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ScienceDirect

Physics Procedia 69 (2015) 524 – 529

Physics Procedia

10 World Conference on Neutron Radiography 5-10 October 2014

Quantification of water absorption and transport in parchment

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Abstract

Neutron radiography was utilized to quantify water absorption and desorption in parchment at the High Flux Isotope Reactor CG-1D imaging facility at Oak Ridge National Laboratory (ORNL). Sequential 60s radiographs of sections of a 15th century parchment were taken as the parchment underwent wetting and drying cycles. This provided time-resolved visualization and quantification of water absorption and transport in parchment.

Selection and peer-review under responsibility of Paul Scherrer Institut

Keywords: parchment; water absorption; neutron radiography; quantification.

1. Introduction

Parchment was the writing material of choice for thousands of years, but its storage and preservation is still a matter of concern for conservation scientists. Since parchment is not tanned like leather, it is very susceptible to water damage – by exposure to either liquid or high humidity (Hansen et. al 1992). Low humidity dries the parchment out and causes mechanical stresses that can tear it. High humidity, on the other hand, can denature the collagen – producing gelatine. Gelatinization weakens the parchment, making it easier to deteriorate. XRD (Wess and Orgel 2000) and differential scanning calorimetry (Badea et. al 2012) have been used to examine the collagen structure of parchment and to detect gelatinization of parchment. However, these methods cannot quantify or map water content in parchment. While there is a significant body of knowledge on the treatment of parchment and methods to detect gelatinization, little is known about the quantity of water and its movement in and out of

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Selection and peer-review under responsibility of Paul Scherrer Institut
doi:10.1016/j.phpro.2015.07.074

parchment, which would serve to help conservators in best preserving these cultural heritage pieces. Recent studies using image spectroscopy have been employed to monitor deterioration of parchment (Marengo et. al 2011) and to map water content in parchment (Bearman et. al 2012).

Neutron radiography was used here to visualize the absorption and desorption of water from a sample of 15th century French parchment at the High Flux Isotope Reactor CG-1D beamline, Oak Ridge National Laboratory (Bilheux et al. 2014; Crow et al. 2011). Utilizing water calibration studies for the CG-1D beamline (Kang et. al 2013), water movement in and out of the parchment could be quantified. Neutron radiographs were taken as the parchment was exposed to wetting and drying cycles (high and low humidity, respectively). The radiographs were normalized to the starting dry parchment sample and therefore only represent water content in relation to this dried state. Two sections of the parchment were cycled through environments of relatively high and low humidity – one section was dried with the aid of silica gel packets and another was dried by allowing it to come to equilibrium with the ambient humidity level in the Cold Guide Hall at the High Flux Isotope Reactor. In the latter, the holding frame was varied to alter the water absorption pathway.

2. Experimental

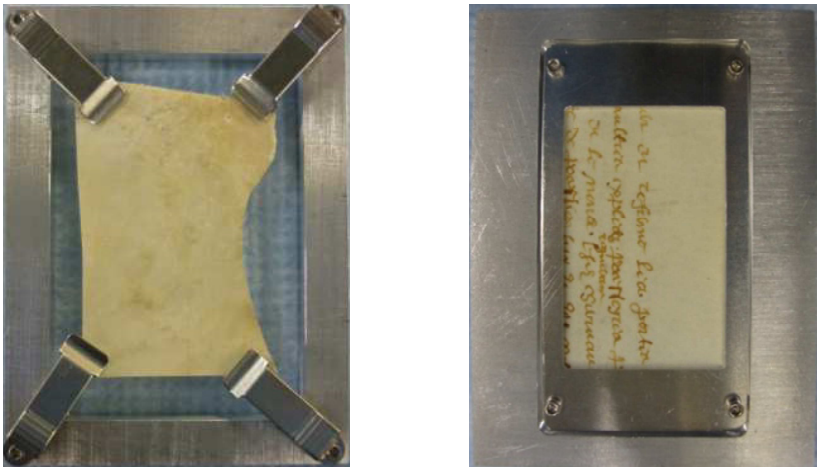
The parchment sections used in the present study were taken from a 15th century French parchment sample, 30 x 45 cm, with an average thickness of 150μm. Parchment section **1** is a corner section (7.8 x 5.3 cm) with a folded edge. Parchment section **2** was taken from the top edge (8.5 x 5.5 cm) and has writing on both sides. While neutron radiography is a non-invasive, non-destructive technique, the preparation of the sample and the environmental conditions surrounding the experiment were not. Sections were cut from a larger piece of parchment and were exposed to extreme humidity conditions.

