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Preliminary investigation of change of pore structure due to salt precipitation during evaporation in brick with X-ray computed tomography

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Abstract

Pore clogging by salt crystallization changes mass transport phenomena. This paper reports on our preliminary attempt to quantify the spatial distribution of salt crystals through time in fired clay brick during drying by using synchrotron radiation X-ray computed tomography (CT; beamline BL20B2 at SPring-8, Japan) that

allows fast 3D imaging with high spatial resolution. Specimens saturated with NaCl solution are dried at ambient conditions and scanned at certain moments in time. The mass loss of the specimens during drying is also measured gravimetrically to investigate the macroscopic effect of salt precipitation on water evaporation, and is used for the evaluation of the validity of the image analysis. Since the amount of salt precipitation identified from CT images and the amount of salt precipitation calculated from gravimetric measurements is almost the same, the usefulness of synchrotron X-ray CT imaging for verifying the temporal evolution of crystal formation in our type of porous bricks is confirmed. Image analysis suggests that salt precipitation on the surface of the material (i.e. efflorescence) mainly occurs during the first 2 hours of drying, and inside the material (i.e. sub-efflorescence) in the latter stages of drying.

Keywords: synchrotron X-ray CT, water evaporation, sodium chloride, salt precipitation

1 Introduction

Salt weathering is a phenomenon in which materials are physically destroyed due to the build-up of crystallization-induced stress, and it is one of the degradation factors of brick or stone buildings and sculptures. This deterioration phenomenon has been confirmed in many historical sites which have been constructed in brick like Mohenjodaro (Fodde 2008) and at the Hagia Sophia in Istanbul (Mizutani et al. 2015). A prediction model for salt weathering is very useful to develop an appropriate conservation and restoration plan in different climate conditions. The frame work for numerical analysis including salt precipitation and deformation has been constructed recently, and several numerical models have begun to be proposed (Koniorczyk et al. 2012 and Derluyn et al. 2014). However, there are many points where the physical phenomena related to salt precipitation in the porous material itself are still unknown.

One of the issues for improving the numerical analysis of salt weathering is how to express the

change in mass transport properties due to the precipitation of salt. The influence of salts dissolved in solution on the mass transport properties such as the sorption isotherm and the moisture diffusivity have been well studied. It should be noted that there are exceptions (Pel, L et al. 2000), but in many cases the explanation is given by considering the increase of viscosity and density with salt concentration, and the decrease of surface tension as well as of water vapor pressure. On the other hand, although the significance of the effect of pore clogging due to salt precipitation on mass transport is obvious from experimental research in the literature (Espinosa-Marzal et al. 2013), the quantitative relationship between the crystal formation and the change in the mass transport properties is not known because of the difficulty in quantifying the salt crystals in the porous material. In order to relate salt precipitation and change of transport properties, it is needed to determine where and how much salts precipitate and how the salt crystals change the pore network structure. The pore network structure is mainly characterized by the connectivity, the pore size distribution and the tortuosity, which are directly affecting gas and liquid transfer in porous media. In this paper, a series of drying experiments using fired clay brick saturated with NaCl solution is performed to quantify the salt precipitation on the surface and inside the porous media, as well as its influence on mass transport. Synchrotron radiation X-ray computed tomography, which offers 3D images with high spatial resolution, is used to quantify the spatial distribution of salt crystals through time in fired clay brick during drying. Gravimetric measurements of the mass loss during drying are also performed to investigate the macroscopic effect of precipitated salt on the drying rates. In addition, the total amount of salt crystals is calculated from the mass loss, and is compared with the results obtained from CT image analysis.

2 Materials and methods

2.1. Materials

Japanese fired clay brick (red brick produced by Okamoto co.) was used for the experiments. The

capillary moisture content of this brick is 0.17 [m³/m³], and the total open porosity is 0.24 [m³/m³]. Figure 1 shows the result of mercury intrusion porosimetry (Autopore IV9500) of a dry brick. Most of the pores are capillary pores (pore diameter between 1µm and 1mm (Klopper 1985)), implying that the moisture uptake transfer in this brick is primarily caused by capillary forces.

