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High-speed neutron radiography for monitoring the water absorption by capillarity in porous materials

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Abstract

Fluid flow through porous natural building stones is of great importance when studying their weathering processes. Many traditional experiments based on mass changes are available for studying liquid transport in porous stones, such as the determination of the water absorption coefficient by capillarity. Because thermal neutrons experience a strong attenuation by hydrogen, neutron radiography is a suitable technique for the study of water absorption by capillarity in porous stones. However, image contrast can be impaired because hydrogen mainly scatters neutrons rather than absorbing them, resulting in a blurred image. Capillarity results obtained by neutron radiography and by the European Standard 1925 for the determination of the water absorption coefficient by capillarity for natural building stones with a variable porosity were compared. It is illustrated that high-speed neutron radiography can be a useful research tool for the visualization of internal fluid flow inside inorganic building materials such as limestones and sandstones.

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Keywords: Capillarity; Water absorption; EN 1925; Neutron radiography; Fluid flow

1. Introduction

Moisture transport in porous media plays a role in a wide variety of processes of environmental and technological concern. Moisture inside building stones can lead to their degradation due to freeze/thaw cycles, causing the formation of cracks and spalling. Moreover, the solvent action of water and its dissolved impurities like sulphate, carbon dioxide and nitrate chemically attack building stones. Therefore, an understanding of moisture transport in natural building stones is important for their conservation.

A parameter that characterizes the tendency of liquids to spread spontaneously over solid surfaces is capillary absorption [1]. A test that directly measures the rate of capillary absorption is the European Standard EN 1925 [2], which determine the amount of water uptake per square meter as a function of the square root of time. However, the determination of water absorption by this gravimetric method does not give a very good picture of the distribution of the absorbed liquid inside the building stones. Although there is probably a linear dependence between the capillary rise (m) and the capillary coefficient ([g/m^2]/$\sqrt{}$s)), the correlation factor strongly depends upon the combination of liquid and solid phases [3]. Since the pore network is not shaped like parallel tubes but is a complex

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network with many orientations, the relation between the capillary rise and the capillary coefficient cannot be accurately modeled. The pore surface topology is far more complex so that, as the air/water interface moves through the porous medium, there are many orientations of the local interface which may be stable despite the smallness of the pore size [4].

The internal structure of porous materials is usually such that liquid water is absorbed spontaneously through capillary forces and once absorbed, freely migrates under the action of these same capillary forces. Additionally, these capillary forces are important for water retention inside porous materials. Therefore, capillary force and the velocity of capillary rise, which depends on parameters like density of the liquid, average pore radius, surface tension of the liquid, contact angle, and gravitational acceleration constant g, is one of the basic factors that is determined when characterising a given type of building stone.

In order to determine the capillary forces inside building stones, the European Standard EN1925 was created, based on the fact that a wetting liquid which is brought into contact with a porous material spreads within the open porosity due to surface tension and capillary action until it reaches an equilibrium state. Even isolated liquid regions will eventually come to an equilibrium state since vapour diffusion will transfer liquid by local processes of vaporisation and condensation, thus equalising the vapour pressure throughout.

When the water is absorbed into a dry porous material by capillary forces, some air will be trapped in this process. Most of this air will be displaced ahead of the advancing wetting front and will be expelled, but small fractions of air, originally present throughout the pore network, might become disconnected and isolated. This especially happens when water invades blind pores and also when small regions of the pore network become encircled by the invading water. Although local 'mechanical' equilibrium is established, no equilibrium is accomplished with respect to diffusion. Due to the difference in concentration of dissolved gas in the pore network a gradient will exist between trapped gas pockets and the boundary. It is this gradient that drives a long-term diffusion that causes the air pockets to contract and eventually to disappear.

Besides the determination of the water absorption coefficient by capillarity from a porous material, the visualisation of this internal process could be an important extra tool for the understanding of this phenomenon. Neutron radiography could offer a good method for visualising capillary rise, because H atoms strongly scatter neutrons, while comparatively little scattering is caused by the minerals in most natural building stones. Neutron radiography has already been used to map water content distribution for a wide range of porous construction materials [5–12].

In this study high-speed neutron radiography was performed as a visualisation technique for water movement inside porous building stones. Neutron radiography consists of several important components: (a) the irradiation of the sample by a neutron beam; (b) the detection of the signal; (c) the transformation of the signal information into an image and (d) the further image processing. In general a radiograph is produced by passing penetrating radiation through the material being tested and measuring how much of the signal is attenuated versus transmitted. By using neutrons, the distribution of hydrogenous products and their dynamic behavior can be visualized with a high contrast inside stone material, due to the high sensitivity of neutrons to hydrogen. Therefore neutron radiography and consequently tomography documents a strong contrast between wet and dry regions of partially saturated porous materials. This is an important advantage over X-ray tomography, which provides comparatively little contrast due to the low attenuation of water and the high attenuation of natural building stone. Often contrast agents (like iodine or strong bromine solutions, see [13]) are required to detect the fluid inside the stone when using X-ray CT, possibly affecting the very phenomena that are under investigation.

Because dynamic neutron radiography could offer a better alternative for visualising the effect of capillary forces in porous media, this technique was explored in this study and compared with results from the more traditional technique, the European standard EN 1925.

2. Materials and products

Although there is a wide variety of natural building stones, in this study, two types were selected based on their high porosity and their monomineralic composition. The first is a highly porous bioclastic limestone from Maastricht (Maastrichtian, Upper Cretaceous), while the second one is a quartz arenite of Upper-Landenian age (Palaeocene, Palaeogene), known as the sandstone of Bray [14].

Maastricht limestone is a well-sorted clastic carbonate rock with an average grain size ranging from 0.125 to 0.25 mm (calcarenite) and an average porosity of 52%. Besides some glauconite grains, a small amount of quartz grains, opaque minerals and some iron oxide, it mainly consists of skeletal components of foraminifera, ostracodes, sponges, bryozoa and brachiopods, all cemented with some calcite spar. The average pore-size diameter, determined with Mercury Intrusion Porosimetry (MIP), was 39.0 μm, ranging from 0.0045 μm to 57.7 μm, while the average threshold pressure is located at a diameter of 45 μm [14].

Apart from some accessory grains of rutile, zircon, opaque minerals and chert, Bray sandstone mainly consists of quartz grains. Varying amounts of iron oxide coat the grains. The average grain size of the Bray sandstone is between 0.125 mm and 0.25 mm. Depending on its degree of cementation, the Bray sandstone shows more 'sandy' varieties with a consequent higher porosity of on average 17.6% and more 'quartzitic' varieties that are highly cemented with an average porosity of 6.6%. The pore diameter determined with MIP ranged from 0.0045 μm to 25.3 μm with an average of 15.7 μm and the average threshold pressure is situated between 11 μm (for the more quartzitic