using neutrons with a wavelength of 1.31 Å

Type of material	μ (cm^{-1})
Water	2.54
Brick	0.155
Kaolin clay	0.67
Aluminium	0.10

uring the transmission for pure water and dry material, respectively. In Table 1 coefficients are given for the materials used in this study. The moisture content can be determined by measuring the transmission I through a sample of given thickness d.

2.2. Experimental set-up

The experiments described in the present study were performed at the 2 MW reactor of the Interfacultair Reactor Instituut of the Delft University of Technology, The Netherlands. The experimental set-up is shown in Fig. 1. For the experiments a monochromatic neutron beam was used with a wavelength of 1.31 Å (corresponding to an energy of 47.7 MeV), which was selected by means of (002) reflection at a zinc crystal. To get a high spatial resolution a narrow neutron beam, with typical dimensions of 1 × 30 mm, was made with a collimator made up of a combination of boron and cadmium. This beam has an intensity of 3.8×10^8 m^{-2} s^{-1}. After passing the sample the neutrons were detected by a ^{3}He proportional detector. This detector has a very high detection efficiency for thermal neutrons and a low one for gamma rays. To correct for the divergence of the neutron beam and to discriminate against multiple scattered neutrons a second collimator, identical to the first, was placed in front of the detector.

In the present study, drying experiments were performed to determine the moisture diffusivity. The sample was placed in an aluminium container with an open top. Dry air was blown over the sample, thus
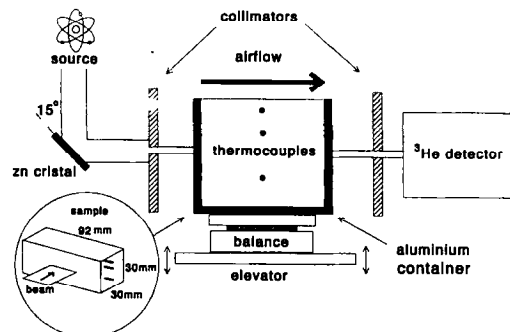

Fig. 1. Experimental set-up.

creating a one-dimensional drying experiment. To get the spatial distribution of the moisture content the sample was moved through the beam by an elevator. Thermocouples were inserted in the sample to monitor the temperature evolution in the course of drying. The sample was placed on a balance in order to compare the overall weight loss with the one obtained from the integral of the moisture profiles.

3. EXPERIMENTAL RESULTS

First of all, the experimental installation (beam profile and performance of the elevator) was checked by moving a Plexiglas cube (containing an abundant amount of hydrogen) with small steps into the neutron beam. The result is shown in Fig. 2. In this case the neutron beam has an effective height of 0.7 mm. The straight line indicates that the intensity of the beam is uniform. If this was not the case, the determined moisture content is not necessarily identical to the average moisture content of the measuring volume.

Drying experiments were conducted with two different types of porous material: machine moulded clay brick and kaolin clay. The initial moisture content of the kaolin clay was chosen such that no shrinkage occurred during drying. A non-uniform grid was used in determining the spatial distribution of the moisture content. Since the steepest profiles are expected to occur near the drying surface, a grid was used ranging from 0.5 mm near the surface to 2 mm at the bottom of the sample. Measuring the moisture content at a specific point with an inaccuracy of about 1% takes approximately 40 s. Measuring one entire moisture profile takes about 30 min. A moisture concentration profile at a specific time is deduced by interpolating

the moisture content as function of time for all points. The results for the two experiments, one for the brick and one for the kaolin clay, are given in Fig. 3.

There is clearly a difference in the evolution of the moisture concentration profile for these two materials. For the brick, moisture concentration profiles are rather flat for a long period of time in comparison to the profiles for kaolin clay, indicating that the moisture diffusivity for the brick must be much higher at the same moisture content. After approximately 10^4 s, the surface of the brick is completely dry and a receding drying front starts to develop. For kaolin clay no receding front is observed.

