Chemical Engineering Science 109 (2014) 204–211

Contents lists available at ScienceDirect

Chemical Engineering Science

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

Paradoxical drying of a fired-clay brick due to salt crystallization

Sonia Gupta [a], Hendrik P. Huinink [a,*], Marc Prat [b,c], Leo Pel [a], Klass Kopinga [a]

[a] Transport in Permeable Media, Department of Applied Physics, Eindhoven University of Technology, PO Box 513, 5600 MB Eindhoven, The Netherlands
[b] INPT, UPS, IMFT (Institute de Mecanique des Fluides de Toulouse), University de Toulouse – Allee Camille Soula, F-31400 Toulouse, France
[c] CNRS, IMFT, F-31400 Toulouse, France

HIGHLIGHTS

- Influence of crystallization on the formation of receding drying front.
- Critical moisture content is not an intrinsic property of a porous material.
- Critical moisture content varies depending on the initial drying rate.
- Initial drying rate influences efflorescence type formed at the material surface.

ARTICLE INFO

Article history:
Received 25 October 2013
Received in revised form
20 January 2014
Accepted 22 January 2014
Available online 31 January 2014

Keywords:
Drying
Porous media
Crystallization
Efflorescence
Salt weathering
Critical moisture content

ABSTRACT

We investigated how salt crystallization inside a porous building material influences the formation of a receding drying front. Nuclear Magnetic Resonance (NMR) is used to measure non-destructively both hydrogen and dissolved sodium ions during drying process. In this study, we focused on the influence of NaCl on the drying behavior of porous media. The results show that salt changes the drying behavior. At low relative humidities (RH∼0%), the drying rate of a brick saturated with NaCl solution is much lower than the drying rate of water saturated brick. Moreover, the presence of salt suppresses the development of a receding front. In this case homogenous drying of the material continues till very low saturation values. This is due to salt crystallization near the surface of the brick that causes blockage of the pores. This blockage reduces evaporation rate at the surface and allows maintaining a continuous hydraulic connection between the surface of the porous medium and the liquid present inside the material till low saturation values. In the case of a salt solution saturated brick, increasing relative humidity to 55% and 70% leads to a paradoxical situation where the evaporation rate is greater for 55% and 70% RH than for 0% RH. The paradox is explained by the impact of evaporation rate on the efflorescence microstructure, leading to the formation of a blocking crust for sufficiently high evaporation rates and non-blocking efflorescence for sufficiently low evaporation rates. The fundamental difference between the two types of efflorescence is demonstrated from a simple imbibition experiment. Using a simple continuum scale model of drying, critical moisture content was determined and all the essential features of the experimental results are validated. It is shown that critical moisture content can be very low in the case of fired-clay brick due to the low threshold of the pore space, which is consistent with the relatively large pore size distribution of the fired-clay brick.

1. Introduction

Salt crystallization inside the pores of a building material can cause severe damage (Scherer, 1999) and is known since ancient times (Taber, 1916). Salt can enter a building material along with moisture in various ways, such as capillary rise of ground water, absorption of sea water, atmospheric pollution. Almost all water that enters building materials will leave by evaporation or drying. Due to drying salt crystals will crystallize inside the pores of a building material.

Drying of porous media in the presence of salt is of interest not only to understand the crystallization mechanisms and related salt damage problems, but also in the field of soil physics (Nachshon et al., 2011), injecting CO_2 in aquifers (Peysson et al., 2011), and related studies. Vegetation, plant growth and soil organisms can be severely limited in salt-affected land. Despite the above mentioned problems, drying of porous materials in the presence of salt is still poorly understood. This is due to the complex phenomena involved, e.g., moisture and ion transport phenomena and phase transitions. Therefore, a better insight into the drying process of

* Corresponding author. Tel.: +31 40 247 5375; fax: +31 40 243 2598.
E-mail address: h.p.huinink@tue.nl (H.P. Huinink).

0009-2509/$ - see front matter © 2014 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ces.2014.01.023

salt containing porous media is required to understand various salt related damage problems.

Lewin (1982) proposed that the location of salt crystallization is determined by a dynamic balance between the rate of escape of water at the surface (evaporation or drying) and the rate of supply of water to the surface. If salt crystallizes inside the material, i.e., the so-called sub-florescence, it can cause damage. On the other hand, if salt crystallizes outside the material, also called efflorescence, it may be unesthetic but it is not harmful for the buildings. The location of salt crystallization is in general determined by the competition of diffusive and advective fluxes. Generally it is characterized in terms of a Peclet number (Pe) (Huinink et al., 2002).