Brick specimens of 5mm x 5mm x 8mm were prepared, and an epoxy resin was applied on the lateral sides, making these surfaces water and vapor tight. This ensures a one-dimensional mass transfer during the drying experiments. Drying was monitored after three or four saturating specimens under each of four conditions: (1) with pure water, (2) with an unsaturated NaCl salt solution of 8g NaCl/100g water, (3) with a saturated NaCl salt solution and (4) with a 2nd saturation with saturated NaCl salt solution after experiment (2).

The monitoring was conducted both gravimetrically and through X-ray CT scanning at beamline BL20B2. The gravimetric measurements were performed outside of the X-ray CT hutch, at 25°C and 35.8 % RH, whereas the climate inside the hutch was slightly warmer, being 26.5°C and 33% RH on average. The specimen were placed in a holder as shown in Figure 2. The specimen was hold by an upper polypropylene cup in which the pure water or salt solution was absorbed. Then the upper cup was mounted in a lower cup with pure water (condition (1)) or NaCl saturated solution (conditions (2) - (4)) in order to suppress evaporation from the bottom surface of the sample.

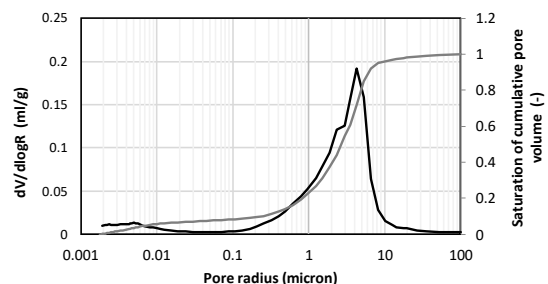


Figure 1: Pore size distribution of the brick sample at the beginning of the experiment measured by mercury intrusion porosimetry.

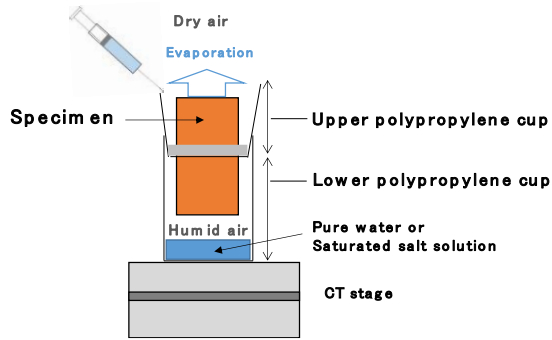


Figure 2: Schematic of the experimental setup.

2.2. Gravimetric experiments

The specimens were saturated with water or salt solution, by injecting the liquid into the upper cup with a syringe (Figure 2). The liquid was absorbed for 30 minutes to ensure capillary saturation. Thereafter, the solution in the upper cup was wiped with a tissue and the brick was dried via the upper surface of the test sample. The time curve of the accumulated amount of evaporation was measured with a balance with a 0.0001g accuracy (Sartorius AG). Only the mass of the upper part, consisting of the upper polypropylene cup and the specimen was measured so as not to be affected by the mass loss of the solution in the lower polypropylene cup. The mass was measured at intervals of about 15 minutes during the initial stage of drying, and thereafter at 30 minutes intervals.

2.3 X-ray CT experiments and image processing

Synchrotron radiation X-ray CT imaging was performed at beamline BL20B2 (SPring-8, Hyogo prefecture, Japan) using a deflection electromagnet as a light source. The critical energy at BL20B2 is 28.9 keV, and the beam size is about 75 mm (horizontal) \times 5 mm (vertical). A Hamamatsu Photonics' ORCAFlash4.0 CCD-camera was used as the image detector. Scans were acquired with a spatial resolution of 2.74 $\mu\text{m}/\text{pixel}$, the duration for one scan acquisition was about 5 minutes. The resulting 3D reconstructed volume of each scan consists of $2048 \times 2048 \times 1700$ voxels ($\pm 6.5 \times 6.5 \times 4.7$ mm in width \times depth \times height). In order to cover the full height of the specimen, three scans were taken at different heights and stitched together to

obtain a 3D reconstruction of the complete brick specimen.