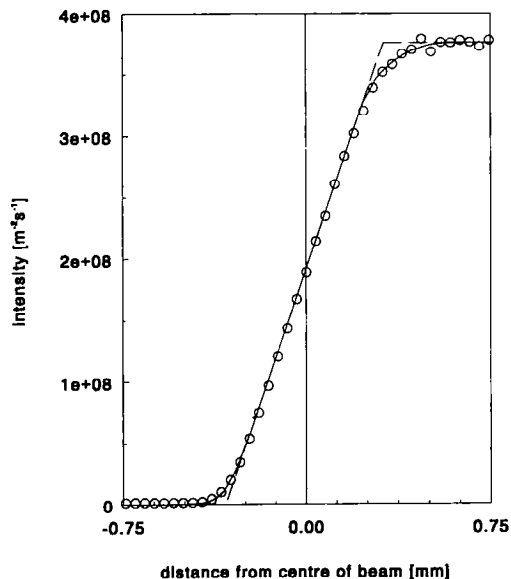From the integral of the measured moisture concentration profiles, the average moisture content of

Fig. 2. Cumulative intensity of the neutron beam as a function of its height; $x = 0$ is the centre of the beam. In this case the effective height of the beam is 0.7 mm.
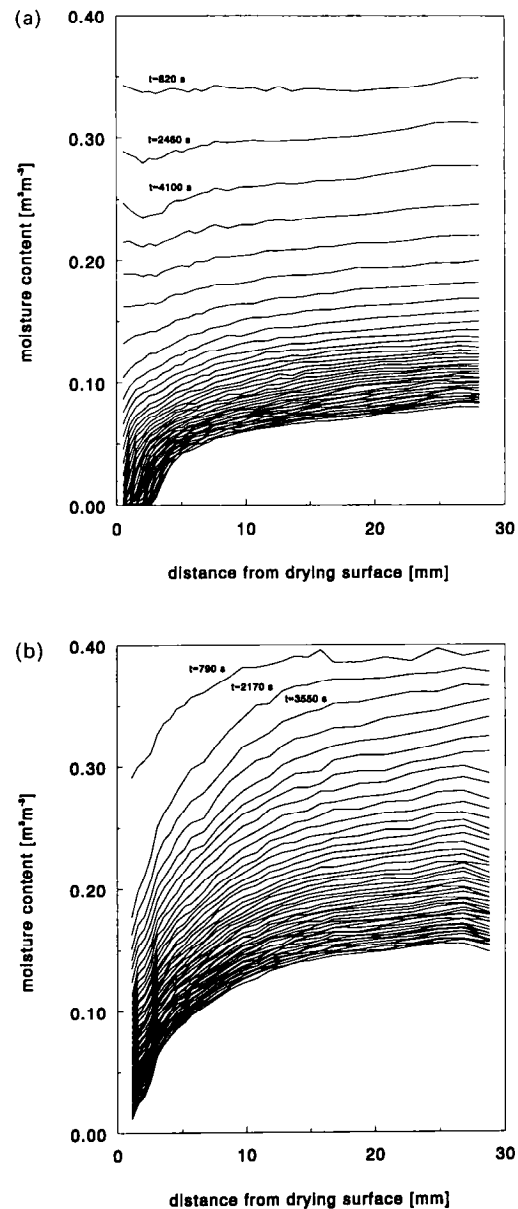
Fig. 3. Moisture concentration profiles for brick (a) and kaolin clay (b). The time between subsequent profiles is 1640 s for the brick and 1380 s for kaolin clay.

the sample as a function of time can be constructed. This curve can be compared with the one obtained directly from the weight loss of the sample. Results are given in Fig. 4 for the same experiments as in Fig. 3.

For both materials there is good agreement between the average moisture content determined from the experimental profiles and determined from the weight loss. The difference in this curve for the brick compared to kaolin clay reflects the already mentioned difference in the evolution of the profiles. For the brick, the point where a receding front starts entering the material (at 10^4 s) corresponds to the point where the drying rate starts to decrease rapidly. For kaolin

clay no receding drying front is observed and therefore the decrease in average moisture content is much smoother.

During the experiments it was not possible to obtain isothermal drying conditions with respect to the sample. Therefore, temperature profiles in the samples were measured during the experiment. The results are given in Fig. 5.

