A gravitational procedure to measure the diffusion coefficient of mater in porous materials : a case study on concrete

Citation for published version (APA):

Zanden, van der, A. J. J., & Taher, A. (2014). A gravitational procedure to measure the diffusion coefficient of mater in porous materials : a case study on concrete. *Drying Technology*, 32(6), 708-712.
<https://doi.org/10.1080/07373937.2013.856915>

DOI:

[10.1080/07373937.2013.856915](https://doi.org/10.1080/07373937.2013.856915)

Document status and date:

Published: 01/01/2014

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
- The final author version and the galley proof are versions of the publication after peer review.
- The final published version features the final layout of the paper including the volume, issue and page numbers.

[Link to publication](#)

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license above, please follow below link for the End User Agreement:

www.tue.nl/taverne

Take down policy

If you believe that this document breaches copyright please contact us at:

openaccess@tue.nl

providing details and we will investigate your claim.

This article was downloaded by: [Eindhoven Technical University]

On: 01 May 2014, At: 02:32

Publisher: Taylor & Francis

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK

Drying Technology: An International Journal

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/ldrt20>

A Gravitational Procedure to Measure the Diffusion Coefficient of Water in Porous Materials: A Case Study on Concrete

A. J. J. van der Zanden^a & A. Taher^a

^a Department of the Built Environment, Eindhoven University of Technology, Eindhoven, The Netherlands

Published online: 16 Apr 2014.

To cite this article: A. J. J. van der Zanden & A. Taher (2014) A Gravitational Procedure to Measure the Diffusion Coefficient of Water in Porous Materials: A Case Study on Concrete, Drying Technology: An International Journal, 32:6, 708-712, DOI: [10.1080/07373937.2013.856915](https://doi.org/10.1080/07373937.2013.856915)

To link to this article: <http://dx.doi.org/10.1080/07373937.2013.856915>

PLEASE SCROLL DOWN FOR ARTICLE

Taylor & Francis makes every effort to ensure the accuracy of all the information (the "Content") contained in the publications on our platform. However, Taylor & Francis, our agents, and our licensors make no representations or warranties whatsoever as to the accuracy, completeness, or suitability for any purpose of the Content. Any opinions and views expressed in this publication are the opinions and views of the authors, and are not the views of or endorsed by Taylor & Francis. The accuracy of the Content should not be relied upon and should be independently verified with primary sources of information. Taylor and Francis shall not be liable for any losses, actions, claims, proceedings, demands, costs, expenses, damages, and other liabilities whatsoever or howsoever caused arising directly or indirectly in connection with, in relation to or arising out of the use of the Content.

This article may be used for research, teaching, and private study purposes. Any substantial or systematic reproduction, redistribution, reselling, loan, sub-licensing, systematic supply, or distribution in any form to anyone is expressly forbidden. Terms & Conditions of access and use can be found at <http://www.tandfonline.com/page/terms-and-conditions>

A Gravitational Procedure to Measure the Diffusion Coefficient of Water in Porous Materials: A Case Study on Concrete

A. J. J. van der Zanden and A. Taher

*Department of the Built Environment, Eindhoven University of Technology,
Eindhoven, The Netherlands*

A new procedure is presented with which the diffusion coefficient of water in partially saturated porous materials can be measured. The first step in the procedure is the creation of a non-equilibrium situation inside a sample by placing it into a centrifuge. In the second step, the mass of the sample is measured by hanging it from two cables, each connected to a balance. The comparison of the time evolutions of the measured masses and the masses as predicted using Fick's second law gives the diffusion coefficient.

Keywords Concrete; Diffusion; Diffusion coefficient; Porous materials; Water transport

INTRODUCTION

Many materials are porous and can absorb water. The presence of water can cause damage (for instance, the biodegradation of wood) where the growth of mold needs the presence of some water. Also well-known is the deterioration of fired-clay bricks when water in the pores freezes and scales can peel off from the brick. It is therefore important to understand the processes involved in the transport of water in porous materials.

