DETERMINATION OF LIVING ORGANISMS ON STONE WALLS OF THE ARCHITECTURAL HERITAGE BUILDINGS IN SOUTHERN HERZEGOVINA (STOLAC, POČITELJ, HUTOVO)

Original scientific paper

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Abstract

This paper deals with the issue of biocolonization on stone walls of architectural heritage buildings. By applying an interdisciplinary approach, the authors shed light on this phenomenon from the perspective of heritage protection theory and from the aspect of stone conservation. The problem of biodegradation of historical stone is placed in the context of comprehensive procedure of diagnosing and conservation planning, which, while applying an interdisciplinary scientific approach and state-of-the-art technology, must rely on the doctrine of conservation.

Biological colonization was examined on three historical monuments of different age, form and function located at three different sites in Southern Herzegovina. Different research methods were used, including the review of literature, analysis of the existing documentation, and field and laboratory work.

Finally, the paper identifies types of microorganisms found in the samples taken and proceeds to discuss the results from the aspect of their effect on stone material, suggesting avenues for further research.

Keywords: *Architectural heritage, Stone, Conservation, Biodegradation, Deterioration, Remedial treatment, Preservation, Počitelj, Ošanici, Methodology, Diagnostics*

Introduction

Note: during the research and writing this paper, both authors were employed with the International Burch University Sarajevo.

The issue of biocolonization on old stone walls of the architectural heritage buildings is an under-researched topic in the conservationist milieu of Bosnia and there is almost no scientific or professional literature dealing with the subject in any systematic fashion. There are a few possible causes for frequent misperceptions and poor understanding of the problem, including the lack of solid scientific platform and virtually no papers presenting the relevant experience and results of empirical research, e.g. probes in situ or laboratory testing. This lack of relevant information not infrequently results in inadequate treatment of the products of biocolonization. Aggressive cleaning of large surface areas of walls of buildings classified as architectural heritage has even become trendy, although unfortunately it is not always aimed at saving a monument by, for instance, removing the devastation caused by human activity (graffiti), uncovering decorative finish hidden beneath the layer of moss or lichen or removing harmful products that are a result of chemical mechanisms of degradation (disintegration or dissolution), but the aim is rather to refresh the stone, i.e. to attempt to restore original visual characteristics to the material of which the monument is made. As part of these stone-cleaning campaigns, which in some cases are entirely independent projects supported by substantial resources and funding, it often happens that traces of all surface alterations, including deposits, products of decay and even patina, as well as all traces of living organisms, are arbitrarily removed without any prior analysis of its potentially harmful effect. Apart from being methodologically wrong, this approach and uninformed decisions directly resulting from it may cause speeding up of the degradation process and even lead to new physical damage to the monument.[1]

1 “A number of authors have emphasized the damage that can be caused by cleaning: loss of surface, staining, deposition of soluble salts, or making the stone more vulnerable to

Still, we are very far – both in our general approach and in this paper – from advocating a romantic view that has its roots in the history of architecture. It is well-known that extreme interpretations of Ruskin's theory sometimes implied complete passiveness and leaving the ruins "to perish in their beauty" in the belief than only an intact and untreated original is valuable and that the visible traces of decay will add to the value of the object because they ensure presence of history through the *"scars of time"*. [2] The aim of this research is not to try to prove that plants, algae and fungi cannot have harmful effects on a monument's stone structure and that they should not be removed since a greenish film or biological growth in the cavities between stone blocks should be seen as part of a historical character of a monument. Through the methodology that we applied and the results of our research, we wish to point to the necessity of making informed decisions when it comes to remedial treatment of stone surfaces, be it in relation to biocolonization or more generally in response to registered alterations. Accordingly, and in line with the existing principles of modern conservation, we believe that both in the area of prevention and when it comes to active effect on the structure's material, it is necessary – prior to taking any concrete intervention – that mechanisms of the effect of agents to which the material is exposed and their possible consequences be clearly determined. Interpretation of the above in the specific context of our research, implies that we first need to identify living organisms that colonized the stone and then establish what is happening on the surface (or even substrate) of the stone material as a result of their colonization.

pollutants or biological growths. They include Maxwell (1992); MacDonald, Thomson, and Tonge (1992);Young and Urquhart (1992); Andrew, Young, and Tonge (1994); Maxwell (2007); and Delegou and others (2008).", Stone Conservation, An Overview of Current Research, Eric Doehne and Clifford A. Price, The Getty Conservation Institute, 2010, p. 30.

2 "(…) how is the new work better than the old? There was yet in the old some life, some mysterious suggestion of what it had been, and of what it had lost; some sweetness in the gentle lines which rain and sun had wrought. There can be none in the brute hardness of the new carving." The Seven Lamps of Architecture, John Ruskin, 1849.

" Maneuvering between the idea of reconstruction,which crops up sometimes even today, and the occasionally advocated idea of letting the ruin "perish in beauty" (...) the conservation plan must seek the correct path for each individual case (..)",International Principles of Preservation, Michael Petzet, ICOMOS, 2009, p. 20.

Manifestations and consequences of activities of different kinds of living organisms on the same petrographic type are different and the mere presence of living organisms on stone walls cannot a priori, in each and every case, be characterized as a direct cause of degradation. Accordingly, the efficiency of a certain type of treatment, e.g. biocide can vary significantly from one case to the other depending on the concrete circumstances.

On the other hand, recent research has introduced the term "bioremediation" in the conservation of architectural heritage too, particularly through experimental technologies of biocleaning and bioconsolidation.[3] It is obvious that in recent years there is an increasing focus on the issue of biological growth on historical stone, both in terms of coming up with more efficient and targeted interventions aimed at preventing their harmful effects without detrimental side effects and finding alternative cleaning and consolidation technologies.

In light of all of the above, we believe that the issue requires due attention and is to be analyzed within a comprehensive diagnostic-analytical procedure. With respect to determining the causes and level of degradation caused by the physical-mechanic or chemical mechanisms, scientific methodology requires both laboratory and in situ research, and similarly, upon observation of living organisms on the historical stone walls, the required minimum is to identify the types of these living organisms in order to look into their potential effect or take it into consideration in the assessment of synergetic effect of different factors.