The temperature of both materials decreases rapidly at the beginning of the drying experiment, since heat is subtracted from the material due to drying. As a result of this, heat will start flowing toward the sample since the samples are fitted in an aluminium container. In this case the heat flow to the sample is a three-dimensional problem.
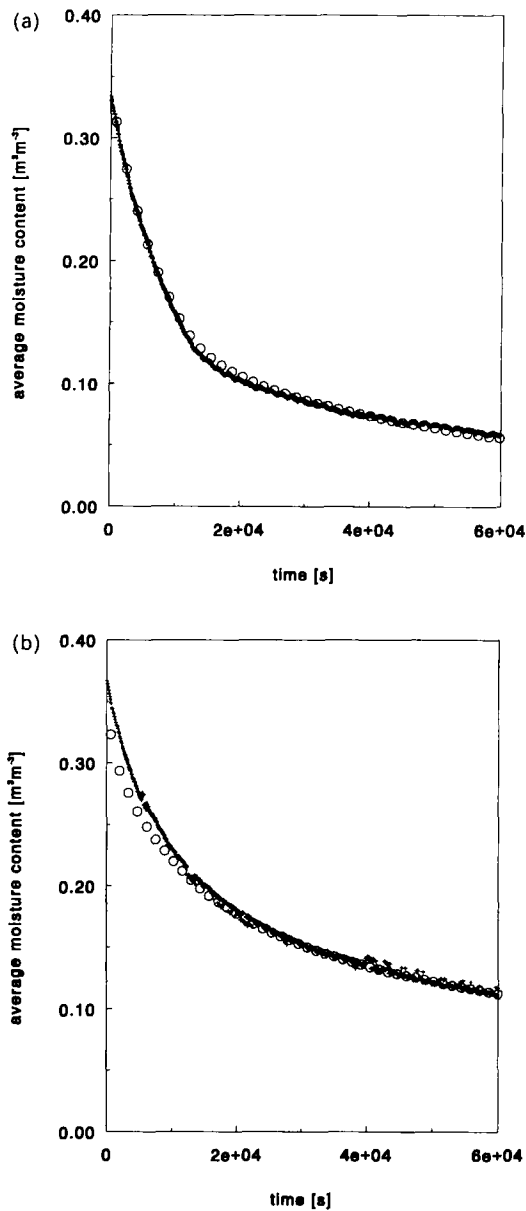

FIG. 4. Average moisture content as a function of time for a brick (a) and kaolin clay (b). (O) Determined from the integral of the moisture concentration profile. (+) Determined directly from the decrease of the weight of the sample.
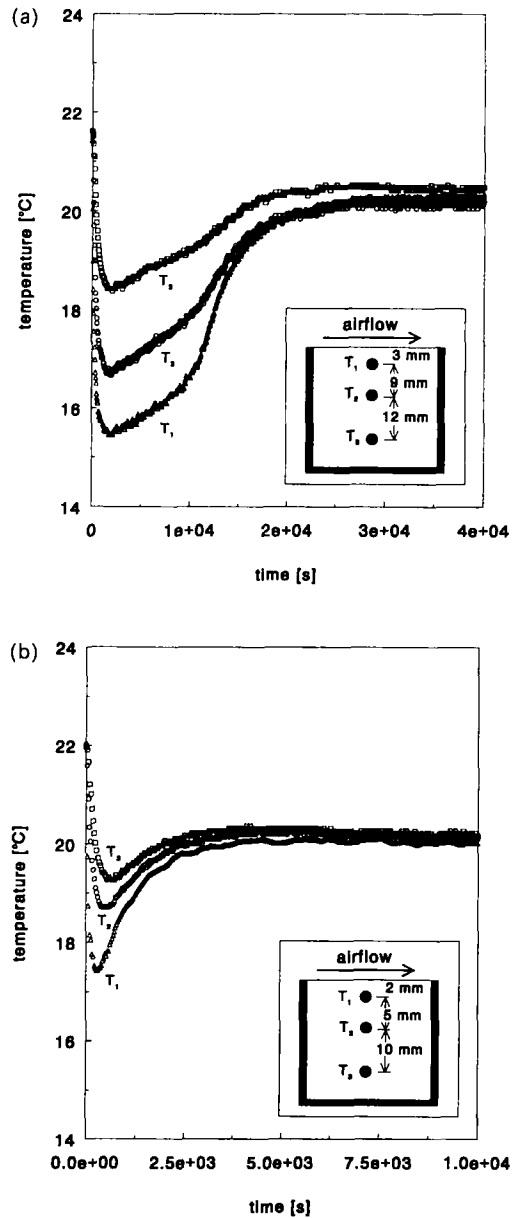

