



# Absorption of impinging water droplet in porous stones



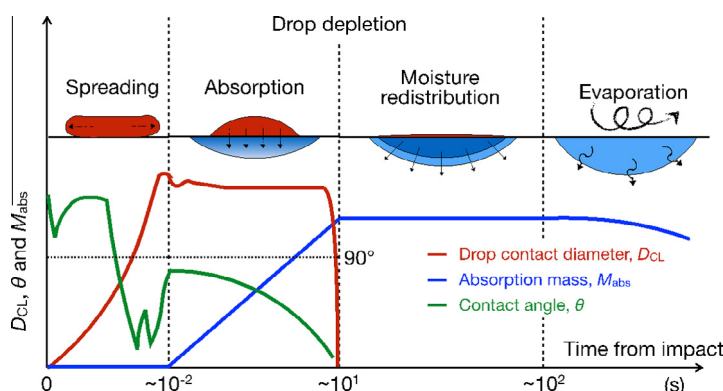
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## GRAPHICAL ABSTRACT



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## ABSTRACT

This paper presents an experimental investigation and numerical analysis of the absorption of water droplets impacting porous stones. The absorption process of an impinging droplet is here fully characterized from spreading to evaporation in terms of absorbed mass during droplet depletion and moisture content distribution in a time-resolved manner for three different natural stones. High-speed imaging and neutron radiography are used to quantify moisture absorption in porous stones of varying moisture properties from deposition until depletion. During impact and spreading, the droplet exhibits a dynamic non-wetting behavior. At maximum spreading, the droplet undergoes pinning, resulting into the contact radius remaining constant until droplet depletion. Absorption undergoes two phases: initially, absorption is hindered due to a contact resistance attributed to entrapped air; afterwards, a more perfect capillary contact occurs and absorption goes on until depletion, concurrently with evaporation and further redistribution. A finite-element numerical model for isothermal unsaturated moisture transport in porous media captures the phases of mass absorption in good agreement with the experimental data. Droplet spreading and absorption are highly determined by the impact velocity of the droplet, while moisture content redistribution after depletion is much less dependent on impact conditions.

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## 1. Introduction

The phenomenon of drop impact on porous media is ubiquitous in nature and is associated with mechanisms found in various industrial applications [12,32]. When a liquid droplet impacts on

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a permeable surface, it spreads on the surface and is absorbed into the porous material due to capillary action. The spreading behavior of the impinging droplet on the surface is known to depend on the properties of the liquid, i.e. density, viscosity and surface tension, impact conditions such as drop size and impact velocity, and the surface wettability and roughness [32]. Absorption is governed both by the properties of the liquid and of the porous medium, i.e. porosity, pore size, wettability [13]. Once the deposited droplet is completely depleted from the surface, the liquid further redistributes within the porous medium due to capillary forces and evaporation occurs at the surface [25].

Although spreading of drop impacting solid impervious surfaces has been the subject of numerous experimental, analytical and numerical studies over the last few decades as presented in two review papers [18,32], the fate of droplets impacting porous media is still far from being understood. Such study needs to consider jointly the droplet dynamics and the porous medium characteristics. In a recent study, we have identified that the spreading of impacting droplets on porous stones is accompanied by hydrophobic dynamic contact angles and a pinning of the droplet at maximum spreading [15,16].

However, the absorption of deposited droplets on porous media is much more rarely studied. Given the difficulty of knowing the status of the liquid within the porous media, absorption studies focused first on the depleting droplet, such as in [6] where the contact radius variation during droplet depletion is modelled. Similarly, Clarke et al. [5] determined the absorption of deposited droplets into microporous filter membranes by measuring the volume of deposited droplet remaining on the surface and proposed an analytical model using Lucas-Washburn equation, assuming only vertical liquid uptake and using a constant permeability. Hapgood et al. [13] investigated the depletion time of droplets deposited on various powder bed substrates and compared the experimental results with the analytical model from [6] by taking into account the measured pore size and wetting properties of the powder bed substrates. However, with different means, described next, of investigating the liquid distribution within the porous media, several experimental studies showed that droplet absorption does not follow Washburn's law for liquid uptake in granular media [11,14,25]. Using a porous medium, Ben Jazia et al. [2] also showed deviations from the expected square root time-dependence as shown by Washburn's law with experiments of drop absorption into hydrophilic nanoporous media formed by polystyrene microbeads assemblies. For this work, they calculated the absorption volume from the drop volume remaining on the surface using the spherical cap approximation. Such unsaturated behavior deviation from Washburn's law was also found in several numerical studies [20,23,26,31]. Markicevic and Navaz [19] showed numerically the transition from fully to partially saturated flow during droplet absorption.