First, a three-dimensional CT imaging of a dry specimen was performed. After that, the specimen was wetted in the same way as for the gravimetric experiments, and the wet sample was imaged. Subsequently, several X-ray CT-scans were performed during the drying process, i.e. after 30 minutes, 60 minutes, 120 minutes, 180 minutes and 480 minutes of drying. The specimen was kept on the CT-stage until the end of the scanning series, and extreme care was taken to ensure that the specimen position did not change, particularly during solution injection and wiping.

The image processing starts with reducing the influence of noise, for which a median filter was used on the 3D reconstructed volumes of the brick specimen. The gray values of the CT images reflect the density and atomic number of the components present in the specimen. The specimen consists of solid brick and its pore space, which can be partly or fully filled with fluid (water or salt solution) and salt crystals. Each of these components have a different gray value distribution, and by thresholding gray value ranges, the different components can be extracted, i.e. segmented, from the images. However, in our case, the overlap of the gray values of the substances is large, as shown in Figure 3 containing four phases determined from different CT images of condition (3). Therefore, the solid phase was segmented from the CT images of the dry brick, and subtracted from the images acquired during drying. The salt crystal phase is then segmented based on grayvalue thresholding of these differential images.

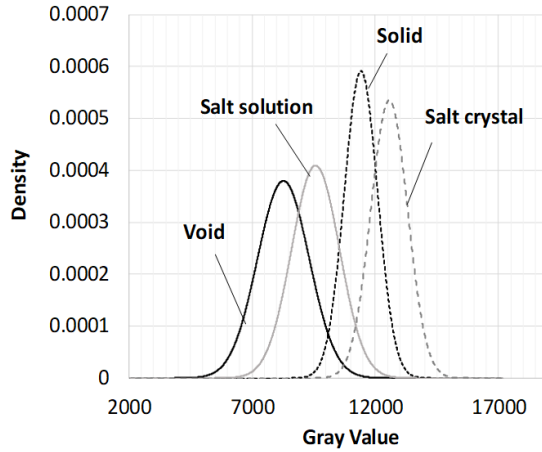


Figure 3: Gray value of each substance determined from CT images of condition (3).

3 Results and Discussion

3.1 Gravimetric experiments

Figure 4 shows the accumulated amount of evaporation from the brick specimen. The evaporation rate becomes slower as the salt content increases. After 300 minutes from the start of drying, 89% of the initial absorbed water was dried on average in the three cases of pure water, while 77% was dried for the unsaturated sodium chloride solution (8g/100g water), 53% for the saturated solution and 33% for the saturated solution during the second cycle. Though it is reported that salt precipitation may increase or decrease the evaporation rate depending on precipitation patterns affecting the pore size distribution and pore shape of porous media (Eloukabi et al. 2013), in our case, the salt in the specimen causes a clear decrease.

The influence of salt was also quantified on two different regimes during drying (Desarnaud et al. 2015, Scherer 1990 and Shokri et al. 2011): *capillary regime* characterised by a fast constant evaporation at the initial stage of drying where capillary force is active and *diffusive regime*, characterized by a small evaporative flux, where

evaporation rates of the capillary regime and the diffusive regime are calculated and shown in table 1 for the brick specimen. The average evaporation rate for salt containing brick specimens decreases in both the capillary and diffusive regime when compared to a brick containing pure water, indicating that salt inside and outside the material affects the movement of both the liquid and gas phase. Especially in cases (3) and (4) for bricks containing saturated NaCl salt solution the evaporation rate in the diffusive regime becomes slow.

