ARE OUR MONUMENTS MELTING AWAY? EXPLORING THE IMPACT OF CLIMATE CHANGE ON STONE SURFACE FINISHES OF BELGIAN HERITAGE BUILDINGS

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Introduction

With the climate change and its ensuing challenges, we fear adverse repercussions for the materials and structures of our heritage buildings. These concerns include increased rainfall in European countries combined with acidification due to pollution, primarily caused by the rise in CO_2 and the presence of SO_X and NO_X compounds from human activities (IPCC 2023; Saiz-Jimenez et al. 2004). This acidic environment particularly impacts limestone, which is sensitive to the interaction of calcium carbonate ($CaCO_3$) with acid solutions (Basu et al. 2020).

Investigating material alteration involves understanding of phenomena affecting the intrinsic properties of rocks and plays an essential role in the diagnosis and preservation of stone masonry heritage buildings (Trudgill and Viles 1998). This analysis helps us to better understand the behaviour of these limestones and serves as a reference for further investigation in other acidic environment expositions.

Weathering of ashlar, especially in historic buildings, has been observed for several decades (Brimblecombe 2000; Cartwright et al. 2008; MEDISTONE and Bromblet 2010). By controlling independent parameters, it is possible to understand how the material behaves under controlled weathering conditions (pH solution, period cycling). In particular, exposure to acidic atmospheres is a concern that has been studied by many scientists (De Kock et al. 2017; Gibeaux et al. 2018; Menéndez 2018; Rodríguez et al. 2023; Salvini et al. 2022; Vagnon et al. 2021; Vázquez et al. 2015; Yan et al. 2022), with repetitive cycles enabling the correlation of various parameters such as porosity, water absorption rate, roughness, mass loss, pH values...

Methods

Two ornamental limestones were subjected to short artificial exposure: Belgian Blue Stone (BBS - Carboniferous in age) and Gobertange Stone (GS - Lutetian in age). Both rocks are found on historical monuments and newly built constructions in Belgium. BBS is composed of approximately 96% calcite, 1-10% magnesium carbonate, less than 2% quartz, others iron minerals and numerous fossils, mainly crinoids (De Barquin 2001). GS is principally composed of 73-87% calcite, 11-25% quartz and others iron minerals (Pierre et Marbres de Wallonie asbl 2024)

30 x 30 x 31mm cubes have been used for the tests. Density and porosity of samples are first determined. Two test conditions were chosen: exposure to urban synthetic rainwater composed of a mixed solution of 600 ml of HNO_3 (10-5 mol/l) and 400ml of H_2SO_4 (5.10-6 mol/l) carried at pH 5 (Eyssautier et al. 2016; Gibeaux et al. 2016); and to an exaggerated exposure of a stone kitchen worktop with an acid source like vinegar (acetic acid CH_3COOH 7% - pH 2.44). The experiments were carried out during 5 cycles of immersion and drying, without continuous agitation. (Bureau de Normalisation 2003; Xie et al. 2004). Between each cycle, the solution is renewed, starting from a known pH, and measured at the end of each phase. To ensure that the pH remains almost constant during the test, the stone specimens are alternatively immersed in approximately 130mL solutions for 24 hours (passive immersion) and then dried in a ventilated oven for 24h at 100°C.

These tests focus on the characterizing the evolution of exposed surfaces in terms of surface roughness and specific area, by using 3D optical profilometry in laboratory (Nikiema 2024). Other indicators, such as mass loss and porosity variation, are also explored.

Results

Both rocks have different initial properties: porosity less than 1% for BBS and 6-10% for GS. There is a general increase in porosity for all tested samples. Concerning the mass, each phase is characterised by a mass loss, especially with the acetic acid test. Nonetheless, GS seems to lose mass more significantly (rate of cumulative mass lost compared to initial mass after 5 cycles: BBS – 38% / GS – 43 %). After 5 cycles in immersion with acetic acid, each face of the cubes reduced by approximately 2.5mm for BBS and 4mm for GS (Figure 1-a).

A change in texture is observed for the test with acetic acid test during which partial dissolution of the rock matrix occurs. For BBS, the predominantly calcite matrix around the fossils in the rock dissolves first, revealing the fossils and increasing roughness (Phase 1 Surface roughness, all the irregularities characterising the surface, Sa = 3.843µm (std. DV 0.343) - Phase 5 Sa = 105.611 µm (std. DV 14.721)). For GS, the alteration process appears different, with more selective dissolution, revealing quartz grains (Phase 1 Sa = 11.225µm (std. DV 3.801) - Phase 5 Sa = 57.777 µm (std. DV 7.245)).

The tests in the synthetic acid rain do not show a significant trend in mass loss or change in roughness over just 5 cycles, for both rocks (Figures 1,2).

