STONE RECESSION IN CULTURAL HERITAGE INVESTIGATED BY LABORATORY AGEING TESTS

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ABSTRACT

Carbonate rocks (limestones, marbles) are among the most commonly used building materials (both as dimension and ornamental stones), and are highly vulnerable to weathering, especially in polluted areas. One of the main issues in the evaluation of cultural heritage vulnerability is the quantification of the deterioration rate. With this goal in mind, and in the frame of the HYPERION Project, the water-driven recession of stone in cultural heritage was simulated and measured in the laboratory by exposing a set of carbonate rocks, historically exploited and used in northern Italy, to accelerated ageing tests by immersion in rainwaters with different compositions. The tests were run cyclically in an environmental test chamber, alternating wetting and drying phases; at fixed intervals, the recession of each sample was quantified by bulk weight-loss measurements and surface mapping at the confocal microscope. The relationship between the different mineralogy and texture of the stones and the rate and areal development of their recession was investigated, finding that, besides water pH, calcite grain size is the most important controlling decay factor. Correction coefficients were also calculated for obtaining more reliable recession estimations from recession equations known in the literature. The findings of this study can be exploited for predicting stone deterioration in cultural heritage constrained by the relevant environmental context and the expected future climate change.

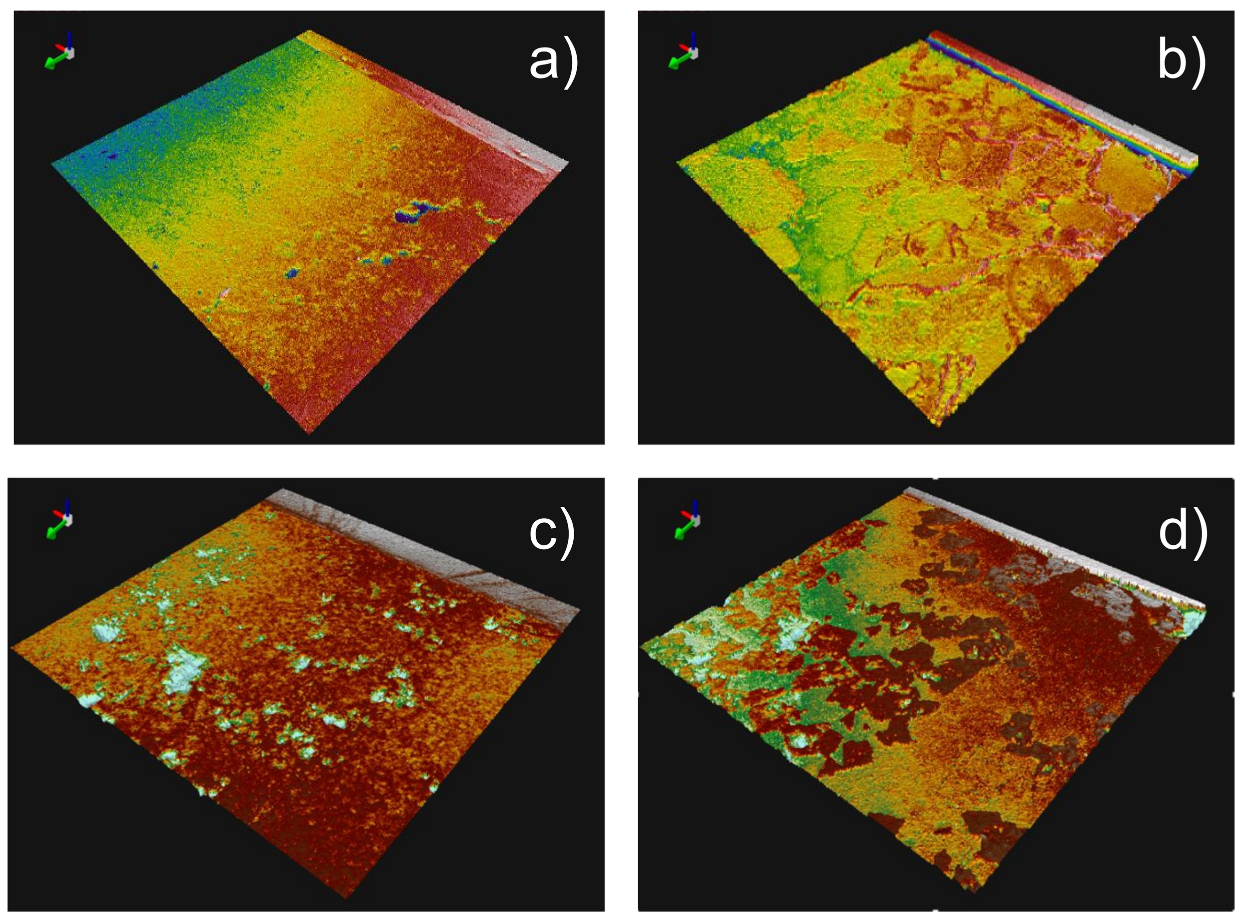


Figure 2. 3D surface model of a sample of Chiampo Paglierino stone (a and b) and of Botticino stone (c and d) before the ageing test (left) and after 240 immersion cycles in the Stresa water (right). The side of the scanned area is about 3 mm. On the opposite side is the stainless still plate used as a reference in the evaluation of stone recession.

