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HOLISTIC APPROACH TO CLEANING AND PROTECTION OF STONE FAÇADES OF 20TH CENTURY ARCHITECTURAL HERITAGE

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Abstract. *Modern urban areas represent our recent past with great aesthetic and heritage value which should be maintained and preserved for the future. One of the best recognized landmarks of the city of Novi Sad, capital of Vojvodina region positioned in northern Serbia, is the architectural masterpiece Banovina Palace in which today resides provincial government. It is an elongated, closed, horseshoe-shaped structure, built in late 1930s in a modern style and influenced by expressionist architecture. Two-floor high with a tower on one end the building dominates the scenery and symbolizes the appearance of a large ship on the Danube River which flows in the immediate vicinity.*

The façade of this famous building is covered with white marble from Adriatic island of Brac. After almost 80 years of exposure to weathering, urban pollution and manmade devastation, the façade was in need of cleaning, consolidation and protection. In the attempt to prepare for the European Capital of Culture 2021, the City of Novi Sad started ambitious project of local heritage revival. Due to awareness of lack of understanding about stone nature and behaviour, stone cleaning and protection usually raises concerns and cautious. The Laboratory for Materials in Cultural Heritage, Faculty of Technology, was invited to join the project to preform holistic characterisation of the stone, reveal deterioration mechanisms, and propose conservation methodology.

Using mobile laboratory with non-destructive techniques coupled with laboratory testing, the stone façade was comprehensively analysed. Various deterioration patterns were identified including patina, dirt deposits, eroding surfaces, yellow layers and black crusts, large amounts of carbonaceous particles responsible for black appearance of stone surface, soluble salts, microbiological corrosion, residue of inadequate graffiti removal, façade paints, acrylic binders, as well as mechanical damage of stone panels. Based on laboratory testing of a number of cleaning, consolidation and protection techniques and products, the *most promising ones were also tested in situ [1]. Products ranging from traditional conservation approaches to innovative solutions like self-cleaning photocatalytic coating as final protective layer, were selected [2]. Resulting from research the methodology for cleaning, consolidation and protection was established in 2017 and implemented in 2019, where the laboratory acted again as scientific supervision and control of the conservation works. The presented approach allowed deep understanding of the complex problem and guaranteed responsible conservation strategy; therefore it stands as an example of 20th century architectural heritage preservation.*

1 INTRODUCTION

Stone cleaning and protection presents an ongoing challenge for architectural conservators through the world for hundreds of years. Since early antiquity natural stone is used in architectural projects in many forms for its attractiveness, durability and strength. For façade coverage thinner stone blocks of standardised dimensions are deliberately chosen by architects as readily available option for outstanding appearance of the buildings. As natural material, stone is prone to weathering and deterioration, which have been observed as dramatically increased in the last century due to changes in our way of urban living and micro environment (local climate, atmospheric conditions and air pollution). The need to preserve architectural heritage in general has many implications including cultural, historical, economic and ecological values [3]. Preservation and maintenance of stone masonry raise public and policy-makers concern over the investments required to enjoy clean buildings.

The cleaning of stone may change its colour and surface characteristics, and affect quality and appearance, in some cases inducing irreversible damage if approaches, materials and methods are inadequately chosen. Therefore the stone cleaning should be performed with the utmost caution, aiming to deliver visually improve surface appearance, but not jeopardize the fragile character of the surface (not to make the cure worse than the disease). Also the need for cleaning should be carefully considered. The appearance of the stone building surfaces could be rather pleasantly weathered then dirty, or the micro environmental conditions could be unfavourable, meaning that the effects of cleaning would not last very long.

All potential materials and methods for stone cleaning should be priory studied for damage or adverse reaction in laboratory and on test panels. There is a number of inadequate approaches including abrasive methods and specific chemical cleaners (strong acidic and alkaline agents), including partial cleaning and lack of putty and mortar joints treatment [4]. The laboratory testing should record physical and chemical characteristics, establish short and long term effects of cleaning and monitor changes of the stone surfaces. The selection of materials and methods should also take into account the impact on environment and operator.

Various deterioration patterns and mechanisms may occur on stone surfaces. They include patina, dirt deposits, eroding surfaces, yellow layers and black crusts, large amounts of carbonaceous particles responsible for black appearance of stone surface, soluble salts, microbiological corrosion (colonisation of algae, lichens and mosses, bacteria, moulds), graffiti, façade paints, acrylic binders as well as residues of their inadequate removal, iron and copper staining, and manmade mechanical damages of stone surfaces [5-8].