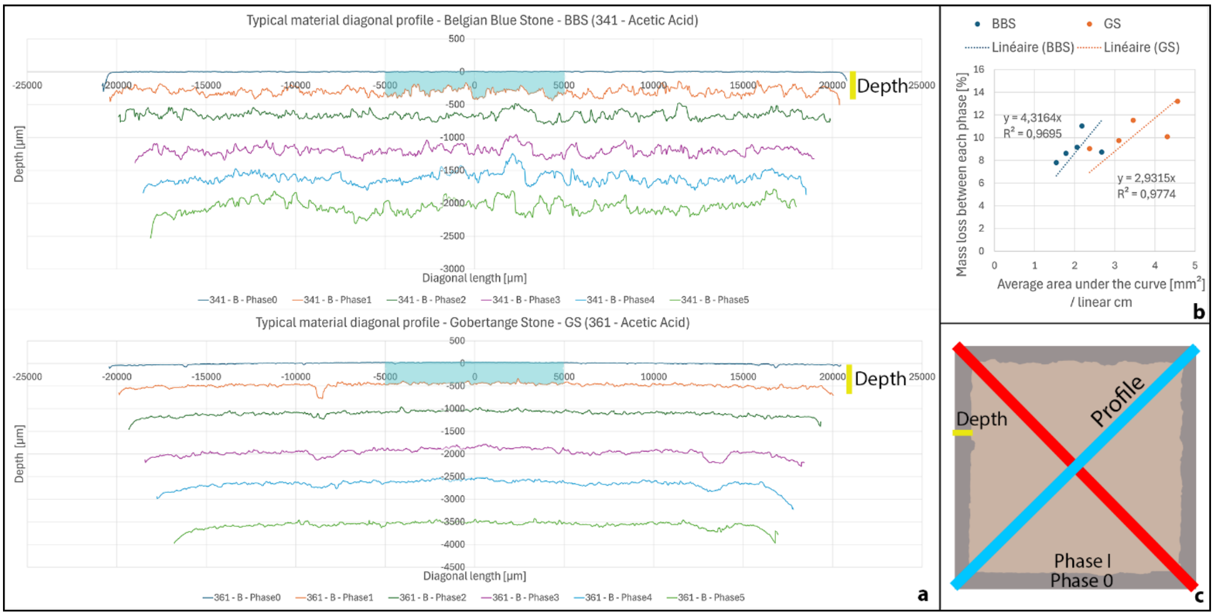

Figure 1. (a, c) Set of typical profiles for immersion of the two stones studied in acetic acid - (b) Correlation between mass loss and area under the curve per phase

Conclusion

These preliminary tests highlight key parameters, such as porosity and main matrix composition to analyse and help to understand the weathering mechanisms of building stones under cyclic acid rains attack.

Results highlight the five immersion/drying cycles had a greater impact with 7% dilute acetic acid (pH 2.44) than with synthetic acid rain (pH 5). This underscores the importance to pursue the efforts of industrialized countries to reduce polluting gases and to protect stone surfaces from household acid attacks, which are irreversible for our stone thresholds and worktops. However, the study, which involved a limited test campaign, does not allow to emphasize the aggressive nature of pH=5 rain. Further tests are necessary on these materials to determine the long-term impact of these pollutants.

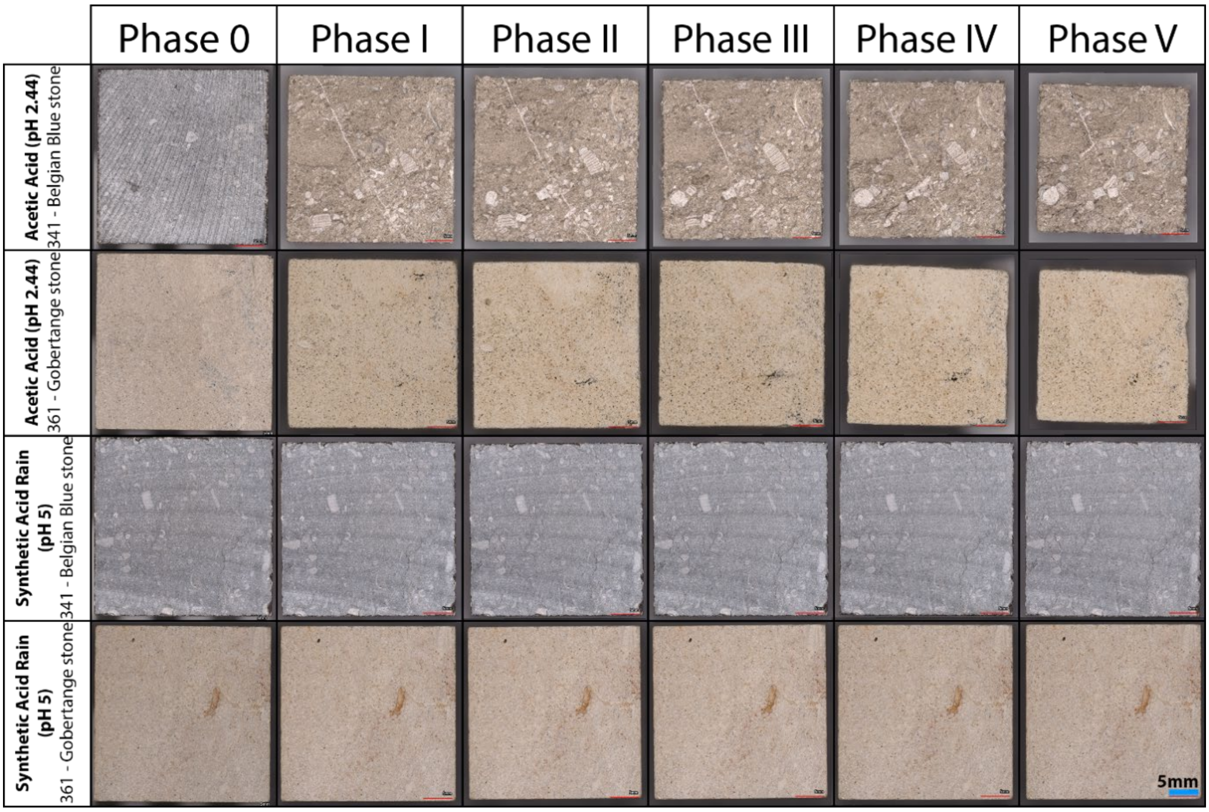

Figure 2 - Appendix. Mapping and evolution of surface finishes following the test phases. There is a noticeable reduction in the size of the samples and a change in texture for both stones following immersion in an acetic acid solution. The surface almost does not change when immersed in a synthetic acid rain solution.

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