2.1. Set-up and Data collection

An aluminium (Al) chamber (25 x 10 x 10 cm^3) was designed to house the parchment samples since aluminium is relatively transparent to neutrons. The chamber was constructed with 1mm thick Al and a reasonably tight-fitting lid. A temperature and humidity probe (OaktonLog: RH/TempLog) rested on grated aluminium approximately one inch from the top of the chamber. In order to keep the parchment in the same place between wetting and drying cycles, a removable tray was placed at the bottom of the chamber. This tray was filled with water during the wetting cycles of both **1** and **2**. For the drying cycles of Parchment **1**, silica gel packets were placed in the tray to aid in the extraction of water. For the drying cycled of Parchment **2**, the tray was removed from the chamber so that the humidity level in the chamber could reach equilibrium with the relative humidity level in the Guide Hall.

Two types of frames were designed to hold the parchment. Frame **A** pinched the corners of the parchment while Frame **B** covered all the edges of the parchment and left only the center directly exposed (Figure 1). By varying the frames used, water absorption in areas could be restricted to evaluate the influence of edges on the rate of water absorption. Parchment **1** was placed in a pinch frame **A**. In the first cycle of Parchment **2**, frame **B** was used to restrict the area of the parchment directly exposed to humidity in the chamber. For the second cycle of **2**, frame **A** was used and all four edges were covered with aluminium foil during the wetting cycle to prevent water absorption at the edges. During the second drying cycle of **2**, the top portion of the aluminium foil was removed to compare the rate of water desorption at the covered and uncovered edges. Lastly, in the third cycle of **2**, frame **A** was utilized, allowing all the edges to be exposed.

The frame was attached to the side of the humidity chamber, and the chamber was placed such that the parchment surface was perpendicular to the beam at a distance of 7.5 cm from the detector. Sequential 60 s radiographs were recorded to evaluate water uptake over time. Prior to the cycling of Parchment **1**, **1** was flattened in the frame and allowed to dry in the humidity chamber with silica gel packets prior to taking the baseline images. If the parchment was dried without the frame, the edges would begin to curl. Parchment **2** was imaged after reaching equilibrium with the relative humidity level in the HFIR Guide Hall.



(A) (B)

Figure 1: Frames used to hold the parchment in the humidity chamber: (**A**) pinch frame with **1** and (**B**) solid frame with **2**.

2. 2. Image analysis

All image processing was performed using iMARS (J.-C. Bilheux et al., this issue). The collected raw image files of the wet and re-dried parchment were normalized to dry parchment images collected prior to the wetting-drying cycling. Normalization is applied to correct for background noises, inhomogeneities of the beam profile and detector, and fluctuations in the neutron flux. This is generally done by using open beam (no object present, measuring the transmission of the chamber and dry parchment), $T_{\text{dry parchment and Al chamber}}$, and dark field (beam and camera shutters off), $T_{\text{dark field}}$, images. The normalized transmission of the water, T_{water}, is given by:

$$T_{water} = f_r \frac{T_{parchment\ during\ cycling} - T_{dark\ field}}{T_{dry\ parchment\ and\ Al\ chamber} - T_{dark\ field}} \tag{1}$$

where f_r is a rescale factor to correct for fluctuations in the neutron beam and $T_{\text{parchment during cycling}}$ is the transmission of the parchment during the wetting or drying cycle.

Once the normalized transmitted intensity was extracted, the water content could be examined. The normalized transmission of each pixel was first converted to water volume per pixel. The thickness of water, x, at each pixel was calculated from the transmission through water:

$$ln(T_{water}) = \Sigma x + \beta x^2 \tag{2}$$

where Σ is the attenuation coefficient of water and β is a correction parameter, which corrects for beam hardening and scattering effects. These factors were determined experimentally (Kang et. al 2013) to be Σ=5.542cm^{-1} and β = -2.140cm^{-2} and were implemented in MATLAB to convert the transmitted intensity of each pixel to water thickness, x:

$$x = -\frac{\Sigma}{2\beta} - \sqrt{\left(\frac{\Sigma}{2\beta}\right)^2 - \frac{1}{\beta}\ln(I_{water})}\,. \tag{3}$$