For a better understanding of the absorption process inside porous media, observing directly the liquid content redistribution in the porous medium is required. Absorption in porous media has been studied with several non-destructive techniques, namely X-ray, neutron and gamma-ray radiography, magnetic resonance imaging (MRI) and nuclear magnetic resonance, and with destructive techniques. Reis et al. [25,27] used MRI measurement (of temporal resolution 3.2 min and spatial resolution 78  $\mu\text{m}$ ) to investigate the evaporation of droplets in glass beads substrates. They showed that the general shape of liquid redistributed within the porous media resembles a half spheroid and is comparable to CFD numerical simulation results [25,26]. Jung et al. [14] observed the absorption of impinging droplet with X-ray radiography (with temporal resolution 6.4 s and spatial resolution 9  $\mu\text{m}$ ). They documented the migration of the moisture front inside packed sand samples and distinguished two water migration regions: inertia

influence region and inertia non-influence region based on velocity of the moisture front. D'Onofrio et al. [11] measured the penetration depth of a nerve warfare agent (VX) sessile droplet into sand using a container of varying depth with detector paper on its bottom to measure the time when the liquid reaches the bottom. They showed a good agreement between numerical simulation using continuum model and their experiments. However, the above-mentioned MRI or X-ray studies had low temporal and spatial resolutions to fully capture the absorption process of a droplet in a porous medium.

In this study, we aim to capture the full absorption process of an impinging droplet on natural porous stones by using high-speed imaging, neutron radiography and numerical simulation. Neutron radiography allows visualizing the absorption process inside porous stones. Given neutron radiography limited temporal resolution, we complement this non-destructive technique with high-speed camera measurements for determining the remaining volume on the surface and thus the absorbed fraction. In addition, absorption is studied with finite-element modeling to evaluate the moisture content distribution inside the porous stone. Therefore, in contrast to previous studies, the absorption process of an impinging droplet is continuously characterized during spreading, absorption, evaporation and redistribution, in terms of absorption mass and moisture distribution, in a time-resolved manner.

## 2. Materials and methods

### 2.1. Porous stone samples and characteristics

Three natural porous stones are selected for the droplet impact experiments: Savonnières, Meule and Pietra serena. Savonnières is a highly porous limestone used historically in building façades (e.g. railway station Gare de l'Est, Paris or sculptures of cathedrals of Aachen and Cologne) and thus still in use for restoration purposes [9,10]. The Meule sandstone (grès à meules) is composed of quartz grains (74%), clay and other secondary mineralization constituents and has also been used as a building material (e.g. tower of Strasbourg cathedral) [22]. Pietra serena is a fine-grained and compact sandstone used for building details (e.g. decorative elements of Pazzi and Medici Chapels, Florence). Fig. 1 shows microscopic images of the three stone surfaces.

The porous stones are cut in cubes of  $20 \times 20 \times 20 \text{ mm}^3$  for characterization and drop impact tests. The properties characterized are the bulk density  $\rho_{\text{bulk}}$  defined as the ratio of the dry mass to the total volume, the open porosity  $\Phi$  defined as the ratio of the volume of open pores to the total volume and the saturated water content  $w_{\text{sat}}$  defined as the ratio of the total mass of water filling the open pore space to total volume. Also, the water absorption coefficient  $A_{\text{cap}}$  is determined by measuring the absorption mass rate per unit surface in a free water uptake experiment. The capillary water content  $w_{\text{cap}}$  equals the water content when the water-front reaches the top of the sample during capillary absorption.

The three stones are selected in order to understand the influence of porosity and uptake characteristics on the dynamics of droplet spreading and absorption. The measured properties of the three stones are summarized in Table 1. Savonnières shows the highest porosity, fastest water absorption and largest capillary water content compared to the other stones. The capillary water content is smaller than the saturated water content due to air entrapment during imbibition from a free water surface. Pietra serena shows the lowest porosity, water uptake coefficient and capillary water content. The fraction of the pore space filled by water in the capillary saturated state in Savonnières, Meule and Pietra serena is respectively 56%, 72% and 80%. More details on pore

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