This research involves determination of living organisms on stone walls of three important architectural heritage monuments on three different locations in Southern Herzegovina: Ošanići near Stolac, Hutovo and Počitelj. This research was conducted in 2013/2014 and it involved different methods: the review of literature, field observation and documentation, sampling and laboratory work. Apart from resulting in the insight that may be used in future conservation efforts on these heritage sites, this research formulated – in the methodological and factual sphere – a solid platform for further development of this topic, to be undertaken soon.

3 The safety of biocleaning technologies for cultural heritage, Pilar Bosch-Roig, Giancarlo Ranalli, Microorganisms pro- and against cultural heritage, A Frontiers Research Topic, 2014, http://frontiersin.org/microbiotechnology,_ecotoxicology_and_bioremediation

1. Biocolonization or biodegradation?

Terminological differences and nuances in the conservation vocabulary reflect very well the essence of our paper. The following terms are in use:[4]

- "Biocolonization" – refers, exclusively, to presence of living organisms on stone surfaces and does not have connotations relating to its potentially harmful effect. It implies "colonization of the stone by plants and micro-organisms such as bacteria, cyanobacteria, algae, fungi and lichen (symbioses of the latter three)."
- "Biofilm" – which is defined as mono- to multilayered microbial colony attached to surfaces with varying thickness of up to 2 mm. By itself, this notion does not suggest a necessarily harmful influence on the material.
- "Biodeterioration", as a term, clearly points to alterations that have the character of stone degradation (or of other material), i.e. depreciation of its characteristics.

It is clear based on the above definitions that any type of colonization of the stone by living organisms cannot a priori be considered harmful. One recent research concerns the protective role of lichen on oolitic limestone buildings. Observing the forms of rock decomposition as a result of atmospheric conditions indicated that products of lichen colonization can modify and "... fill the superficial porous network with a dense network of lichenised fungal hyphae." The results of capillary coefficient measurement on samples suggest that "... endolithic organic matter can waterproof the stone and could act as a sulfate contamination barrier." [5]

In his overview of current research on stone conservation, Price reports that "[w]hile additional work is needed, research in this area suggests that some surface patinas may be an effective natural protection for carbonate stones, while other biofilms, particularly in polluted environments, may be deleterious." [6]

Finally, the concept of "bioremediation", mentioned earlier in the text, implies natural treatment based on specific metabolic activities of micro-organisms, for instance, "the use of microbial cells and enzymatic activity to remove organic material", or the application of certain bacteria to remove harmful salts through denitrification and sulfate reduction. [7]

2. Manifestations of biodegradation on built-in historical stone

The growth and activity of micro-organisms on stone elements of historical buildings, that is, on stone material of artworks, may result in five basic types of alterations:

- Bioweathering
- Staining or color alteration,
- Surface alterations (pitting, etching, stratification, etc),
- Biocorrosion, and
- Transformation of crystal into small size one." [8]

The types of damage according to internationally accepted classification applied in the area of stone conservation that we can relate to the above-mentioned alterations on stone material include:[9]

4 The terms "biocolonization" and "biofilm" are interpreted according to definitions provided in: ICOMOS-ISCS : Illustrated glossary on stone deterioration patterns, ICOMOS International Scientific Committee for Stone (ISCS), 2008, p. 64.

5 Bioprotective Role of Lichens on Oolitic Limestone Buildings , Nicolas Concha-Lozano, Jacques Pages, Gisel de Billerbeck, Pierre Gaudon, Dominique Lafon, 12th International Congress on the Deterioration and Conservation of Stone, Methods and Materials of Cleaning, Conservation, Repair and Maintenance; The Role Of Biology in Stone Conservation, Session XIII, 2012, Proceedings.

6 Stone Conservation, An Overview of Current Research, Eric Doehne and Clifford A. Price, The Getty Conservation Institute, 2010, p. 21.

7 Heterotrophic microorganisms in deteriorated medieval wall paintings in southern Italian churches, O. Pepe, L. Sannino, S. Palomba, M. Anastasio, G. Blaiotta, F. Villani, G. Moschetti, Elsevier, Microbiological Research 165, 2010, pp. 21-32.

8 The visible manifestations of activity of microorganisms on historical stone are described in literature in more or less detail. The review of consequences of their activity is provided here according to: "Microbially induced deterioration of architectural heritages: routes and mechanisms involved", Tikam Chand Dakal, Swaranjit Singh Cameotra, Environmental Sciences Europe, 2012, pp. 5-13.

9 Classification - types of damage according to: ICOMOS-ISCS: Illustrated glossary on stone deterioration patterns, MONUMENTS AND SITES XV, 2008.

1. Microkarst,

2. Discoloration, with sub-types:

- Coloration
- Bleaching (or fading)
- Moist area
- Staining

3. Pitting

4. Delamination

Note: Efflorescences and biological colonization can be detected in-between the laminae.

5. Erosion

Note: Erosion may have natural and/or anthropogenic causes. It can be due to chemical, physical or/and biological processes.

6. Film, sub-type: Biofilm

7. Patina, sub-type: Oxalate patina

Note: Calcium oxalates are formed as a result of depositing of calcium carbonate with the help of the release of oxalic acid which is a metabolic by-product of lichen and fungi.

8. Crust, sub-type: Black crust

9. Encrustation

Note: The term encrustation is used when the feature is clearly due to a precipitation process, following any kind of leaching. If there is no evidence of leaching and precipitation, the term crust will be employed. [10]

3. Bioreceptivity

The intensity of growth of microorganisms, that is, potential of a rock for biocolonization depends on a number of factors such as "... mineral composition, nutrient availability, pH, salinity, surface texture, moisture content, porosity, permeability, climatic and micro-environmental conditions." [11]

In 1995, Guillitte introduced and explained the notion of bioreceptivity, distinguishing between primary, secondary and tertiary bioreceptivity.[12]

The primary bioreceptivity is connected with the initial potential for colonization of a rock – in this case intact – which implies its relevant characteristics. The alterations that appear over time on the surface of built-in stone as a result of environmental factors may have a significant effect on the initial bioreceptivity, and it is in this fact that we find explanation for "secondary bioreceptivity". Finally, determination of "tertiary bioreceptivity" depends on anthropogenic factors – potential use of consolidants, protective coating or other type of conservatory treatment on historical stone. The link between the use of consolidants or protective coating and changing the degree of bioreceptivity has already been proven. While, for instance, "... alkoxysilane consolidants have been shown to slow or suppress growth" [13] on stone material, the use of some types of water repellents is not efficient from the aspect of passivization and stopping of biodegradation, and "some of them could [even] be a source of nutrients for microorganisms." [14]

If we put definitions of primary, secondary and tertiary bioreceptivity, as provided by Guillitte, in the context of practical conservation methodology, we would interpret them in the following manner: primary bioreceptivity is determined by the type of rock and when it comes to establishing the causes and mechanisms of biodegradation, it is necessary

10 Descriptions of deterioration patterns according to: ICOMOS-ISCS: Illustrated glossary on stone deterioration patterns, MONUMENTS AND SITES XV, 2008.