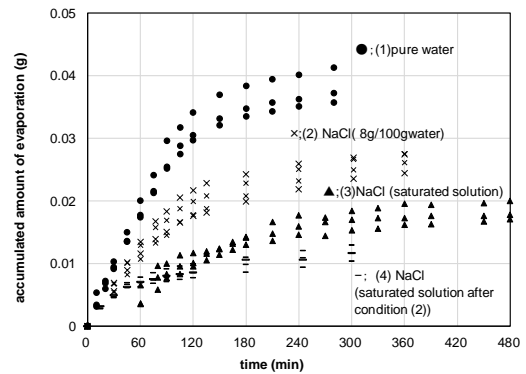


Figure 4: Accumulated amount of evaporation over time, obtained from gravimetric measurements.

3.2 X-ray CT experiments

In this section, we focus on the image analysis to quantify the spatial distribution of salt crystals for the experiment with NaCl saturated salt solution (condition (3)). The analysis of the other experiments is ongoing.

The total open porosity of the whole sample identified by image analysis is 0.195 [m³ / m³] which corresponds to about 81% of the total open porosity of 0.240 [m³ / m³]. This difference seems to be reasonable considering the ratio of voids below the spatial resolution (2.74 μm/pixel), as shown in Figure 1, that cannot be identified. Figure 5 shows the comparison of the

Table 1: The average evaporation flux per unit surface area

	(1) pure water	(2) NaCl (8g/100g)	(3) saturated NaCl salt solution	(4) saturated NaCl salt solution after condition (2)
Capillary regime [g/(min•m ²)]	9.93	6.55	-	4.98
Diffusive regime [g/(min•m ²)]	1.06	1.0	0.53	0.63

vapor diffusion is dominant. The average

evolution as a function of time of the total

amount of precipitated salt obtained from gravimetry and from CT image analysis. The amount obtained from gravimetry is calculated from the amount of water evaporation measured, assuming that salt crystals precipitate as soon as the salt concentration exceeds the saturated concentration. The amount of salt identified by image analysis is 1.68 times larger at 60 minutes, 0.91 times at 120 minutes, 0.87 times at 180 minutes and 0.92 at 480 minutes. Although there would be some differences between specimens in Figure 5, the overall trend of salt precipitation can be rather sufficiently identified, which would confirm the suitability of the use of synchrotron X-ray CT imaging for verifying the temporal evolution of crystal formation in porous materials and its influence on the liquid transport phenomena.

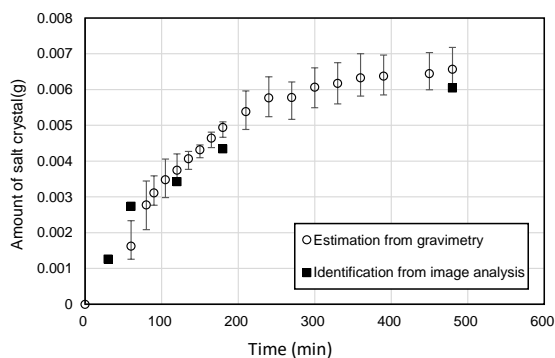


Figure 5: Total amount of precipitated salt as a function of time for samples of condition (3). The data points of estimation from gravimetry give the values averaged for three specimens as well as the minimum and maximum values.

Figure 6 shows the time evolution of the spatial distribution of salt crystals identified by the image analysis, focused at the evaporation surface and the inside of the brick. The salt precipitation occurs on the surface of the material (i.e. efflorescence) during the initial 120 minutes, and thereafter, proceeds inside (i.e. sub-efflorescence). This result indicates that salt solution could be transported to the surface by capillary force up to 120 minutes, whereafter the drying front recedes, causing salt precipitation inside. Figure 7 shows segmented images of the horizontal cross sections taken at a depth of 0mm in Figure 6 at 60 minutes and 480 minutes after the start of drying. Salt precipitation only occurred in large pores at 60 minutes, but was found in smaller pores at 480 minutes. This result seems to be qualitatively correct as it is consistent with the theory of evaporation, saying that evaporation is likely to empty first large pores. Later on in the evaporation, salt precipitation increases in smaller pores, which becomes more difficult to identify due to the limit in spatial resolution. This may be the reason why the identified salt amount values by image analysis become smaller than the values from gravimetry after 120 minutes. This difference should be taken into account when the influence of salt precipitation on the mass transport phenomenon is discussed further using the whole data set obtained from the drying experiments with X-ray CT in the future.