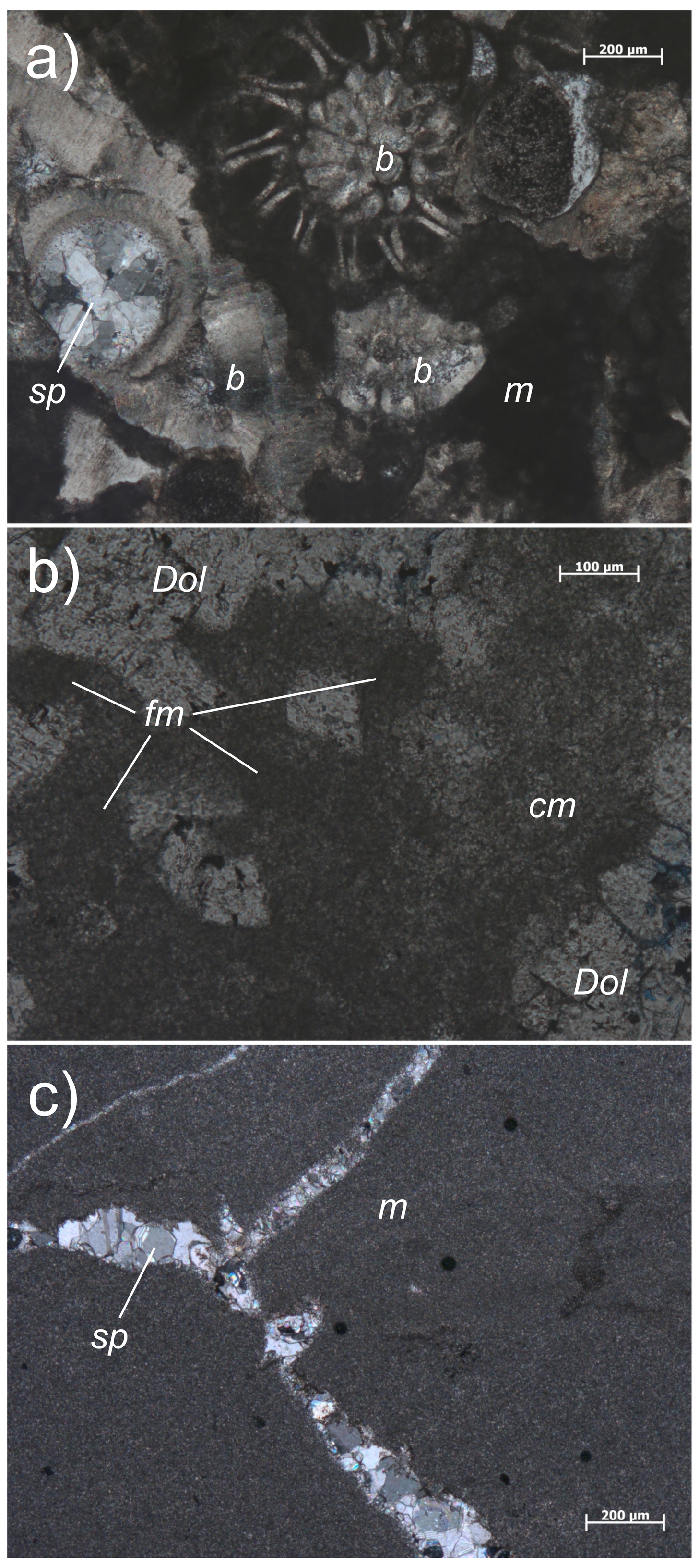


***Figure 1.** Representative photomicrographs of thin sections under cross-polarised light: a) Typical textural elements of Chiampo Paglierino stone (5x) consisting in bioclasts (b), micrite domains (m) and foraminifera chambers filled by sparry calcite (sp); b) Botticino stone (10x) is characterized by domains made of fine-grained micrite (fm) and coarse-grained micrite (cm), containing abundant large dolomite crystals (Dol); c) Orsera stone (5x) is mainly made of micrite (m) with fenestrae and fractures sealed with sparry calcite (sp).*

Materials and methods

The materials to be subjected to the accelerated ageing tests were selected among those most frequently used in the built environment of northeastern Italy. Eleven carbonate rock types were chosen, covering a large range of petrographic and textural features (**Figure 1**): Vicenza (Nanto and Costozza varieties), Carrara marble, Verona (Red and Brown varieties), Asiago, Istria (Orsera variety), Aurisina, Chiampo (Ondagata and Paglierino varieties), and Botticino. They were subjected to accelerated ageing tests in an environmental test chamber for simulating the effect of rainfall, using two different water compositions corresponding to rainwater chemistry in the cities of Bologna (pH ~ 7) and Stresa (pH ~ 6) in Italy.