11 The relevant literature provides, in more or less detail and with some terminological differences, the same factors that influence the susceptibility of stone material to biocolonization, that is, its potential for the growth of microorganisms. The factors indicated in the text are cited from: „Microbially induced deterioration of architectural heritages: routes and mechanisms involved", Tikam Chand Dakal and Swaranjit Singh Cameotra, Environmental Sciences Europe 2012, p. 5.

12 Biodegradation of Cultural Heritage: Decay Mechanisms and Control Methods, Piero Tiano, p. 2.

13 Alkoxysilanes and the Consolidation of Stone, George Wheeler, Getty Trust Publications, 2005, p. 74.

14 "The results of most laboratory studies indicate that some of these water repellents and consolidants—including organosilanes, silicones, acrylics, epoxies, and polyvinyl acetates—appear to have practically no effect on the growth of microorganisms on stone and that, on the contrary, some of them could be a source of nutrients for microorganisms (Price 1975; Bradley 1985; Richardson 1988; Salvadori and Nugari 1988; Santoro and Koestler 1991; Krumbein et al. 1993).", Biodeterioration of Stone in Tropical Environments, Rakesh Kumar, Anuradha V. Kumar, The Getty Conservation Institute, 1999.

that mineralogical-petrographic and chemical characteristics be considered within a diagnostic procedure. Laboratory examination of undegraded samples would be one of the necessary steps to be taken here, especially if the presence of microorganisms that have a harmful effect on historical stone is confirmed. Secondary bioreceptivity would involve the analysis of degraded samples given the synergetic effect of chemical, physical-mechanical and biological agents. Deposits, products of decay and reaction with external agents, differential erosion and other changes to the texture of stone surface are a result of the effect of different factors that may, depending on the concrete conditions, influence in one way or the other the growth of living organisms, the intensity of colonization and spatial distribution of different species. Finally, determining the level of tertiary bioreceptivity would imply establishing any prior use of water repellents or consolidants, which may also be relevant for diagnostics in this context.

4. Field and laboratory work

4.1. Selection of locations

Convinced that the analysis of the effects of biodeterioration and determination of optimum conservatory treatment must be done in the context of general assessment of the condition of stone walls that cannot exclude the agents of physical, chemical and anthropogenic character, in making a final decision on the selection of locations we particularly took into consideration the existence of prior analysis of the condition of historical objects that we wanted to include in our research. For two out of three architectural heritage monuments (Ošanići and Počitelj), there were detailed conservation analyses of stone walls: highly relevant photo-material that also included mapping of deterioration with the relevant classification (Mujezinović, 2005, 2009, 2010).[15]

One of the factors that influenced our choice was the assumption about a different degree of bioreceptivity, that is, different potential for the growth of vegetation and microorganisms.

The following locations, or objects, were chosen:

1. Hammam (Turkish bath) in Počitelj
2. Hillfort in Ošanići near Stolac – Megalithic wall
3. Hutovo old town

Figure 1: Hammam (Turkish bath) in Počitelj

15 See detailed conservatory analyses in: Study on results of investigative work on Šišman Ibrahim pasha's Hammam in Počitelj, with photos of the monument's condition and the analysis of preservation of the existing stone material, Nermina Mujezinović, Cantonal Institute for the Protection of Cultural-Historical and Natural Heritage, Sarajevo, 2006; Main project of restoration, conservation, partial reconstruction, consolidation and adaptation of Šišman Ibrahim pasha's Hammam in Počitelj into a museum/gallery space, Nermina Mujezinović, IGA plan, Sarajevo, 2007; Program of conservatory protection of the archeological site "Hellenistic urban complex at Gradina (Hillfort) in Ošanići near Stolac", Nermina Mujezinović, National Museum of Bosnia and Herzegovina, Sarajevo, 2009; Hellenistic urban complex at Gradina (Hillfort) in Ošanići near Stolac, Determination and classification of deterioration and the condition of megalithic wall with the towers, photo-documentation concerning different types of degradation and graphical documentation, drafts 1- 8, Nermina Mujezinović, National Museum of Bosnia and Herzegovina, Sarajevo, 2010. See also the following papers: Hellenistic town of Daorson – Methodological approach to protection and preparation of the project of conservatory-restoratory works, Nermina Mujezinović, Journal of the National Museum, National Museum of Bosnia and Herzegovina, 2010, Sarajevo and Šišman Ibrahim pasha's hammam in Pocitelj: researching, condition analyses and revitalization project, Nermina Mujezinović, ICOMOS Bosnia and Herzegovina, FIRDUZ-BEY'S HAMMAM: Revitalization of the archaeological site, Sanković Simčić, Vjekoslava (ed.), Mulabegović Ferhad, Mujezinović Nermina, Pravidur Andrijana, Sanković Simčić, Vjekoslava, Simović Snježana, Šekarić Branka, Škarpa Dubreta Darija, Sarajevo: ICOMOS National Committee in Bosnia and Herzegovina, 2013, ISBN 978-9958-9094-6-7, p 62-77.

Figure 2: Hutovo old town, detail

Figure 3: Hillfort in Ošanići near Stolac – Megalithic wall

4.2. Field work and determining the sampling positions

Samples in all three cases were taken from walls, in different areas, where macroscopic examination enabled us to register visible effects of colonization and activity of living organisms, as well as different types of surface alterations. Having done careful observation of stone surfaces and potential different origin and types of stone, we selected our sampling positions. In cases where the available documentation and prior analyses pointed to clear manifestations of stone degradation and prior presence of algae, lichen and fungi (Počitelj and Hutovo), it was possible to make informed decisions about the relevance of samples and adopt a targeted approach in determining the sampling positions.