FIG. 5. Temperature distribution as a function of time for brick (a) and kaolin clay (b).

For the brick there is a temperature difference of 3–4°C in the sample during the first 10^4 s of the experiment. The temperature increases rapidly after this time, when the receding front starts entering the material. At this point the drying rate decreases considerably and so does the heat subtracted from the material; consequently, the maximum temperature difference decreases rapidly. From 2×10^4 s the drying process can be regarded as isothermal with respect to the sample. For kaolin clay the situation is somewhat different. Here, too, there is a rapid decrease of the temperature at the beginning of the experiment. However, the lowest value for the temperature at the surface has already been reached after 600 s. After 4×10^3 s the temperature gradient in the material has become very small and the experiment may be considered as isothermal. No sudden increase of the temperature is observed, which is in agreement with the fact that no receding drying front is present.

4. MOISTURE DIFFUSIVITY

As discussed in the preceding section, the experiments can be regarded as isothermal some time after the start of drying (for the brick after 2×10^4 s and for kaolin clay after 4×10^3 s). In this isothermal region the coupled set of equations for heat and mass transfer reduces to a single partial differential equation describing the transport of moisture with the gradient in moisture content as the driving force. For the one-dimensional problem encountered during the experiments and neglecting the influence of gravity, this equation reads:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x}\left(D_\theta \frac{\partial \theta}{\partial x}\right). \tag{3}$$

In general, the moisture diffusivity D_θ is a function of moisture content. With the help of equation (3) and the experimental moisture concentration profiles, it is possible to derive the dependency of the moisture diffusivity with moisture content. For this, equation (3) is written as:

$$D_\theta = \frac{\displaystyle\int_1^{x'} \frac{\partial \theta}{\partial t}\,dx}{\left.\dfrac{\partial \theta}{\partial x}\right|_{x'}}. \tag{4}$$

In this equation, use has been made of the fact that the partial derivative of θ with respect to place is zero at the vapour tight bottom ($x' = 1$) of the sample. Note that this condition is confirmed experimentally, as can be seen from the experimental moisture concentration profiles. Results for the moisture diffusivity as a function of moisture content are given in Fig. 6.

For the brick, the results of two experiments are combined: the experiment discussed here and an additional one under identical external conditions using a different brick (of the same type and batch,
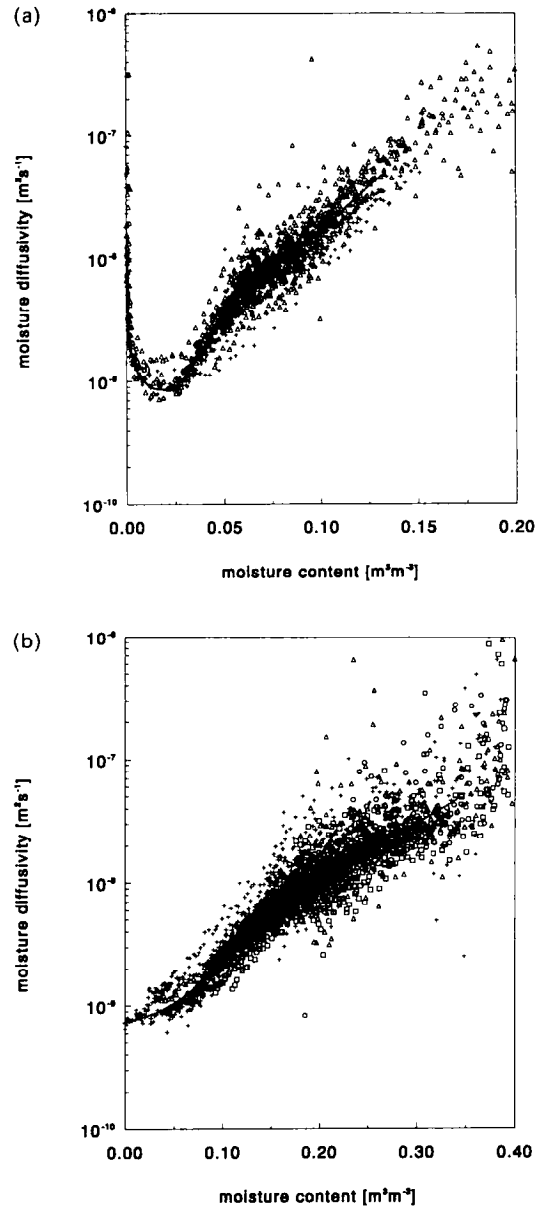
Fig. 6. Moisture diffusivity as a function of moisture content for brick (a) and kaolin clay (b). (+) Experiment discussed in this study. (O, □ and △) Other experiments under identical conditions. (——) Guide to the eye. The figure for the brick contains approximately 1000 data points and for kaolin clay approximately 3000.