Carbonate stones, such as limestone and marble, are much more readily to dissolve in cleaning solutions compared to silicate-based stones, what requires specific approach to their cleaning. Likewise, decolouration and soiling of carbonate stone surfaces usually presents not only a layer of dirt, but an organic film physically adhered to the surface as well. In these cases chemical reactions, such as gypsum formation, bond the soiling agents to the surface even more tightly and thus increase the risks of making the stone surface more vulnerable after the cleaning treatment [9].

To guarantee evidence based cleaning and protection of stone, an interdisciplinary team of scientists and professionals is required, capable to understand the stone structure, reveal decay mechanisms, "heal" the stone and prolong the life of stone artefact, enabling its functional and aesthetic properties to last.

The case study presented in this paper describes the use of inspection methods, non-destructive techniques, laboratory and in situ testing on the stone façades of significant architectural heritage of XX century. The whole cycle started from the original substrate characterisation and the establishment of conservation methodology for stone façade cleaning, repair and protection in 2017, and continued with the scale up, implementation and control of contractor's works in 2019, with excellent results obtained. Therefore, it stands out as a good practice example of both an interdisciplinary project and management approach to architectural conservation.

2 BRIEF HISTORY

The Banovina Palace is nowadays a building complex of the Provincial Government and the Assembly of the Autonomous Province of Vojvodina. It was built in a modern style and influenced by expressionist architecture. Shaped by the aspiration to symbolize the economic power and prosperity of the Danube Banovina, the Palace still fascinates with its monumentality and appearance. Construction started in 1935 and completed in 1939. The Banovina Palace stands as the most famous work of the Serbian architect Dragiša Brašovan (1887-1965). In the period from 1939 to 1941, the Palace was the centre of the Danube Banovina (regional administration unit) and it's Governor. After the occupation of the Kingdom of Yugoslavia in 1941, the Banovina Palace was the headquarters of the Military Administration and National Police. After the Second World War the provincial Government and the Assembly again reside in the complex, while throughout the mid-fifties, in the representative rooms of the Banovina Palace there was also the Military Club in which various cultural events were organized.

The Banovina Palace (Figure 1) is one of the most beautiful buildings of the twentieth-century architecture in Serbia. The complex dominates the urban scenery and symbolizes a massive ship cruising on the Danube River, which flows in the immediate vicinity. Locals often call it a "white cruiser" because of its shape and white marble façade. It was built on the area of 28.000 square meters. The building was erected along a new boulevard, in the shape of a horseshoe base, 185 meters long and 42.5 meters wide. The height (except for the "tower") is about 20 meters (the building has five floors, from the basement to the attic), and has five entrances, two of which are central, representative and official. The entire architectural composition of the palace is accentuated by a 42 meter high tower on the northeast corner. The complex consists of two buildings: the palace with 569 departments, mostly office spaces, and the residence with large assembly hall and 147 rooms, some of which were luxuriously decorated at the time of construction. The building façade is covered with natural stone panels made of white marble from Adriatic island of Brac (Pučišća quarry).

Preparing to take over the prominent role of European Capital of Culture 2021 the City of Novi Sad opted for intensive development of its cultural and creative potentials aiming to strengthen cultural vitality of the city and enable urban regeneration. After almost 80 years of exposure to weathering, urban pollution and manmade devastation, façades of the Banovina Palace were craving for cleaning and repair. Being aware of the challenges natural stone cleaning and conservation bring forward, the Laboratory for Materials in Cultural Heritage was invited to join the project with its mobile equipment and expertise in historical materials conservation.

Figure 1: Banovina Palace after the treatment in 2019

3 FEASIBILITY STUDY

The feasibility study was carried out by the Laboratory for Materials in Cultural Heritage, Department of Materials Engineering, Faculty of Technology, University of Novi Sad. The aim was to determine the most suitable approaches (materials and methods) to cleaning, repair and protection of stone façades. The study started with visual and non-invasive in-depth in situ assessment of the degradation types and intensities, and deterioration mechanisms identified on the façade stone surfaces. The study also considered all available alternative materials and methods available on the regional market, including water washing, poultice cleaning, chemical and abrasive cleaning, repair mixtures and anti-graffiti protection and self-cleaning coatings [2,10]. Treatment trials were carried out in laboratory and in situ on the selected test panels (small façade areas). The findings of trials and investigations were elaborated in the Feasibility Report. The most appropriate forms of cleaning, repair and protection were recommended.