We faced objective limitations in conducting our field work, mainly in the form of physical accessibility of objects and related hazards. The improvised door in the wall surrounding Šišman Ibrahim pasha's hammam in Počitelj was locked during our field work, blocking our access to the object. The inner yard space is completely overgrown with bushes and high vegetation growing next to the walls, which also blocked our access. Finally, entering the hammam was not possible either.[16]

16 In the course of the investigative work that we conducted a few years ago in relation to establishing the authentic elements of the interior of Šišman Ibrahim pasha's hammam, under the preserved parts of the˝ floor and backfilled material within the hammam, we discovered hypocaustum (floor heating systems) in two rooms in which physical material was mostly preserved in its original form. After that, the hammam was closed and its doors remain closed to this day.

At the location of Hutovo – a desolate, relatively isolated architectural complex which is not in use, nor is it maintained in any way, we faced an entirely different type of danger. Being situated in an environment entirely free from permanent human activity the complex proved conducive for the intensive growth of living organisms, including different types of snakes.

After we spotted them in several places during our initial examination of walls inside the complex, we made a decision to take our samples from the stone wall on the outer side of the complex near the road. Although more recent in origin and physically detached from the hillfort, this wall was built using the same type of stone, and based on visual observation we registered the presence of the same or similar species.

4.3. Sampling and microscopic analysis

The samples of lichen, moss and stains (black stains) were taken by removing the layer from the stone surface. They were placed in clean sterile containers and stored at the temperature of +4 degrees Celsius until further analysis.

The algal samples were taken by scraping the surface layer and placing them in dry and sterile closed containers. Then, the algal samples taken from the stone surface were resuspended in ultra-clean water with a few drops of Lugol's solution. These samples were then stored at the temperature of +4 degrees Celsius. They were kept in the laboratory at this temperature until final analysis.

All samples were taken to the laboratory on the same day.

In the laboratory, the algal samples were shaken up and then a few drops were pippeted out into the well unit, covered with a cover glass and observed under a light microscope.

5. Discussion

5.1. Bioreceptivity

The three locations are situated in the unique zone of Mediterranean climate of flatland Herzegovina, and hence there is no potential for the growth of significantly different species. Small variations in the average annual precipitation, maximum and minimum temperatures, wind frequency, direction and speed cause minor micro-climatic differences that may have more influence on intensive stone degradation caused by other agents than on biodeterioration, which would be a result of more intensive colonization of stone by certain types of vegetation and microorganisms because of conducive climatic conditions.

The altitude for these three locations is different, as is the position of stone structures in relation to the nearest road communication and the settlement to which they gravitate, that is, a larger settlement in the surrounding area that may potentially create specific microlocation conditions due to the presence of pollutants. While Hutovo old town and Šišman Ibrahim pasha's hammam in Počitelj are located either next to or in the immediate vicinity of the road, Hillfort in Ošanići is located at a great distance from the road and it can be accessed only by a dirt road. As for urban environment, this is applicable only in the case of hammam which is situated in the heart of the old town of Počitelj and not too far from another larger town (Čapljina). Hutovo old town is located in an off-the-road area quite distant from a small settlement. The hillfort in Ošanići is located on an isolated plateau in a natural ambience free from any human activity.

The analyzed stone structures are of different age, original purpose and form. None of them are in use. Hutovo and Ošanići are architectural complexes that have the character of archeological sites, while the hammam in Počitelj is an abandoned building that is not in use nor is it maintained.

The stone structures of Hellenistic town of Daorson in Ošanići near Stolac, including the megalithic defense wall with towers and town gates, were built in prehistoric fortified settlement that existed in continuity from the beginning of the early (17/16th century BCE) until the late Bronze Age (9/8th century BCE).

Šišman Ibrahim pasha's hammam, old Ottoman bath, is mentioned in historical records in connection with the name of secretary to the Grand Vizier Fadil Ahmed pasha Ćuprilić, Ibrahim, son of Omeraga, born in Počitelj. A few of the buildings in Počitelj are mentioned in connection with his name, among others han (inn) and hammam. According to Evliya Celebi, these two buildings were constructed by Šišman Ibrahim pasha in 1665.

Hutovo is mentioned in written sources in 1525. A massive round tower, later surrounded by ramparts, is known to have been in existence in 1714 as a derebend /Transl. note: from the Turkish word derebend meaning a mountain pass or gorge/ tower in the Stolac captaincy. Hajji Mehmed bey turned the tower into a fort before the Hutovo captaincy was separated from Vidoška captaincy in 1802. Two of the eight stone slabs erected in various places in the fort record the dates 1796 and 1806. These dates mark the time when a small fortified town was created by erecting ramparts around the tower.[17]

Samples were taken from stone blocks of three different types of walls on three different locations. These blocks differ in their natural (petrographic variety) and artificial characteristics (types of blocks: rough, partly-dressed, dressed stone blocks; dimensions and type of masonry).

Megalithic wall in Ošanići complex is a dry stone wall built with huge stone blocks (standard size of blocks is 0.7-0.9 m thick and 1 m long). The stone blocks on the sides forming the joints are finely cut while most of the blocks on the outer facing sides (excluding the town gates) are roughly dressed. The texture varies from one stone block to the other.[18]

17 Historical information about Hutovo, Hutovo (the Hajji bey's fort) in Hutovo, historic site, Decision on designating Hutovo as a national monument of Bosnia and Herzegovina, available at www.kons.gov.ba.

18 It is noted in many relevant research studies that surface roughness is among factors that have an influence on bioreceptivity. "The surface roughness of concrete has also been previously suggested to influence biofouling susceptibility; specifically, smoother surfaces have been observed to harbor less growth (Pinheiro, 2004).", Molecular characterization of microbial communities fouling concrete infrastructures, David John

Depending on the type of deterioration and condition, the degree of roughness differs because of the effects of fragmentation, peeling, erosion, differential erosion, microkarst, and other different manifestations of the loss of cohesion resulting from different factors (chemical and physical).