As an addition to theoretical models (see Wang et al.^[1]), these processes are studied very often experimentally, because every porous material has its own characteristics. The most obvious physical property that can be measured during a process is the mass of a sample (see, for instance, Legros et al.^[2]) where a drying sample loses weight. Another valuable experimental technique is nuclear magnetic resonance (NMR), where the mass distribution inside a sample can be measured. For instance, Pel et al.^[3] measured the water distribution inside a fired-clay brick during the drying process. The advantage of this technique is that the water distribution can be measured inside a sample as a

function of time. Two important disadvantages of this technique are that it is expensive and that much knowledge is needed to use it. Other techniques used to obtain moisture transport properties are measuring the surface temperature and saturation values using an optical metrology (see Belhamri^[4]) or using interior humidity measurements.^[5]

Another technique has been proposed to determine the diffusion coefficient of water in porous materials by van der Zanden and Aghaei.^[6] In this technique, a sample is dried on one side, while it is hanging from two cables, which are each connected to a balance. This way, more information is obtained from the drying experiment compared to the situation where only the total mass is measured with one balance. Van der Zanden and de Wit^[7] studied the accuracy of this drying procedure when the used balances have a given inaccuracy. This technique is similar to the technique proposed by Gaffner,^[8] who measured the mass at one end of a sample, while the other end was resting on a support. He sealed the sample on all sides and created a non-equilibrium situation inside the sample by placing two differently conditioned sample halves against each other. The main disadvantages of this technique are that both sample halves can have different characteristics and that the connection between the sample halves can introduce resistance against mass transport.

The present paper proposes another measuring procedure to obtain the diffusion coefficient of water in porous materials. In the procedure, a centrifuge is used to create a non-homogeneous water distribution inside a porous material. The material is then taken out of the centrifuge and the method of van der Zanden and Aghaei^[6] is then applied to measure how fast the water reaches the equilibrium distribution again.

In the second section, the proposed measuring procedure is described in detail. Next, an equation is derived with which the concentration gradient of a partially saturated sample in a gravity field can be obtained. How the diffusion coefficient is obtained from the measured masses is not described in the present paper, but is presented by van der

Correspondence: A. J. J. van der Zanden, Department of the Built Environment, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands; E-mail: a.j.j.v.d.zanden@bwk.tue.nl

Color versions of one or more of the figures in the article can be found online at www.tandfonline.com/ldrt.

Zanden and de Wit.^[7] Finally, we examine theoretically how the proposed procedure would work out for a sample of concrete.

MEASURING PROCEDURE

In this section, the method of sample preparation and how the measurements must be performed will be described in detail.

The first step is the sample preparation. The sample is most often cut from a larger piece of material. For the presented procedure, a length of 0.1 m and a height and a width of 0.05 m are convenient. Of course, these values are somewhat arbitrary. The measuring procedure assumes that no water can leave or enter the sample during the mass measurements. To ensure this, the sample must be sealed completely. During the sealing process, a small surface area of the sample is left open, which will be used to condition the sample with a known water content. The first part of the conditioning process of the sample is adding a known amount of water to the sample. After all, the diffusion coefficient depends strongly on the water content. Thus, for each meaningful measurement of the diffusion coefficient, the water content at which the diffusion coefficient is measured must be known. The second part of the conditioning is creating a homogeneous water distribution throughout the sample, which can be achieved by leaving the sample for a long time in a conditioned environment. After this, the remaining open surface area of the sample is also sealed. The time needed to reach a homogeneous water distribution can also be done after the sealing of the sample. The result is a sample with a known water content, which is sealed on all sides. During the sample preparation, the masses are measured after every production step. Thus, the masses of the dry sample, the sealing material, and the added water are known.

The second step is that the sample is placed in a centrifuge (Fig. 1) to create a non-homogeneous water distribution inside the sample. The time that it takes to reach this non-homogeneous water distribution depends on the diffusion coefficient of water in the sample.

The third step is the measurement of the mass on two places of the sample, as proposed by van der Zanden and Aghaei.^[6] Both masses are registered as a function of time. The masses, as predicted from Fick's second law, are fitted with the measured masses. The best fit gives the value of the diffusion coefficient. A detailed description of this method was given by van der Zanden and de Wit.^[7]

It has been shown that the diffusion coefficient is very often a function of the water content—for instance, for gas concrete,^[9] for parboiled rice,^[10] and for pineapple.^[11] The preceding steps give the value of the diffusion coefficient only at one water content of the sample. The sample can be used again for another water content. In order to do so, the sealing must be removed at a small surface area and a small

FIG. 1. After the sample preparation, a non-homogeneous water distribution is in the second step created with a (n artificial) gravity field. In the third step, it is measured how the mass distribution in the sample evolves as a function of time.

amount of water must be evaporated from the sample or a small amount of water must be added to the sample. The sealing must then be repaired, and then the sample is ready for a measurement at another water content. It is also possible to use the same sample upside down to investigate possible inhomogeneities of the sample.