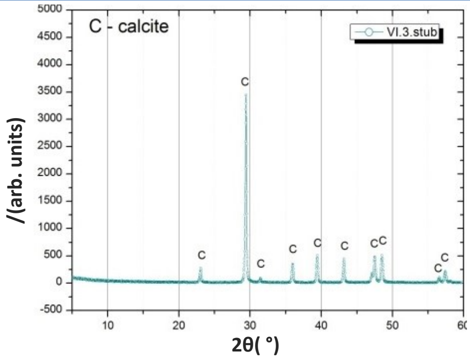
To characterize the original stone, reveal deterioration mechanisms, and establish conservation approaches, the Laboratory used laboratory and in situ testing equipment: stereo optical microscope (OMANO OMXTL/V7 Articulated Boom Microscope), Fourier Transform Infra-Red Spectroscopy (Alpha Bruker Optics), X-Ray Fluorescence Spectroscopy (mobile ARTAX 200 µ-XRF spectrometer, BRUKER Nano), spectrophotometer (CM-700D, Konica Minolta), test stripes for soluble salts detection (Quantofix), microbiological analyses, X-ray diffractometer (PW 1050, Philips), mercury porosymeter (Autopore 9500, Micromeritics), IR thermographic camera (T440bx, FLIR), and drilling resistance measurement (DRMS SINT Technology).

3.1 In situ and laboratory stone characterisation

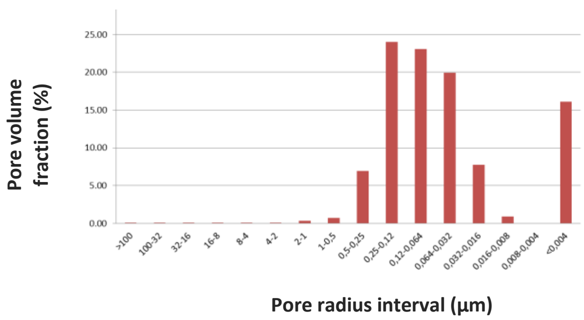
Mineralogical composition determined by X-ray structural analysis (XRD) confirmed that the tested sample of the original stone material is mineral calcite ($CaCO_3$). Calcite is the only mineral identified in this natural stone. These findings were confirmed by XRF analysis (alumosilicate origin with impurities), while Hg porosimetry provided information about material pore sizes and distribution, Table 1.

Table 1: Stone sample characterisation: XRD, XRF and Hg porosimetry

X-ray diffraction (XRD)



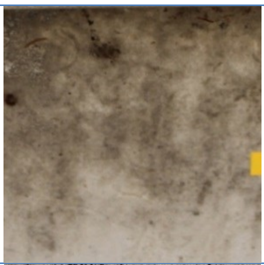
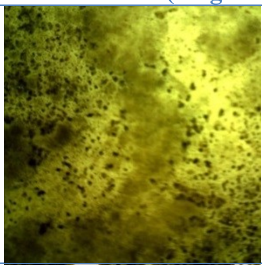
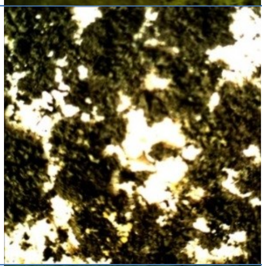
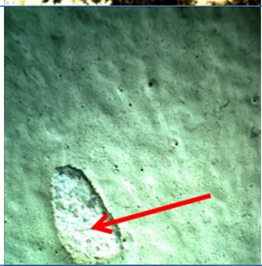
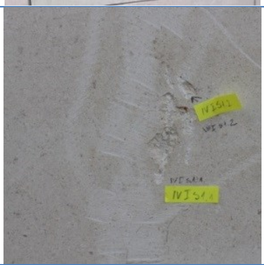
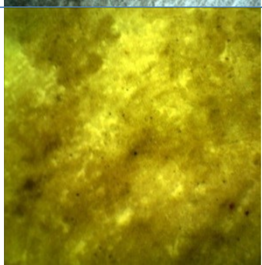
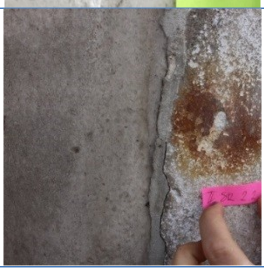
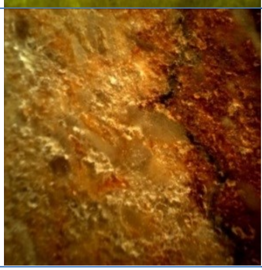
Hg porosimetry



XRF	Element	Al	Si	P	S	Cl	K	Ca	Fe	Zn	Sr	Ba
	(%)	1,013	7,278	0,491	0,200	0,355	0,639	**53,102**	0,054	0,071	0,041	0,026

In situ mapping of characteristic surface filth, deposits, discoloration and microbiological growth was performed on the entire façade area. Based on the mapping 70 zones were selected for detailed visual and microscopy characterisation, spectrophotometry, XRF and FTIR investigation, microbiological analysis and soluble salts identification. Some of the deterioration patterns are presented in Table 2.