In the case of Šišman Ibrahim pasha's hammam, samples were taken from the fence wall built with rough stones of different dimensions and forms joined with mortar. The surface texture is mostly rough, which is partly a result of the cutting of stone and partly of surface deterioration. With respect to entire surface area of the wall, the texture is relatively homogeneous both quantitatively (degree of roughness from one stone to the other) and qualitatively.

The wall surrounding the Hutovo fort is built of partly-dressed stone blocks, more regular in shape, and laminated stones without any binding material. Looking at the entire surface area of the wall, the texture is not homogeneous and surface roughness varies both quantitatively (degree of roughness) and qualitatively (profile and morphology). Sporadically, it is very rough mostly as a result of erosion, differential erosion and other types of deterioration, and some stone blocks are visibly patchy due to the loss of material.

Limestone was used for the construction of all three structures. Based on the visual characteristics of the built-in stone blocks, it may be assumed that the varieties of stone differ in terms of their mineralogical-petrographic and physical-mechanic characteristics, as well as their chemical composition, although it should be said that our best guess is that there are no extreme variations in the relevant values of these parameters.

5.2. Biodegradation

5.2.1. Mechanisms of action

The formation of blue-green algae (cyanobacteria) on surface of walls of the stone structures in the region is facilitated by a gelatinous polysaccharidic sheath of these single-celled organisms.

Giannantonio, A Thesis Presented to The Academic Faculty In Partial Fulfillment of the Requirements for the Degree Master of Science in the School of Biology, Georgia Institute of Technology, August 2008, pp. 8-9. „[A]s a ratio of surface area to projected area... [s]urface roughness can be an indicator of porosity, as well as aggregate size and texture." Ibidem, p. 9.

The sheath acts as a reservoir of water and nutrients during dry spells and prevents drying of algae. At the same time, it serves as the medium for adhesion of the algae to the surface on which they live and for communication between organisms of the same species. Sheaths may be pigmented or not, depending on the type of algae.

The algae, in symbiosis with other microorganisms, create a biofilm on stone monuments that has an impact on the visual characteristics of the architectural heritage monuments. Apart from this aesthetic effect, there are other, more deteriorating, effects caused by these microorganisms.

The first type of deterioration is biogeophysical, which implies mechanical damage caused by biological growth. With the sheath's swelling during humid spells, there is an increased pressure on the stone cracks colonized by the algal growth. The cracks expand and in more serious cases the stone gets fragmented resulting in more damage. Moreover, this process contributes to longer retention of water in the cracks, which in turn enables colonization of stone by many other microorganisms and their uncontrolled growth on the surface of stone structures.

The sheath is directly associated with the other type of deterioration – biogeochemical, because it favors the adherence of airborne particles (dust, pollen, spores) and pollutants (soot, products from combustion of coal, fuel and oil), thus forming deposits that may cause different surface alterations on stone blocks.

Other biogeochemical damage concerns metabolic processes. The biogenic release of corrosive acids (biocorrosion) causes dissolution of stone, sequestering of metallic cations from stone, or the conversion of inorganic substances to acids by redox reactions. Endolithic photosynthetic organisms use these acids (lactic, acetic, glycolic, pyruvic, and other) to dissolve carbonate surface and penetrate deeper into the stone. Cyanobacteria (blue-green algae) have this ability. The algae first colonize a damaged portion of the stone.

Lichen play a major role in biodegradation of stone structures. With their hyphae they penetrate the substratum and damage it further by expanding and shrinking through cycles of drying and moistening. Lichen acids (primarily oxalic acid) dissolve the stone material, and as a result lichen talus accumulates 1-50 percent calcium oxalates depending on the surface (Macedo et al., 2009).

5.2.2. Visible manifestations of biocolonization on stone blocks of the Počitelj hammam, Ošanići hillfort and Hutovo old town

Based on preliminary research and without further laboratory analysis and the necessary in situ tests that would also include instrumental analyses, it is not possible to reach, with any certainty, conclusions about the extent of biodegradation caused by the colonization of stone by identified microorganisms, nor is it possible to exactly quantify their harmful effects. Below are the results of visual observation and our analysis of the existing documentation on the prior condition of stone walls of these monuments.

General condition of the megalithic stone blocks in Ošanići hillfort is not homogenous either quantitatively or qualitatively (this also applies to biological degradation). From one segment to the other, the number, order and position of blocks vary in terms of characteristic pathology, or some type of deterioration of stone material. Almost all types of deterioration are present more or less: discoloration and deposit, detachment, material loss, dislocation, cracks, completely destroyed missing elements and parts. Deterioration caused by anthropogenic factors reflected here in the consequences of previous restoration efforts is a separate deterioration category. (Figure 1, Figure 2).[19]

Biological colonization is most prominent on the north-east facade and on the top of the megalithic wall. Lichen dominate, along with the algal growth while moss is very rarely observed. There is a diffused pigmentation on the stone blocks in the form of white, black and colored stains, and on some surfaces in the wall's interior, visibly moist, we sporadically came across blue-green film. The orientation numerical data about the colonization of the facade by microorganisms and vegetation relating to the condition of stone walls in 2010 is quite illustrative. [20]Out of the entire surface area of relatively preserved megalithic blocks of approximately 5.8 m2, 3.3 m2 was affected by biocolonization while drafts for the remaining approximate area of 2.5 m2 do not indicate the traces of lichen, algae and vegetation in the cavities. On the part of the same wall, approximately 5.2 m2 in size where the blocks have almost entirely lost their shape, biocolonization was registered on the surface area of approximately 4 m2 while the remaining 1.2 m2 of the wall was free of this type of deterioration.

Similar biocommunities were found on the wall surrounding the hammam in Počitelj although biocolonization of stone walls by lichen here is much less represented here compared to the megalithic wall in Ošanići. The growth of plants in the stone cavities, however, is more intensive here. Some sections of the wall evidently retain water and dry harder, and it is in these areas that we detected greenish film on the stone blocks. There is also a sporadic presence of white and yellowish stains affecting a small surface area of the wall.