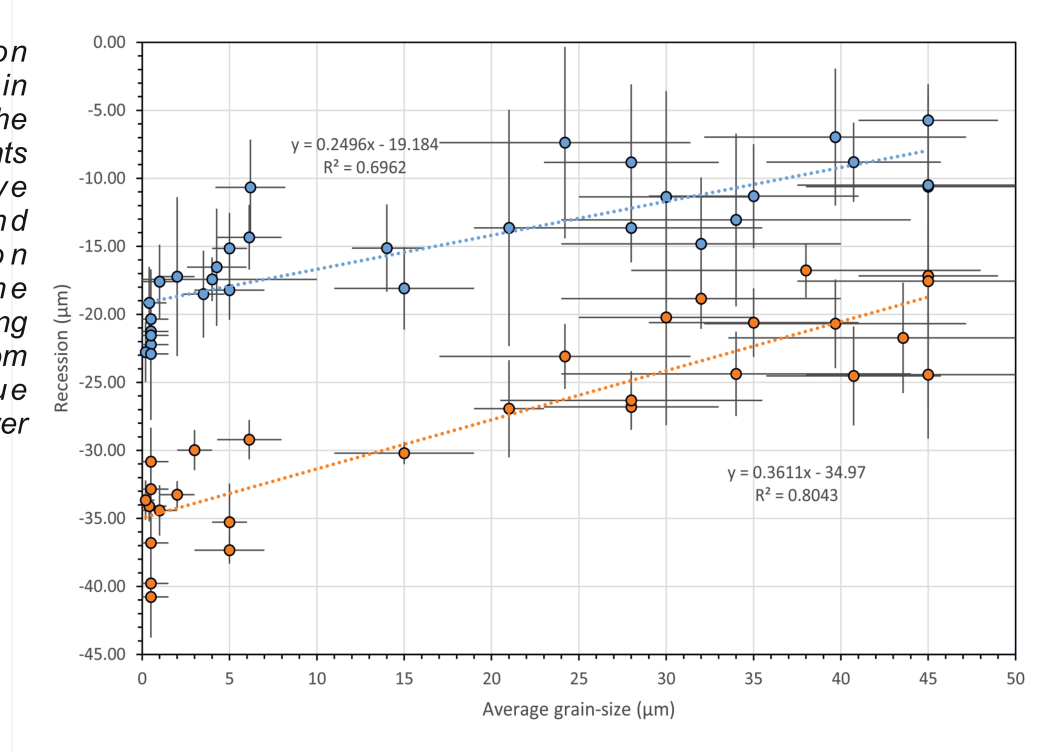
The ageing tests were performed using a benchtop environmental chamber Suntest CPS+ equipped with a Xenon Arc Lamp and an immersion system to cover completely the specimens with water. The samples were subjected to 240 dry-wet cycles, each consisting of one immersion phase and one emersion phase. Each immersion phase lasted 60 minutes, with water temperature of 25°C and under irradiation of 300 W/m2, determining a reference surface temperature of 40 °C, followed by a 180 minutes emersion phase under irradiation of 500 W/m2, determining a reference surface temperature of 70 °C. Bulk stone recession was evaluated considering sample weight loss as a function of the number of wetting cycles. Moreover, direct measurements of recession were performed by confocal microscopy using a confocal laser microscope Olympus Lext OLS4000, which allowed 3D surface reconstruction of the stone surface and evaluation of differential recession as a function of calcite grain size (**Figure 2**).

