

Micropillars formation via Anisotropic Dissolution of Calcite leads to Hydrophobicity

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The study of the physical-chemical and wettability properties of material surfaces has drawn significant attention in recent years. Here, we show that it is possible to change the wettability of calcite surfaces from hydrophilic to hydrophobic through the formation of micro-pillars by anisotropic dissolution of calcite in an aqueous ammonium chloride solution. The formation of micropillars via dissolution of calcite in ammonium halide salt solutions and the use of the resulting needle-like pillars for transdermal drug delivery has been previously reported [1]. However, the formation mechanism of such structures at the nanoscale, and their effect on the wettability of calcite surfaces have not been studied.

In this work, we promoted the formation of iso-oriented calcite micropillars, creating surface roughness and subsequently leading to an increase in water contact angle. The initial phase of anisotropic dissolution was studied using AFM, and surface roughness was determined by confocal microscopy. Using ESEM, we also studied water droplet formation on the treated calcite surface by promoting condensation during saturation-evaporation cycles. The hydrophobic behaviour was verified using static contact angle measurements (4 μL water droplets) showing an increase from $\sim 30\text{--}60^\circ$ for $(104)_{\text{calcite}}$ prior to treatment up to $\sim 135^\circ$ after micropillar development.

Our findings demonstrate that the proposed strategy could be successfully applied to create surfaces with improved weathering resistance by preventing direct water contact through the creation of surface roughness in materials made up of calcite, such as marble, limestone or lime plasters used in the built and sculptural heritage. This approach presents the first surface treatment viable on an industrial scale, which does not require the application of any coatings. The study also contributes to a better understanding of anisotropic calcite dissolution and the effect of surface roughness on wettability, which may have significant implications for industrial and building materials with improved durability.

[1] Long et al., *Chemistry–A European Journal* 20.15 (2014): 4264-4272.