The characteristic manifestation of colonization of the wall in front of the Hutovo fort is mostly pigmentation, that is, the presence of white, black and brownish stains on stone blocks. There are no moist areas on the observed surface area or areas with greenish or blue-green biofilm.

Figure 4

Figure 4 i **5:** Mapping of deterioration with classification of different types, megalithic wall in Ošanići, 2009, prior recording of stone structures was done by a laser scanner. From: Mujezinović, N. 2010. Determination of deterioration patterns and the condition of megalithic wall with the towers; Hellenistic urban complex at Gradina (Hillfort) in Ošanići near Stolac, study on conservation and protection of the archaeological site. Sarajevo: National Museum of Bosnia and Herzegovina.

19 For detailed description of all types of deterioration, see the studies and technical documentation: Hellenistic urban complex at Gradina (Hillfort) in Ošanići near Stolac, Determination and classification of deterioration and the condition of megalithic wall with the towers, photodocumentation concerning different types of degradation and graphical documentation, drafts 1- 8, Nermina Mujezinović, National Museum of Bosnia and Herzegovina, Sarajevo, 2010; and article: Hellenistic town of Daorson – Methodological approach to protection and preparation of the project of conservatory-restoratory works, Nermina Mujezinović, Journal of the National Museum, National Museum of Bosnia and Herzegovina, 2010.

20 We obtained the data about surface areas affected by biocolonization by calculating the areas from mapping of the damage in 2010, Hellenistic urban complex at Gradina (Hillfort) in Ošanići near Stolac, Determination and classification of deterioration and the condition of megalithic wall with the towers, photo-documentation concerning different types of degradation and graphical documentation, drafts 1- 8, Nermina Mujezinović, National Museum of Bosnia and Herzegovina, Sarajevo, 2010. Surface areas here should be understood as projected areas.

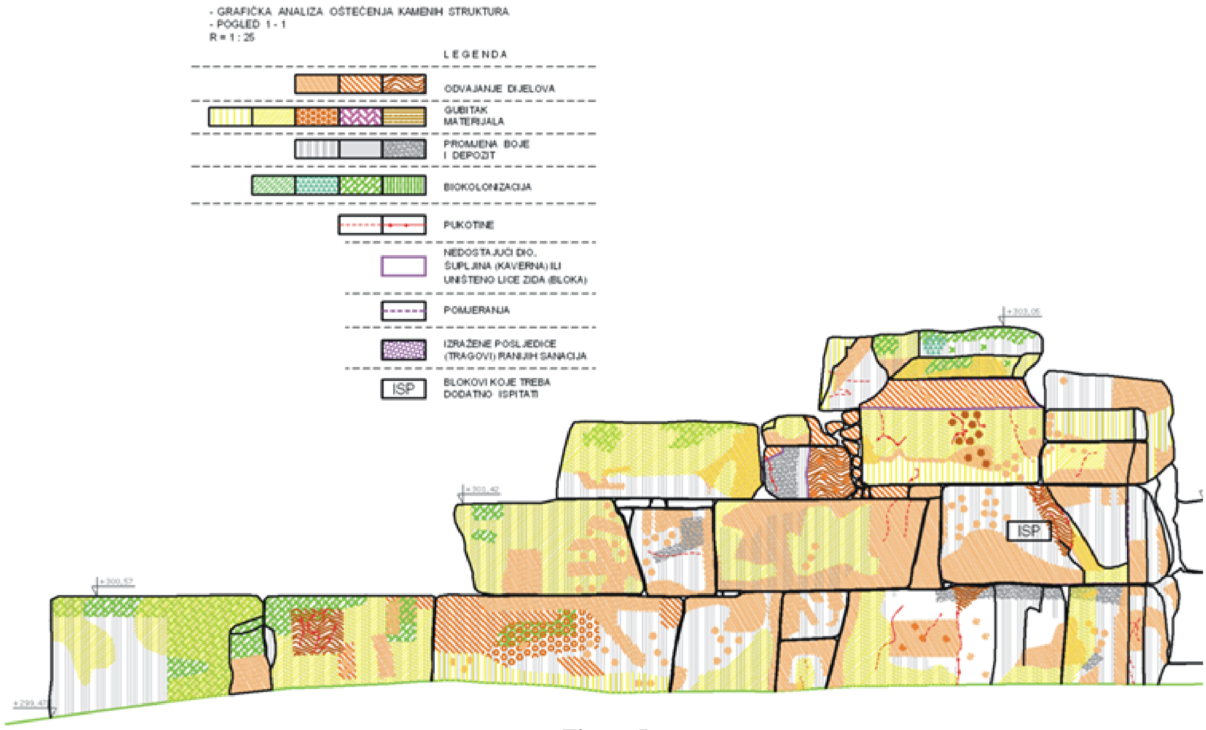

Figure 5

5.2.3. Identified species

At the locations in Ošanići and Počitelj (fence wall), there is a confirmed presence of blue-green and green algae that form a biofilm on stone surfaces affecting the aesthetic appearance of these structures. They result in biogeophysical and biogeochemical types of deterioration, which involves swelling of the algal sheath creating cracks in the stone material that is subsequently further corroded by various acids released by the blue-green algae.

In the samples taken from the wall in front of the Hutovo fort, a type of moss called hair cap moss (Polytrichum commune) was registered. Its effect on stone is mostly physical in nature as it penetrates with its stems the surface layer of stone structure and adheres to the substratum.

The identified species are provided in the table below. (Table 1, Figure 6-10)

Table 1

LOCATION	TYPE	DESCRIPTION
POČITELJ “HAMAM I”:	Nitzchia sp.	Diatom
	Scytonema sp.	Blue-green algae
	Pediastrum sp.	Green algae
	Gloeocapsa sp.	Blue-green algae
POČITELJ “Algae (stone) HAMAM VI” / “HAMAM VII”	Phormidium sp.	Blue-green algae
	Scytonema sp.	Blue-green algae
	Cladophora sp.	Green algae
	Pediastrum sp.	Green algae
	Nitzschia sp.	Diatom
	Gloeocapsa sp.	Blue-green algae
HUTOVO OLD TOWN “black moss”	Polytrichium commune	Moss
HUTOVO OLD TOWN “black stains”	*Thiobacillus sp.*	Bacteria
OŠANIĆI II - orange stains /	*Chroococcidiopsis sp.*	Blue-green algae
OŠANIĆI VIII - green algae	*Gloeocapsa sp.*	Blue-green algae

Figure 6. Sampling position "I", Počitelj, hammam

Figure 8. Sampling position "VII", Počitelj, hammam

Figure 7. Sampling position "I", Počitelj, hammam

Figure 9. Sampling position "VIII", Ošanići hillfort, megalithic wall

Figure 10. Sampling position "VIII", Ošanići hillfort, megalithic wall

5.3. Possible conservation treatment, preliminary recommendations

As the first phase of the project, this research involved a preliminary assessment of biodegradation on walls of stone structures in Počitelj, Ošanići and Hutovo through simple determination of the present species.