In the past, a significant progress has been made in understanding the drying kinetics of porous materials, especially focusing on the pore level (Prat, 2007; Prat and Bouleux, 1999). It is widely mentioned that for irregular pore geometry thick liquid films provide an efficient transport of liquid to the surface (Chauvet et al., 2009). The pores inside the brick are generally not perfectly cylindrical and have edges; consequently thick liquid films will be present. These films are known to contribute significantly to the mass transport phenomenon during stage-1 of drying (Laurindo and Prat, 1998). The films supply water to isolated liquid clusters, thereby enhancing the mass transfer in comparison to the vapor diffusion transport mechanism (Laurindo and Prat, 1998). The standard drying behavior of water saturated porous material is well documented in the literature (Hall and Hoff, 2002). However, the drying behavior of the porous material in the presence of salt is still a topic of discussion, especially with regard to the impact of salt crystallization on drying. By varying the mean pore size of the porous material, it has been shown that the crystallization of the salt at the porous material surface could either severely reduce the evaporation rate compared to pure water or have no impact at all (Eloukabi et al., 2013). This led to identify two main types of efflorescence, referred to as patchy (or non-blocking) and crusty (or blocking). As we shall see, distinguishing these two main types of efflorescence is also a key factor for analyzing our results.

The aim of this work is to study the drying behavior of porous materials saturated with the solution. In this study we will focus on NaCl as this is a common salt found in many salt related problems in situ. We will investigate to which extent this drying behavior resembles the standard drying behavior of water saturated materials. In particular, we focus on the effect of salt crystallization inside a porous material on the development of a receding front during drying. To our best knowledge, no study has been reported in the past to explore this aspect of drying, probably because it is difficult to measure the moisture and ion distribution simultaneously in real porous materials. For this purpose a specially designed Nuclear Magnetic Resonance (NMR) set-up with a static magnetic field of 0.78 T was used (Kopinga and Pel, 1994). With this set-up it was possible to carry out non-destructive, quantitative and simultaneous measurements of both the hydrogen and sodium content in a brick sample.

2. Material and methods

2.1. Materials

The material used in this study was fired-clay brick. The red fired-clay brick is of a type typically used for construction in the Netherlands, and had an average porosity (as measured by water immersion method) of 0.32 $m^3\ m^{-3}$, and a pore size distribution ranging from a few tens of nanometers to 100 μm (with 80% of the total pore space corresponding to pores in the range 1–10 μm), as determined by Mercury Intrusion Porosimetry (MIP) (Gupta, 2013).

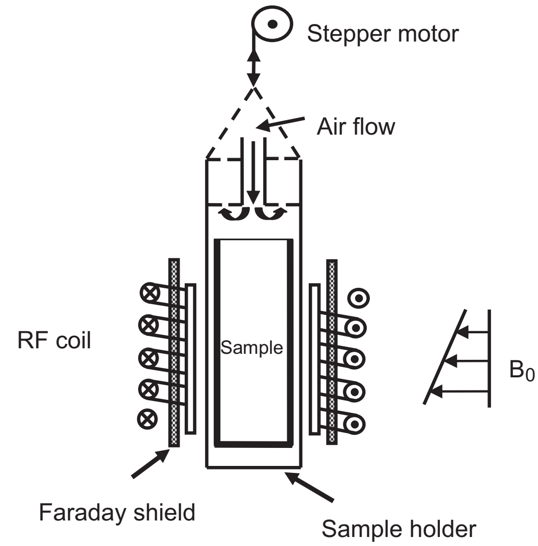

Fig. 1. NMR set-up for the drying experiments with cylindrical samples. The Teflon holder with the saturated sample is moved in the vertical direction by means of a stepper motor. The signal from H or Na nuclei at resonance is received by the RF coil.

2.2. Nuclear magnetic resonance (NMR)

A specially designed nuclear magnetic resonance (NMR) set-up, for non-destructive, quantitative and simultaneous measurement of moisture (^{1}H) and sodium (^{23}Na) profiles in inorganic materials was used. An extensive description of this set-up can be found elsewhere (Kopinga and Pel, 1994). A schematic representation of the set-up is given in Fig. 1. The tuned circuit of the set-up can be toggled between 33 MHz for ^{1}H and 8.9 MHz for ^{23}Na, giving the possibility to quasi-simultaneously measure the H and Na content and thereby the concentration. For the echo time used in the experiment, i.e., T_E=250 μs for ^{1}H and 450 μs for ^{23}Na, only the dissolved Na and H nuclei are measured and no signal is obtained from the nuclei incorporated in the crystals. The magnetic field gradient was chosen so that a slice of less than 2 mm is measured. The cylindrical samples 20 mm in diameter and 40 mm in length were vacuum saturated with water and 3 m NaCl solution. These samples were sealed using Teflon tape on all sides except the top surface and placed in the NMR sample chamber. In this way a one dimensional drying experiment was performed. The samples were then exposed to dry air at a flow rate of 1 l min^{-1}. The relative humidity was varied from 0% to 70%. The sample was moved in the vertical direction using a stepper motor to allow the measurement of moisture and sodium content throughout the sample length. Measuring one profile takes about 2.26 h. As the complete drying experiment takes in the order of a few days, small variations in the moisture and ion profiles during a single scan can be neglected. After each drying experiment was completed the efflorescence formed on the top of the sample was collected and weighed.

3. Results

3.1. Drying behavior of water and salt solution saturated fired-clay brick dried at 0% relative humidity

Initially, the drying behavior of fired-clay brick samples vacuum saturated with water and salt solution (3 m NaCl) was studied. The samples were dried at 0% RH and 1 l min^{-1} air flow rate. Fig. 2a and b shows the measured moisture profiles during drying of water and salt saturated samples, respectively. The recurrent irregularities in the profiles result from inhomogenities