3. Results and Discussion

Radiographs of **1** and **2** after exposure to wetting cycles are given in Figure 2. As mentioned above, the top portion of **1** was folded over. With water absorption, this folded region began to lift. Since the solid frame (A) used for **2** restricted the movement of the parchment, swelling of the parchment with water absorption caused the parchment to curl in areas. Therefore, the average water volume content of the parchment sections were retrieved from several regions of interest (ROIs) that remained flat. The average water volume content per pixel (from representative ROIs, shown in Figure 2) is plotted as a function of time in Figure 3, through wetting and drying cycles. Representative humidity data for the two parchments is also given in Figure 3.

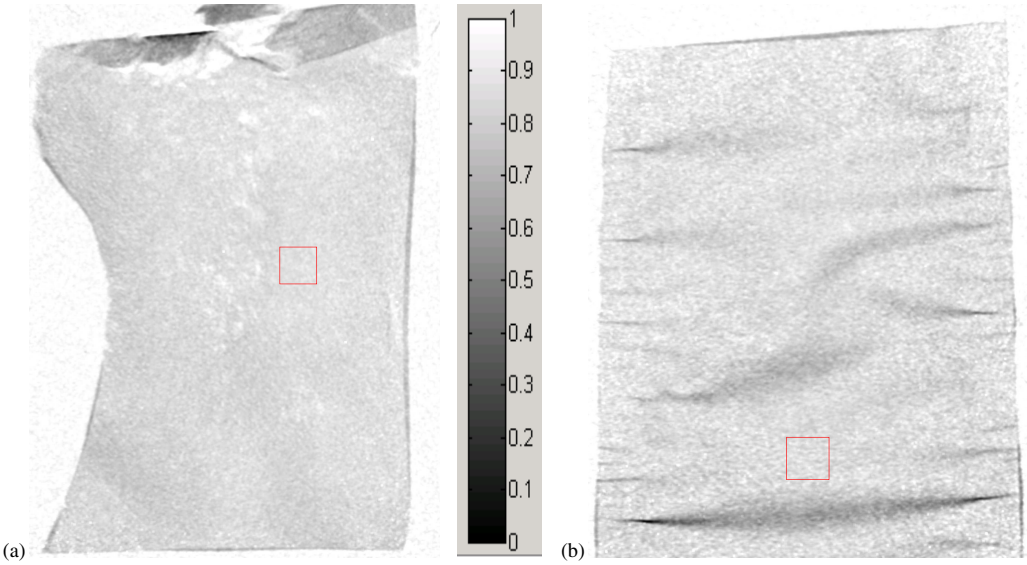


Figure 2: (a) **1** at end of first wetting cycle. (b) **2** at the end of the second wetting cycle. Representative ROIs are displayed.

The average water content absorbed by **2** (7.2 x 10^{-5}mm^3, **2**-second cycle) is much less than that absorbed by **1** (9.5 x 10^{-5}mm^3, **1**-second cycle) because **1** was dried with silica gel prior to the wetting cycle and was therefore at a drier state at the beginning of the experiment and could absorb more water. However, even with the aid of the silica gel packets, **1** does not reach the state of dryness as its starting point (i.e., the water volume does not return to zero), suggesting a degree of gelatinization of the parchment. Parchment **2** does not reach this state until its second cycle after prolonged exposure to high humidity levels. The scattering in the data for **2** is attributed to the instability of the humidity level in the chamber (a secondary humidity probe was inserted into the chamber and chamber could not be sealed as tightly).

The dark edges/outlines observed in the radiographs are due to in-plane swelling of the parchment and not a collection of water at the edges. While the edges of **2** were covered in the first and second cycles, water absorbed by the exposed surface travelled to the covered regions of the parchment, which could not absorb water at the surface.

As can be seen in Figure 3, the slopes of the humidity change do not match the slopes of the water absorption/desorption. In addition, the rate of water absorption in **1** is faster than **2** at the start of their respective cycles. Water absorbed by parchment is related in different ways to its collagen structure depending on the relative humidity (Hansen et al. 1992). The variation in the rates of water absorption may be linked to the variation in how the water is bound to the collagen and may provide insight into parchment gelatinization. This will be explored further in a future publication.