In this context, it may be noted that the effects of biodegradation are relative in the quantitative sense too, i.e. its consequences are not equivalent on stone structures in general, and the stone structures included in our research in particular. The possible material loss caused by the dissolution of calcium carbonate and other metabolic activities of microorganisms on rough stone blocks of the megalithic wall of Ošanići hillfort, some of which are over a meter long and 80 cm thick, does not carry the same weight as in the case of, for instance, a stone statue, fine decorative finish or a very shallow relief (rilievo stiacciato). The effect of biodegradation at the location of Ošanići hillfort is obviously incomparable to the effect of physical-mechanical and chemical agents, which are the primary causes of extreme deterioration. In support of the above, it should be noted that on the stone wall in Hutovo old town, we observed very similar visual manifestations of the growth of living organisms and the level of colonization of the surface area by lichen is equally high (both in Ošanići and Hutovo). The stone blocks in Hutovo, however, are in much better condition so that can not even be compared with previous.

In view of the observations noted above and after having studied the effect of micro- and macro-organisms on the structure and appearance of stone blocks in the walls of historical monuments, we may give the preliminary guidelines on further development of solutions on possible treatment.

Thus, general recommendation would be to remove lichen because the review of literature shows that their deleterious influence is the strongest. However, techniques and degree of intervention to be applied on each particular position should be proposed and evaluated in the following stages of the project.

The algae, blue-green and green, also have a harmful effect, but their removal and aggressive clearing off the stone surface can create conditions conducive for the colonization by lichen, which then more easily penetrate the substratum of such deteriorated stone material. In the long run, this type of cleaning would cause more damage than benefit and hence a very careful assessment should be made in each concrete case.

If understood generally, aforementioned preliminary recommendations, can relate mostly to historical buildings and stone assemblies preserved at the level of ruins, stone structures in archeological parks, and generally uninhabited structures of historical importance. As for walls and other stone elements in the interior of historical residential buildings or structures intended for rehabilitation and use, we surely recommend detailed cleaning and complete removal of all micro- and macro-organisms as they pose a health hazard.

5.4. Further research

The continuation or follow-up on this research would entail a clearer quantification of some forms of biodegradation, but also identifying optimum, non-harmful conservation treatment for stone structures in Počitelj, Ošanići and Hutovo.

As a first step in the next stage of the project, we recommend that the types of lichen that colonized the stone structures included in this research be identified. Methods to be applied should include the review of literature, morphological analyses and molecular research of genetic material of unknown species.

The next step would be to study and test different non-aggressive means for the removal of harmful organisms aimed at identifying optimum fungicides for the removal of lichen and mild bactericides and algaecides for partial or complete removal of blue-green and green algae. This is intended to provide for appropriate, non-aggressive remedial treatment of stone surfaces, tailor-made to the condition of monuments in question, where the potentially harmful secondary effects of the used substances and technologies, or their potentially negative interaction with the existing materials, would be reduced to the minimum.

We also propose the examination of short- and long-term effects of micro- and macro-organisms on the thickness and the level of degradation of stone surfaces by using physical-chemical methods, as well as methods of in-vitro examination of the use of nutrients.

6. Conclusion

In the course of our research we established the presence of microorganisms on all three stone walls of architectural heritage buildings in Počitelj, Hutovo and Ošanići near Stolac, first visually in situ and then in laboratory, mostly of algae, fungi and cyanobacteria that, in symbiosis, create a biofilm on stone surfaces. Depending on their level of bioreceptivity, these stone surfaces are more or less affected by biodegradation and more or less altered as a result of biological activity.

At the locations of Ošanići and Hutovo, there is a dominant presence of lichen, less so in the case of wall of the hammam in Počitelj. In the samples taken from the wall of Šišman Ibrahim pasha’s hammam in Počitelj, we identified diatoms, green and blue-green algae, whereas samples from the hillfort in Ošanići revealed the presence of blue-green algae only. In the samples taken from the surface of stone blocks of the wall in Hutovo old town where visual observation indicated black stains, we identified the presence of bacteria.

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References

[1] Macedo, M. F.; Miller, A. Z.; Dionisio, A. and Saiz-Jimenez, C. (2009). Biodiversity of cyanobacteria and green algae on monuments in the Mediterranean Basin: an overview. Microbiology, 155, 3476–3490 DOI 10.1099/mic.0.032508-0

[2] Gadd, G. M. (2010). Metals, minerals and microbes: geomicrobiology and bioremediation. Microbiology, 156, 609–643 DOI 10.1099/mic.0.037143-0

[3] Rogerio-Candelera, M. A.; Miller, A.Z. and Saiz-Jimenez, C. (2009). Contribution of image analysis to the detection of endolithic microbial communities in limestones used in cultural heritage. COALITION No. 17, ISSN 1579-8410, www.rtphc.csic.es/boletin.htm, p.12-14

[4] Krumbein, W. E. (2002). Patina and cultural heritage – a geomicrobiologist's perspective. In Biodeterioration and its control – biotechnologies in cultural heritage protection and conservation; Proc. 5th EC Conference, Krakow, 16-18 May 2002. 39-47. Krakow, Polska Akademia Nauk.

[5] Doehne, E. and Price, C. A. (2010). Stone Conservation, an overview of current research. Los Angeles, Getty Conservation Institute.

[6] Ruskin, J. (1849). The Seven Lamps of Architecture. New York, John Wiley.

[7] Petzet, M. (2009). International Principles of Preservation. Berlin, ICOMOS.