The proposed procedure is better than the procedure proposed by van der Zanden and de Wit,^[7] because in their procedure, the measured masses depend on their drying flux and the diffusion coefficient, while in the present procedure there is no drying flux. The reaching of the new equilibrium, the time evolution of the measured masses, depends only on the diffusion coefficient.

EQUILIBRIUM CONCENTRATION GRADIENT IN A GRAVITY FIELD

When a partially saturated porous sample is placed into a gravity field, the water in the sample will go down until an equilibrium is reached, where the bottom part will have a higher water concentration than the upper part. In the present section, a differential equation will be derived with which the concentration gradient in the sample can be obtained. It is based on the fact that, in an equilibrium situation, the gradient of the hydrostatic pressure is equal to the gradient of the capillary pressure.

The radius of pores in a porous material can vary a few orders of magnitude ranging, for instance, from 0.01 to 10 μm . For an overview of the pore size distribution of different building materials, and how this distribution is measured, see, for instance, the work of Haynes.^[12] When such a material is partially saturated with water, the water is, due to the capillary forces, mainly in the smallest pores. This has been observed experimentally by Engøy et al.^[13] The larger pores are still filled with air. It is assumed here that both phases (i.e., the water and the air phase) are continuous. This means that every geometrical point in one phase can be

reached from any other point in that same phase through a path that also lies completely in that same phase.

When such a partially saturated sample is placed into a gravity field with acceleration g , the situation of Fig. 2 arises. The water is pulled down by gravity, which results in a higher water concentration at position x than at position $x + dx$. At position x , more pores are filled with water than at position $x + dx$.

In the absence of gravity, such a concentration gradient would lead to a pressure gradient in the water phase and to water flow through the water-filled pores, as is shown by the model of van der Zanden et al.^[14,15] In the presence of gravity, however, the difference between the pressure in the water phase at x and at $x + dx$ is, in an equilibrium situation, because of the assumption of continuity in the water phase, the hydrostatic pressure difference, and is given by

$$p(x) = p(x + dx) + \rho g dx, \quad (1)$$

where ρ is the density of water. An example of a cumulative pore size distribution of a fictional material is sketched in Fig. 3. It is measured by forcing mercury under pressure into a sample, as was proposed by Ritter and Drake^[16] in 1945. From the pressure in the mercury needed to force the mercury into the sample, the average pore radius can be obtained using Laplace's law (Atkins^[17]) and the volume of mercury in the sample is measured. The result is given as a cumulative volume fraction, F . The maximum value of F , $F_{\max}$, is the total volume of the pores expressed as the volume fraction of the porous sample, and is thus for this case the porosity of the concrete. For the sample of Fig. 2, at position x , the volume fraction occupied by water is $F_{\max} - F(x)$. This means that all the pores with a radius smaller than $R(x)$ are filled with water. Likewise for $F(x + dx)$ and $R(x + dx)$.

The density of air is much smaller than the density of water. The hydrostatic pressure difference is in the air

FIG. 2. A partially saturated porous material that is placed into a gravity field has, in an equilibrium situation, more pores filled at position x than at position $x + dx$, when the gravitation points into the negative x direction.

FIG. 3. A hypothetical pore size distribution. The cumulative volume fraction is given as a function of the pore radius.

phase much smaller than in the water phase, and is thus neglected in the derivation below. The pressure in the air phase is set to zero.^[14] This is not the real value, but only pressure differences are relevant for the present reasoning. Then, again using Laplace's law,^[17] the water pressure can be related to the largest filled pore radius as

$$p(x) = -\frac{2\sigma}{R(x)}, \quad (2)$$

and

$$p(x + dx) = -\frac{2\sigma}{R(x + dx)}. \quad (3)$$

where σ is the surface tension of water.