however). For kaolin clay, the results are from four experiments with different samples under identical external conditions, of which one was discussed in the preceding section. In both cases the moisture diffusivity is reproduced well. The fact that the diffusivities at higher moisture contents are less accurate is caused by flat profiles for both materials at these moisture contents. It should be noted that the accuracy of the moisture diffusivity is dominated by the accuracy by which the derivative of moisture content with respect to place can be determined.

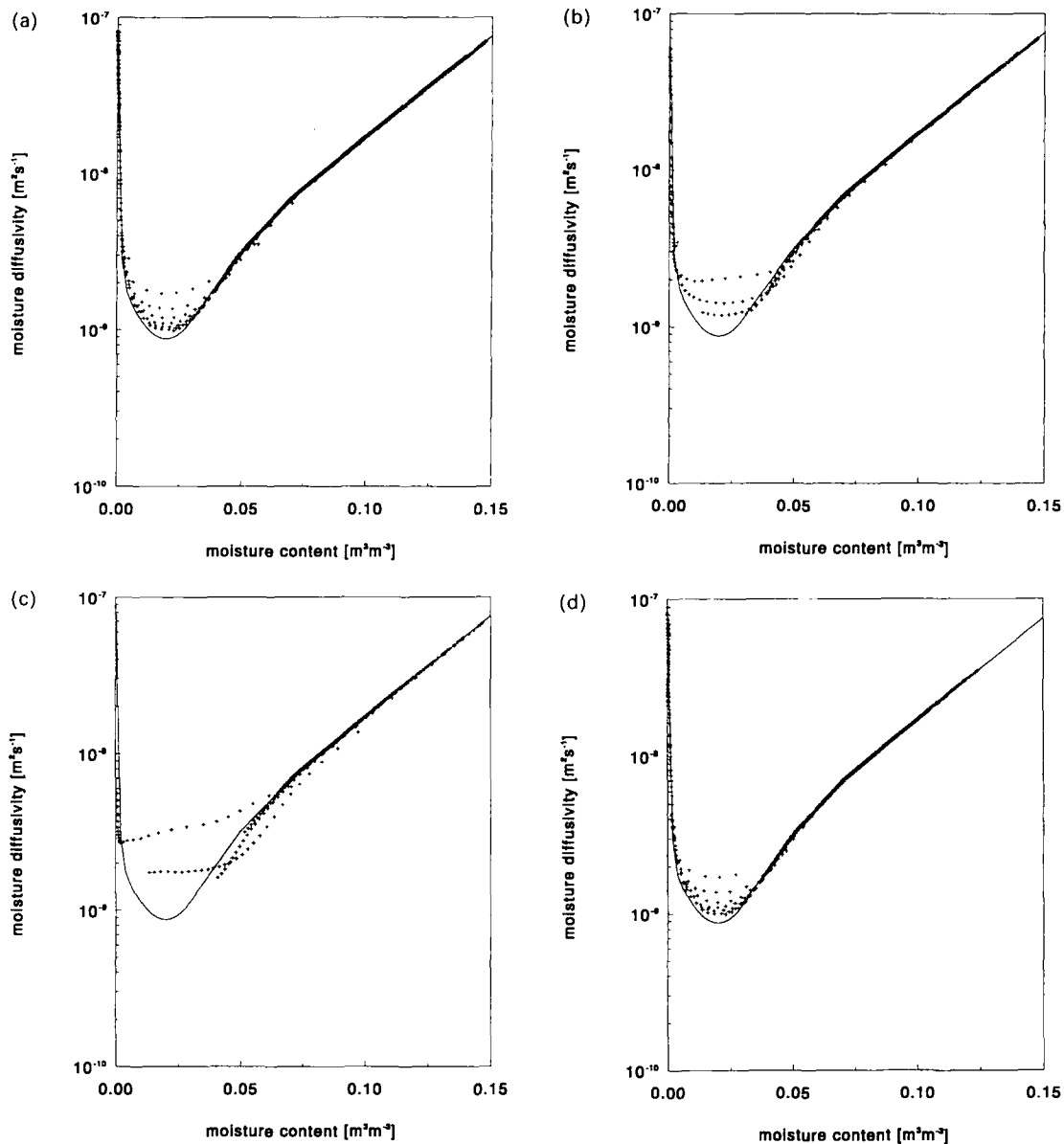The diffusivity for the brick shows a minimum