[8] Bosch-Roig, P.; Ranalli, G. (2014). The safety of biocleaning technologies for cultural heritage. Microorganisms pro- and against cultural heritage, A Frontiers Research Topic, http://www.frontiersin.org/microbiotechnology,_ecotoxicology_and_bioremediation

[9] ICOMOS International Scientific Committee for Stone - ISCS. (2008). Illustrated glossary on stone deterioration patterns. Champigny / Marne, ICOMOS.

[10] Concha-Lozano, N.; Pages, J.; Billerbeck, G.; Gaudon, P. and Lafon, D. (2012). Bioprotective Role of Lichens on Oolitic Limestone Buildings. In Methods and Materials of Cleaning, Conservation, Repair and Maintenance; The Role Of Biology in Stone Conservation, Congress Preprints, 12th International Congress on Deterioration and Conservation of Stone, Session XIII. New York, International Institute for Conservation of Historic and Artistic Works.

[11] Pepe, O., Sannino,L., Palomba, S., Anastasio,M., Blaiotta,G. Villani, F. and Moschetti, G. 2010. Heterotrophic microorganisms in deteriorated medieval wall paintings in southern Italian churches. Microbiological Research, 165: 21-32.

[12] Dakal, T. C.; Cameotra, S. S. (2012). Microbially induced deterioration of architectural heritages: routes and mechanisms involved. Environmental Sciences Europe, 24, doi:10.1186/2190-4715-24-36 .

[13] Tiano, P. 2002. Biodegradation of cultural heritage: Decay mechanisms and control Methods. In 9th ARI-

ADNE Workshop “Historic Material and their Diagnostic”. Prague: ARCCHIP. http://www.arcchip.cz/w09/w09_tiano.

[14] Wheeler, G. (2005). Alkoxysilanes and the Consolidation of Stone. Los Angeles, Getty Publications.

[15] Kumar, R., and Kumar, A. V. (1999). Biodeterioration of Stone in Tropical Environments: an overview. Los Angeles, The Getty Conservation Institute.

[16] Giannantonio, D. J. (2008). Molecular characterization of microbial communities fouling concrete infrastructures (M.Sc. Project / Thesis, School of Biology, Georgia Institute of Technology).

[17] Mujezinović, N. (2011). Hellenistic town of Daorson – Methodological approach to conservation and preparation of the project of conservation - restoration works. Journal of the National Museum,52: 307-362.

[18] Mujezinović, N. (2013). Šišman Ibrahim pasha’s hammam in Počitelj: researching, condition analyses and revitalization project. In FIRDUZ-BEY’S HAMMAM: Revitalization of the archaeological site. Edited by V. Sanković Simčić. Sarajevo, ICOMOS National Committee in Bosnia and Herzegovina.

SOURCES

[1] Mujezinović, N. (2006). Study on results of investigative work on Šišman Ibrahim pasha’s Hammam in Počitelj, with survey of the monument’s condition and the analysis of condition of the existing stone material. Sarajevo, Cantonal Institute for the Protection of Cultural-Historical and Natural Heritage.

[2] Mujezinović, N. (2007). Main project of restoration, conservation, partial reconstruction, consolidation and adaptation of Šišman Ibrahim pasha’s Hammam in Počitelj into a museum/gallery space. Sarajevo, IGA plan.

[3] Mujezinović, N. (2009). Program of conservation of the archeological site Hellenistic urban complex at Gradina (Hillfort) in Ošanići near Stolac. In Hellenistic urban complex at Gradina (Hillfort) in Ošanići near Stolac, study on conservation and protection of the archaeological site. Sarajevo, National Museum of Bosnia and Herzegovina.

[4] Mujezinović, N. (2010). Determination of deterioration patterns and the condition of megalithic wall with the towers; photo-documentation concerning different types of degradation and graphical documentation, drafts 1- 8. In Hellenistic urban complex at Gradina (Hillfort) in Ošanići near Stolac, study on conservation and protection of the archaeological site. Sarajevo: National Museum of Bosnia and Herzegovina. Historical Commission to preserve National Monuments, Hutovo’s town (Hajjibeg town), the historic, Decision on designating Hutovo as a national monument of Bosnia and Herzegovina, www.kons.gov.ba.

[5] Kalajdžić, A. (2013). Determination of living organisms on the stone walls of the architectural heritage buildings in the southern Herzegovina (Undergraduate project, International Burch University, Faculty for Engineering and Information Technologies, Department for Genetics and Bioengineering).

DETERMINACIJA ŽIVOG SVIJETA NA KAMENIM ZIDOVIMA OBJEKATA GRADITELJSKE BAŠTINE U JUŽNOJ HERCEGOVINI (STOLAC, POČITELJ, HUTOVO)

Sažetak

Rad se bavi problematikom biokolonizacije na kamenim zidovima objekata graditeljske baštine, koja je vrlo malo naučno i stručno obrađivana u Bosni i Hercegovini. Primjenjujući interdisciplinarni pristup, autori rasv-

jetljavaju ovaj fenomen iz dva ugla: sa stanovišta teorije zaštite baštine, te s aspekta konzervacije kamena kao discipline koja razvija istraživanja orijentirana problematici degradacije, liječenja i preventivne zaštite fizičke građe kamenih spomenika. Problem biodegradacije povjesnog kamena stavljen je tako u kontekst cjelovite procedure dijagnostike i planiranja konzervatorskih intervencija, koji bi se, primjenjujući interdisciplinarni naučni pristup i najsavremenije tehnologije, morali oslanjati na doktrinu konzervacije.

Biološko naseljavanje istraživano je na tri povjesna objekta različite starosti, forme i funkcije, smještena na tri lokaliteta južne Hercegovine. Primijenjene su različite istraživačke metode: pregled literature, analiza postojeće dokumentacije, te rad na terenu i u laboratoriji.

U konačnici, identificirane su vrste mikroorganizama pronađenih u uzorcima, a rezultati prodiskutovani s aspekta njihovog uticaja na fizičku građu, daljih istraživanja te preliminarnih preporuka za razvijanje solucija konzervatorskih tretmana.

Ključne riječi: *prednapinjanje, ušteda, konstrukcija, rasponi, BBR*

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