Substitution of Eqs. (2) and (3) into Eq. (1) gives

$$\frac{-2\sigma}{R(x)} = \frac{-2\sigma}{R(x + dx)} + \rho g dx, \quad (4)$$

which can be written as

$$\frac{-2\sigma}{R} = \frac{-2\sigma}{R + dR} + \rho g dx. \quad (5)$$

Eq. (5) can be written as

$$\frac{-2\sigma}{R} = \frac{-2\sigma}{R} \left(\frac{1}{R + \frac{dR}{R}} \right) + \rho g dx, \quad (6)$$

which can be approximated as

$$\frac{-2\sigma}{R} = \frac{-2\sigma}{R} \left(1 - \frac{dR}{R} \right) + \rho g dx, \quad (7)$$

which can be simplified to

$$\frac{dR}{dx} = \frac{-\rho g R^2}{2\sigma}. \quad (8)$$

For F as a function of x , Eq. (8) can be used to obtain

$$\frac{dF}{dx} = \frac{dF}{dR} \frac{dR}{dx} = \frac{dF}{dR} \left(\frac{-\rho g R^2}{2\sigma} \right). \quad (9)$$

The result of this section, Eq. (9), gives the gradient of the cumulative volume fraction F and, because the water concentration is given by ρF , essentially also the concentration gradient in a partially saturated porous material that is placed into a gravity field. If this gravity field is artificially created with a rotating motion, the acceleration g is a function of position x .

THEORETICAL APPLICATION ON A CONCRETE SAMPLE

A hypothetical experiment is considered on a sample with length L , as shown in Fig. 4. The mass is measured on the left side, M_L , and on the right side, M_R . The (centrifugal) acceleration was pointing in the negative x direction, as in Fig. 2. A water concentration profile is sketched with a low concentration at $x = L$ and a high concentration at $x = 0$.

An estimation will be given of the mass difference that can be expected when the sample of Fig. 4 has been in a gravity field. The sample can be considered to consist of two halves, one on the left side and one on the right side. In each half, there is an average water concentration, which differs from the others. This results in a difference in the masses of the two halves M_L and M_R . To obtain this mass difference, Eq. (9) is used to arrive at the approximation

$$\Delta F \equiv F_L - F_R \cong \frac{L}{2} \frac{dF}{dR} \left(\frac{-\rho g R^2}{2\sigma} \right), \quad (10)$$

from which follows

$$\Delta M \equiv M_L - M_R \cong \frac{L}{2} \frac{dF}{dR} \left(\frac{-\rho g R^2}{2\sigma} \right) \frac{V}{2} \rho, \quad (11)$$

FIG. 4. In an experiment, the total mass of the sample is divided into two, which are measured independently. A non-homogeneous water content is shown, and the average water content of the two halves are shown.

where V is the volume of the sample and ΔM the mass difference.

Eq. (11) has a very simple form, and it is accurate enough for an estimation of the order of magnitude. The precise equations for M_L and M_R follow from the mechanical no-rotation conditions.^[6,7] If the mass difference is not large enough, it can be increased by moving the cables closer to each other. This way, the sensitivity of the measuring procedure can be increased.

This method can now be applied theoretically on a sample of concrete. In Fig. 5, the pore size distribution of a concrete is given, as the result of a computation presented by Ye.^[18] The pores have a size typically in the range 2×10^{-7} – 2×10^{-6} m. An estimation will follow of the mass difference, given by Eq. (11), for a sample that has an easy-to-handle size in an experiment of a centrifugal velocity that can be obtained easily. The diameter of the largest filled pores is set at $1 \mu\text{m}$. From Fig. 5 and after a computation, it follows that dF/dR has the value -10^5 m^{-1} . A convenient sample size is a length of 0.1 m and a width and height of 0.05 m . The density of water is approximately 1000 kg m^{-3} . The surface tension of water is $73 \cdot 10^{-3} \text{ N m}^{-1}$. A centrifuge for drying clothes can easily have a rotation velocity of 20 rps , which implies an angular velocity, ω , of 120 rad s^{-1} . If the sample is placed 0.25 m from the rotation axis, the acceleration,

$$g = \omega^2 r, \quad (12)$$

with r the distance from the rotation axis, which becomes $3.6 \cdot 10^3 \text{ m s}^{-2}$. Using these values in Eq. (11) gives a mass difference of $3.1 \cdot 10^{-3} \text{ kg}$. This mass difference can be measured very easily. For dryer samples, the mass difference is smaller. For instance, for the same sample but with the pores filled up to a pore diameter of $0.1 \mu\text{m}$, dF/dR has approximately the same value as before, but the 10 times smaller pore radius leads, according to Eq. (11), to a 100 times smaller mass difference. For higher water concentrations, the mass difference is larger.

FIG. 5. The pore size distribution of a concrete as found in the literature.

CONCLUSION

A procedure to measure the diffusion coefficient of water in porous materials has been presented. After preparation of the sample, which is the first step, the procedure consists of a second step, where a non-equilibrium situation inside a partially saturated porous sample is created using a centrifuge. In the third step, the sample is taken out of the centrifuge and we measure how fast the water distribution inside the sample reaches its new equilibrium by hanging the sample from two cables, each connected to a balance. The comparison with Fick's second law gives the diffusion coefficient.

