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Porosity change after gypsum crust formation on macro-porous limestones

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The deterioration of stone is a complex process in which physical, biological and chemical mechanisms are involved. In this research, pore structure changes inside two types of porous limestone were analyzed before, during and after strong acid tests with SO_2. Sulphatation and crust formation phenomena on natural building stones exposed to a polluted environment, are largely described in literature. As far as rocks rich in calcium carbonate are concerned, the main processes involved are the dissolution of the calcium carbonate and the formation of gypsum ($CaSO_4.2H_2O$) in presence of an acid atmosphere. The low mobility of this newly formed salt favours its accumulation in porous materials and at the surface of less porous media. The main actor in the process of gypsum crystallization on limestone is the aggressive sulphur dioxide gas (SO_2).

In this study, the Savonnières and Euville limestone were subjected to tests with a strong acid. According to the standard EN 13919:2002E, samples were put in acid environment for 21 days. At the bottom of the container a mixture of 500 $\pm$ 10 ml H_2SO_3 and 150 $\pm$ 10 ml de-mineralized H_2O was added. No airborne particles or oxides of nitrogen (NO_x) were added. Before exposure, after 6 days in the polluted environment and at the end of the test, the two samples were scanned with X-ray computed tomography (X-ray CT) at the Centre for X-ray Tomography at Ghent University, Belgium (UGCT; www.ugct.ugent.be). This visualization technique allows 2D and 3D reconstructions on a micrometer scale of the internal structure of an object without damaging the material. It thus enables to scan the same sample in a sequential way. In order to obtain information about the sample's interior of which the characteristics can be compared before, during and after the test, the same scanning parameters (exposure time, amount of frames, energy, etc.) were used. In addition, the same adjustments like beam hardening correction, normalizing, ring and spot filter, etc. were applied for the reconstruction. Total porosity, open and closed porosity and radial porosity were calculated for each sample by using the in-house developed software program Morpho+.

The analysis of the various scans revealed that the Euville limestone developed a distinct gypsum crust, behind which a secondary porous layer of 100 μm thickness had developed. Inside the sample the porosity decreased by infilling of the large pore spaces with gypsum. However, after 6 days exposure the total porosity of the sample increased from 5.70% to 8.45%. In this case, the formation of secondary porosity behind the newly formed exterior gypsum layer prevailed upon the crystallizing of gypsum inside the pores located in the sample's interior. Also, the firstly formed gypsum crystals prevented the further interaction of the sulphuric acid with the stone material. After 21 days, the total porosity of the sample still reached 8.45%. The results of the radial porosity measurements were also the same after 6 and 21 days, indicating that the secondary porosity and the filling of pores inside the samples were stabilized.

On the other hand, the gypsum crust on the Savonnières limestone was less visible. No secondary formed porous layer was measured and the total porosity decreased from 12.10% to 10.94% after 6 days and further to 10.31% at the end of the test. The decrease of porosity was still measurable at a depth of 500 μm inside the sample. The combination of micro-CT, image analysis and induced weathering tests are a promising combination of tools and techniques that allow for a better understanding of gypsum crust formation and pore structure change just behind the crust and deeper inside the rock sample.