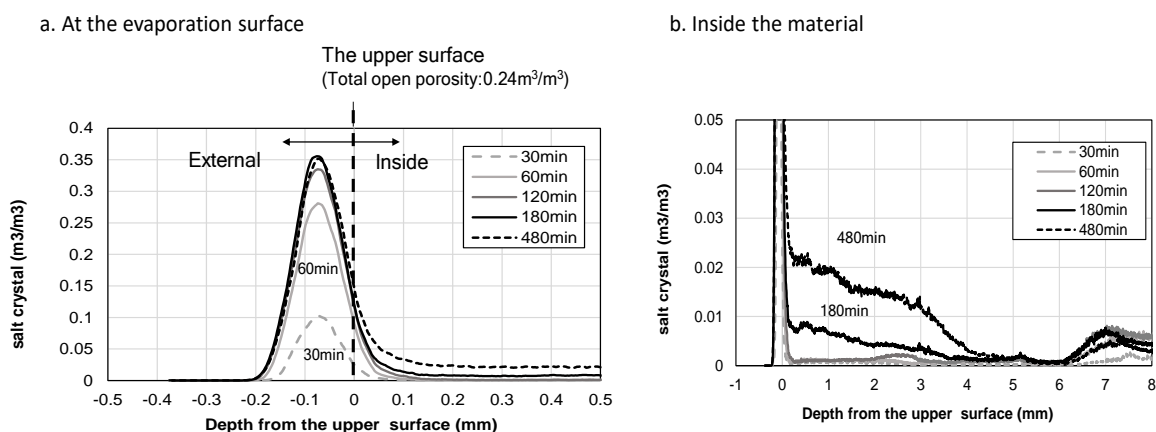


Figure 6: The temporal evaluation of spatial distribution of salt crystals identified by X-ray CT image analysis focused on (a) the evaporation surface and (b) inside the specimen.

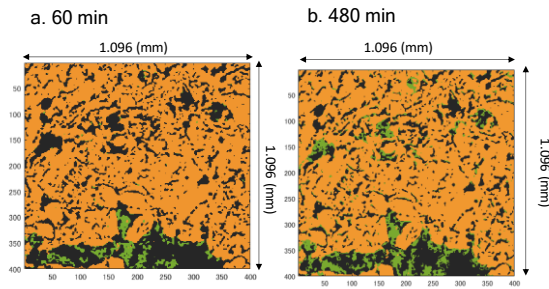


Figure 7: Segmented images (400 x 400 pixels) at (a) 60 min and (b) 480 min after the start of drying (black: void/liquid, orange: solid, green: salt crystals)

4. Conclusion

A series of drying experiments using fired clay brick filled with NaCl salt solution is performed to quantify the salt precipitation on the surface and inside the porous media and its influence on mass transport. The gravimetric tests have shown that the salt reduces the evaporation rate both during the initial capillary drying regime and the subsequent diffusive drying regime. Since the amount of salt precipitation identified from CT images and the amount of salt precipitation calculated from gravimetric measurements were almost the same, the suitability of the use of synchrotron X-ray CT imaging for verifying the temporal evolution of crystal formation in porous materials was confirmed.

In the future, the segmented images consisting of the void, solid and salt crystal phases shown in this paper will be used for the study of influences of the pore size distribution, pore connectivity and tortuosity on the gas diffusivity and moisture permeability. Although the CT images used in the study of this paper were limited, our future study will include more data from the drying experiments presented in this paper.

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