The theoretical application on concrete shows that, for this material, the procedure can be used. In order to show the feasibility of the procedure, the pore size distribution must be known. However, to measure the diffusion coefficient with the proposed procedure on a material, the pore size distribution does not need to be known.

NOMENCLATURE

F	cumulative volume fraction (—)
$F_{\max}$	porosity (—)
g	(gravitational) acceleration (m s^{-2})
L	length of a sample (m)
M	mass (kg)
M_L	mass of left side (kg)
M_R	mass of right side (kg)
p	pressure (N m^{-2})
R	pore radius (m)
r	distance from the rotation axis (m)
V	volume of a sample (m^3)
x	position coordinate (m)

Greek

ρ	density of water (kg m^{-3})
σ	surface tension (N m^{-1})
ω	angular velocity (rad s^{-1})

REFERENCES

- Wang, W.; Chen, G.; Mujumdar, A.S. Physical interpretation of solids drying: An overview on mathematical modeling research. *Drying Technology* **2007**, *25*, 659–668.
- Legros, M.; Yoshida, M.; Sakaguchi, T.; Imakoma, H.; Okazaki, M. Determination method of variable diffusion coefficients of hygroscopic material from drying rate curves of the single particle. *Drying Technology* **1992**, *10*, 733–752.
- Pel, L.; Kopinga, K.; Bertram, G.; Lang, G. Water absorption in a fired-clay brick observed by NMR scanning. *Journal of Applied Physics D* **1995**, *28*, 675–680.
- Belhamri, A. Characterization of the first falling rate period during drying of a porous material. *Drying Technology* **2003**, *21*, 1235–1252.
- Zhang, J.; Hou, D.; Gao, Y.; Wei, S. Determination of moisture diffusion coefficient of concrete at early age from interior humidity measurements. *Drying Technology* **2011**, *29*, 689–696.
- van der Zanden, A.J.J.; Aghaei, A. Determination of the diffusion coefficient of water in brick with a simple mass distribution technique. *Drying Technology* **2009**, *27*, 393–402.
- van der Zanden, A.J.J.; de Wit, M.H. A procedure to measure the diffusion coefficient of water in brick as a function of the water concentration. *Drying Technology* **2012**, *30*, 526–534.
- Gaffner, D. The moment method for measuring moisture diffusivity of porous building materials. In *Proceedings of the Conference Thermal Performance for the Exterior Envelope of the Whole Building X*, Clearwater Beach, FL, December 2–7, 2007.
- Dietll, C.; George, O.P.; Bansal, N.K. Modeling of diffusion in capillary porous materials during the drying process. *Drying Technology* **1995**, *13*, 267–293.
- Elbert, G.; Tolaba, M.P.; Aguerre, R.J.; Suarez, C. A diffusion model with a moisture dependent diffusion coefficient for parboiled rice. *Drying Technology* **2001**, *19*, 155–166.
- Simal, S.; Garau, M.C.; Femenia, A.; Rossello, C. A diffusional model with a moisture-dependent diffusion coefficient. *Drying Technology* **2006**, *24*, 1365–1372.
- Haynes, J.M. Determination of pore properties of constructional and other materials. *Matériaux et Construction* **1973**, *6*, 169–174.
- Engøy, T.; Feder, J.; Jøssang, T. Counter-flow in drying of competing pores. *Physica Scripta* **1991**, *T38*, 99–102.
- van der Zanden, A.J.J.; Coumans, W.J.; Kerkhof, P.J.A.M.; Schoenmakers, A.M.E. Isothermal moisture transport in partially saturated porous media. *Drying Technology* **1996**, *14*, 1525–1542.
- van der Zanden, A.J.J. A possible revision of the results of a model for moisture transport in partially saturated porous media. *Drying Technology* **1995**, *13*, 2227–2231.
- Ritter, H.L.; Drake, L.C. Pore-size distribution in porous materials. *Industrial and Engineering Chemistry* **1945**, *17*, 782–791.
- Atkins, P.W. *Physical Chemistry*, 6th ed.; Oxford University Press: Oxford, UK, 1998.
- Ye, G. *Experimental Study and Numerical Simulation of the Development of the Microstructure and Permeability of Cementitious Materials*; Delft University Press: Delft, The Netherlands, 2003.