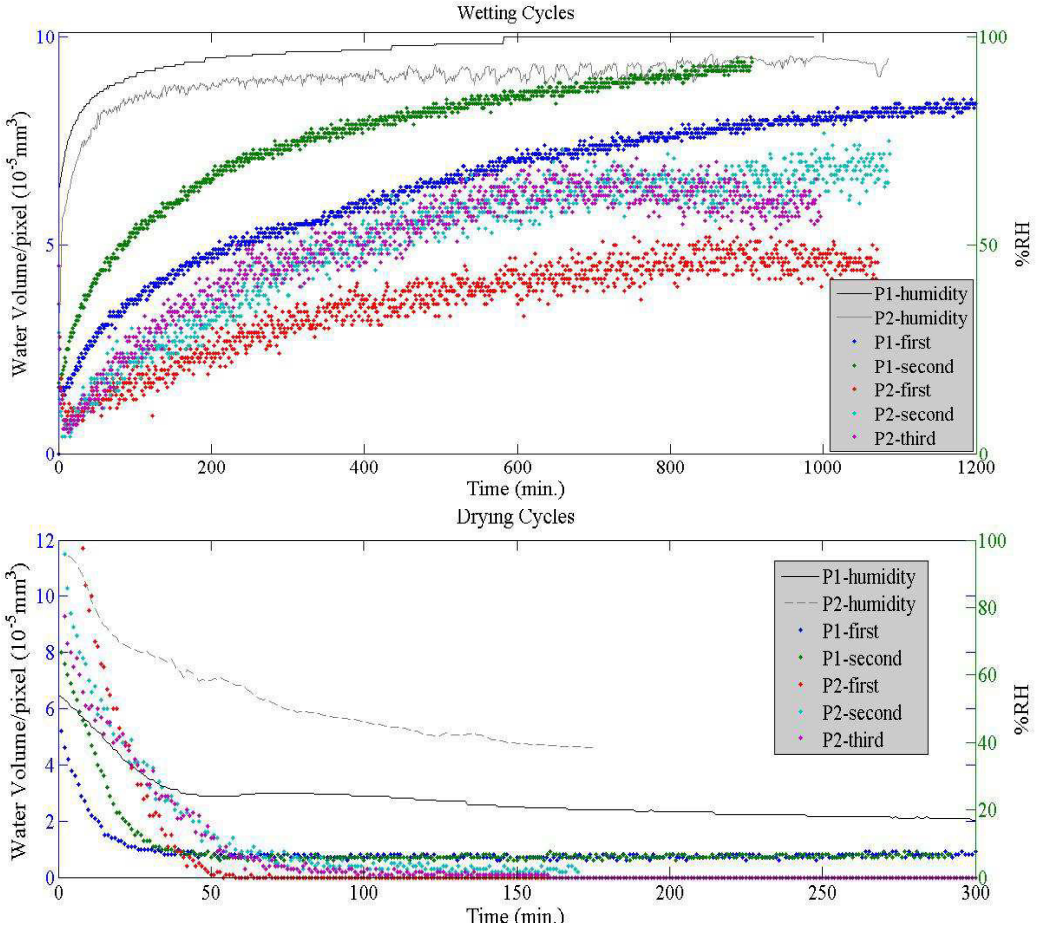


Figure 3: Plot of the average water volume per pixel during (top) wetting cycles and (bottom) drying cycles for selected ROIs in **1** and **2**. The ROIs for **1** and **2** are shown in Figure 2.

4. Conclusions

The aim of this work was to quantify the amount of water absorption and desorption from parchment in humid and a relatively dry environments, respectively. Using the dry parchment images as a baseline, water absorbed from the starting state could be quantified. While water absorption at the surface of certain areas could be halted by application of a frame, this did not preclude water from travelling to these areas. The knowledge gained about the kinetics of water absorption/desorption will provide invaluable information concerning the preservation of such materials and encourage the movement towards viable ways of monitoring such processes.

Acknowledgements

Research at the Oak Ridge National Laboratory's High Flux Isotope Reactor was sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. Department of Energy, which is managed by UT-Battelle, LLC. Funding was provided by ORNL Higher Education Research Experience (HERE) Fellowship and the National Science Foundation, Grant No. DGE0228243.

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