Structural, chemical and hygric characterization of antique Dutch tiles and their susceptibility to salt damage

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1 Introduction

Cultural heritage is exposed to many deteriorating processes around the world. Degradation induced by salt crystallization is one of the main mechanisms that threaten artefacts such as sculptures, ceramics, or frescoes. These artworks are typically made of an assembly of layered materials with different physicochemical properties. Although much progress has been made in recent years to understand the impact of salt crystallization on single porous media, many questions remain unanswered when salt damage occurs in layered materials and on the role of the interfaces between the material layers. This gave the motivation for the JPI-CH project CRYSTINART that aims to develop an integrated approach for modelling and analysis of the decay of artworks due to salt crystallization in layered materials.

In that context, the purpose of this study is to characterize the structural, chemical and hygric properties of antique Dutch tin-glazed tiles from the 19th century, and the effect of these properties on the susceptibility to salt damage. These tiles are ceramics that are mainly made of a mixture of calcium-rich clays coated with glaze. This material is prone to different types of glaze defects induced by salt weathering, they are manifesting by (1) glaze peeling, (2) crazing which is glaze cracking due to high surface tension and (3) shivering which is a process that occurs when the separation between the glaze and the body induces the removal of a portion from the body [1].

2 Experimental study

2.1 Structural, chemical and hygric characterization

The tiles' structure was studied by scanning electron microscopy and X-ray micro-tomography, as depicted in Figure a-b. The tiles consist of a glaze layer of about 380µm thickness and a clay body of about 8mm in height. The glaze itself is in principle non-porous, but air and gas bubbles can be formed during the firing of the tile in the manufacturing process (black square in Figure a-b) [6]. The open porosity of the bi-layered material was determined from a vacuum saturation test (standard NF EN 1936) and also using the X-ray images of the material and amounts to 29%. The full porosity of the clay as well as for the glaze was estimated using CT images of samples of the material, and it was estimated to 26% for the clay part, and 14% for the glaze layer. The tile clay body was analyzed by X-ray diffraction and X-ray fluorescence to characterize its crystallized mineral phases and chemical composition, respectively. The main crystal phases are quartz, calcite, albite and muscovite, and calcium, iron, and lead are abundant. No salts were detected, implying that the source for salt damage must be external.

A common external source of salts is capillary rise [2], [3], [4]. By varying environmental conditions (relative humidity, temperature), the material is then exposed to cycles of drying and rewetting, inducing salt weathering. To characterize the hygric behavior of the tiles, a drying test, as well as a hygroscopic test, was performed with pure water on two types of tiles: (1) with intact glaze, (2) with craquelure (red square in Figure a-b), i.e. with fractured glaze. For the drying test, saturated cylindrical samples of 5mm in diameter and 8mm in height were sealed circumferentially and at the bottom, and subsequently exposed to an environment of 57% RH and 21 °C. As such, drying could only occur via the glaze layer. The samples with craquelure showed a faster drying, which is logical since the vapor permeability is increased in the presence of the fractures. Surprisingly, also the samples with intact glaze dried, indicating that the glaze layer is not fully impermeable. For the hygroscopic test, samples of 2x2x0.8cm were exposed to steps in relative humidity at 23°C, without sealing the surfaces. The hygroscopic moisture content was determined once the samples were in equilibrium with the surrounding environment. The hygroscopic sorption curves are presented in Figure c, showing a change in the hygroscopic behavior of the tile when the glaze is fractured. The higher moisture uptake for samples with craquelure suggests that the fracture network increases the hygroscopic sorption capacity of the tile. Further tests are ongoing in order to elucidate on the moisture transport through the glaze layer.

2.2 Salt weathering

Cylindrical samples (5mm in diameter and 8mm in height) with and without craquelure are being weathered through wetting-drying cycles, using a 5.5 molal NaCl solution and a 1.26 molal Na_2SO_4 solution. The tiles are exposed to the solutions for 10 minutes, then let to dry at 25% RH and 21°C with sealing of all the sides except the one of the glaze. X-ray scans are performed on the tiles before, during and after the salt weathering test to monitor structural changes and crystallization damage.

(a) (b) (c)

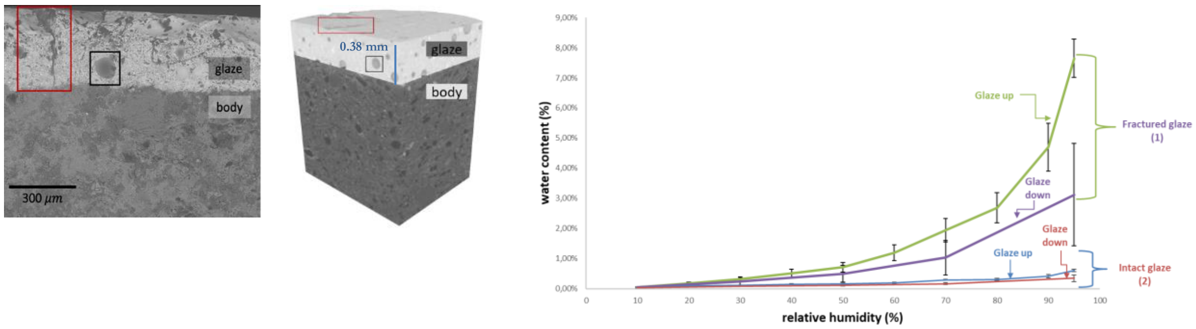

Figure 1: (a)-(b): SEM image and 3D X-ray CT reconstruction of the glaze-body interface. An example of craquelure is marked in the red square and an example of an air bubble within the glaze in the black square. (c) Hygroscopic sorption curve of the Dutch tile: fractured glaze (1), intact glaze (2).

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