Results

1) Average recession resulted to be controlled by the water composition used during the ageing cycles; in particular, there is a clear relation with water pH, which is about 6 for that of Stresa and 7 for that of Bologna (**Figure 3**). Consequently, the samples immerged in the Stresa water, characterized by a lower pH, always show higher average values of recession.

2) Stones with markedly different textural characteristics, especially calcite grain-size, displayed significantly different recession values. The analysis of the 3D surface maps obtained by confocal laser microscopy allowed to separately quantify surface recession in portions of the rock characterized by different textural features, such as those made of micrite respect to those made of sparry calcite, or bioclasts. **Figure 2** shows two representative 3D surface maps obtained by confocal laser microscopy on samples of Chiampo Paglierino stone and Botticino stone before and after the ageing test with Stresa water. In the case of Chiampo Paglierino stone, the evolution of the stone surface during the ageing test shows a clear differential recession of the bioclasts, which remain in relief in respect to the surrounding fine-grained micrite and the sparry calcite observed in veins or along bioclast rims. In the case of Botticino, the dolomite crystals markedly "rise" from the receding surrounding micrite; after 240 ageing cycles with the Stresa water, the dolomite shows an average recession of about 0.5 µm, while the micrite reaches values of about 33 µm (fine-grained portion) and 29 µm (coarse-grained portion), respectively.

***Figure 3.** Correlation between average grain size (accounting for the different textural elements and a representative range 0–45 µm) and average recession measured on all the samples after 240 aging cycles with the water from Bologna (upper blue trend) and Stresa (lower orange trend).*



Correction coefficient

The dataset obtained during this study is useful for providing a broad perspective on stone recession considering together a number of petrographic parameters, that is, grain size, heterogeneity of grain-size distribution, content in clay minerals, porosity, etc. The global effects of these properties on stone recession are considered calculating a coefficient Φ, which is proposed for obtaining more reliable projections and predictive trends of stone deterioration for different types of carbonate rocks. The coefficient Φ is the ratio between the recession of a given stone material and the recession of Carrara Marble in the same ageing conditions. **Table 1** suggests other similar stone materials for which the coefficient might be used.

***Table 1.** Suggested coefficients Φ for various carbonate rocks based on the results presented in this study and calculated from the weight loss measurements. These values are also applicable to other stones characterized by similar mineralogical composition, texture, and grain size.*

Stone	Petrographic classification	Φ	Porosity	Notes	Also suggested for
Carrara Marble	Crystalline carbonate	1.00	Low	Grain size ≈ 100 µm	Marbles from Naxos, Paros, Aghia Marina; Pentelico
Botticino Stone	Dolomitic Micrite/ Crystalline carbonate	1.01	Low	Cal 57%, Dol 43%	Indiana limestone
Verona Stone	Biomicrite Wackestone	1.03–1.32	Very Low	High heterogeneity, nodular limestone	Rouge du Roi, Tardos (Hungary), Adneter (Austria), Moneasa (Romania)
Chiampo Stone	Biomicrite/ Grainstone	1.21-1.22	Low		Rouge Languedoc Beige of Missolonghi
Vicenza Stone (Nanto)	Biomicrite/ Packstone	1.23	High	Micritized fossils, clay components, heterogeneity	Portland, Lecce, Noto

THE PARTNERS

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