FIG. 7. Moisture diffusivities as a function of moisture content : 0.5 mm grid (a), 1 mm grid (b), 2 mm grid (c) and variable grid (d). (——) Diffusivity used in the simulation. (+) Diffusivity obtained from simulated profiles.

around a moisture content of approximately 0.02 m^3 m^{-3}, which is directly related to the occurrence of a drying front. To obtain such a drying front one needs a minimum in the moisture diffusivity. For kaolin clay no receding drying front occurs, which is in accordance with the fact that no minimum is observed. For both materials it is obvious that the dependency of the moisture diffusivity on moisture content does not obey simple relations such as exponential or power-law.

5. ERROR ANALYSIS

To be able to determine the isothermal diffusivity from the experimental profile directly, a fine space grid for scanning has to be used. To investigate what maximum distance between data points is allowed, some numerical simulations were performed. A moisture diffusivity similar to the experimentally determined one for the brick was used for this. Moisture concentration profiles were calculated with several grids of equally spaced points (0.5, 1 and 2 mm). From these profiles the moisture diffusivity was recalculated using the same approach as discussed in the preceding section. The results are given in Fig. 7.

For moisture concentrations above 0.06 m^3 m^{-3} the moisture diffusivity was reproduced correctly for all grids. At lower moisture concentrations, the grids of 1 and 2 mm fail to reproduce the minimum in the diffusivity correctly. The 0.5 mm grid reproduces the

minimum, although with a somewhat higher value for the diffusivity. The 0.5 mm grid was adapted in the experiments, since smaller steps would lead to too much time spent in scanning a single profile. An additional reduction of this time was obtained by using a variable grid in the experiment, ranging from 0.5 mm near the surface to 2 mm at the bottom of the sample. The result of a simulation using this variable grid is also included in Fig. 7. The variable grid reproduces the moisture diffusivity as accurately as the 0.5 mm grid; however, in the experiments it takes about half the time.

6. CONCLUSIONS

Scanning neutron radiography has been shown to be an accurate and reliable method for determining moisture concentration profiles. The moisture diffusivity can be determined directly as a function of moisture content from these profiles. Several drying experiments for a brick and kaolin clay show reproducible isothermal moisture diffusivities. For the brick a minimum in this diffusivity occurs, corresponding to a receding drying front. For kaolin clay the moisture diffusivity increases continuously with increasing moisture content, corresponding to the absence of a drying front. In both cases no simple relation can be used to describe the relation between moisture diffusivity and moisture content. Simulations show that a fine spatial grid for scanning should be used near the surface to be able to determine the diffusivity correctly for low values of the moisture content.

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