Table 2: Deterioration patterns

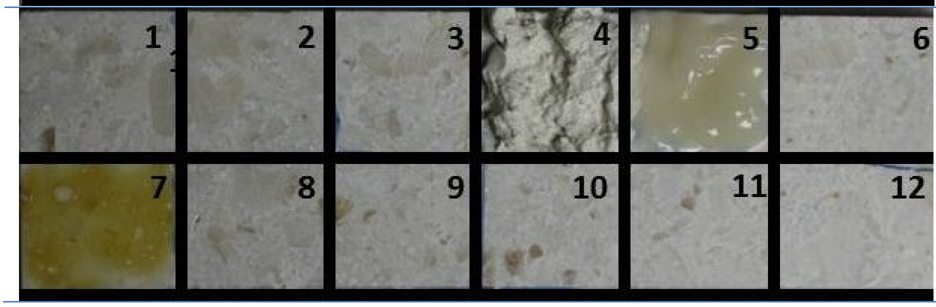
PHOTOGRAPH	MICROSCOPY (mag. *x6.5*)	IDENTIFIED DETERIORATION
		Surface sulphatation with black crusts, eroding surfaces (pitting)
		Advanced microbiological corrosion, soluble salts (chlorides: 19mg/l). Microbiological analysis confirmed the presence of aerobic mesophilic bacteria, a small number of aerobic oligotrophic bacteria, ammonifires, yeasts and moulds.
		Acrylic facade paint based on styrene-acrylic copolymers. Below the paint layer, the stone surface is in a very poor condition due to its degradation in reaction with the facade paint.
		Significant mechanical damage (scratches). No patina or black crusts observed. The values of total colour change (colorimetry) are below the level of visual change detection. An increased content of soluble salts sulphate and chloride.
		Presence of calcium carbonate, calcium sulphate and soluble nitrate salts confirmed by FTIR analysis. The surface layer is missing. Chlorides presence is due to the diffusion of salt ions from technical NaCl used in the winter.
		Iron staining typical for corrosion of iron reinforcement and anchors in the presence of soluble salts (sulphates) under conditions of increased humidity. Deep cracks and missing fragments. Soluble slats (sulphates: 750 mg/l, chlorides: 500mg/l).

The in-depth characterization revealed following deterioration patterns: microbiological corrosion, atmospheric deposits, surface structural alterations (patina, yellow layers and black crusts), layers of paint (acryl – façades paints, and alkyd - graffiti), soluble salts (sulphates, chlorides, nitrates), eroding surfaces (pitting, perforations, fractures and/or missing fragments, cracks), carbonate and gypsum deposits, incompatible repair material, iron staining (on pillars), and mechanical damage.

3.2 Stone cleaning and repair – selection of materials and methods

Based on the deterioration patterns identified, a number of market available materials and methods of stone cleaning were selected for laboratory testing. For testing purposes white marble panels from the same quarry were provided. In deciding on methods, products and techniques considered for treatment were evaluated based on their likely effects on the stone over both a short and long-term period. The promising products (compatible with the mineral substrate based on their technical sheets) were first tested on the panel sample, Table 3. Colorimetric coordinates and surface roughness were tested. Based on the obtained results, the chosen materials and cleaning methods were tested in situ.

Table 3: Laboratory test panel - cleaning

	Laboratory test panel before the cleaning products application
	Laboratory test panel during test application of cleaners

In situ the test panel was established and zoned in 10 areas with characteristic deterioration patterns. The Figure 2 shows 8 test areas on the left with different deposits and dirt, while on the right both test areas present façade paint applied over graffiti layer. The cleaning products and techniques selected in laboratory were applied and relevant measurements taken.

Based on the evaluation of data obtained by microscopy, colorimetry, FTIR and XRF, the water/steam washing was recommended as the first stage of cleaning procedure for the whole façade surface, to be in the following stages combined with complementary techniques in case of stubborn deposits. Steam jet cleaning by spraying the surface of the stone with alkaline hot water/steam (pH 8; 148 ºC) under pressure (4.5 bars) to soften the surface deposits. High temperature was chosen to decrease amount of water and thus avoid saturation of the stone surface, deeper layers and foundation with damp penetration and forcing contaminants deeper into the stone. Based on the type and characteristics of each group of stubborn deposits, the following techniques were recommended: air abrasive cleaning based on fine rounded glass pearls, dry cleansing paste, and graffiti remover spray for safely graffiti clean.

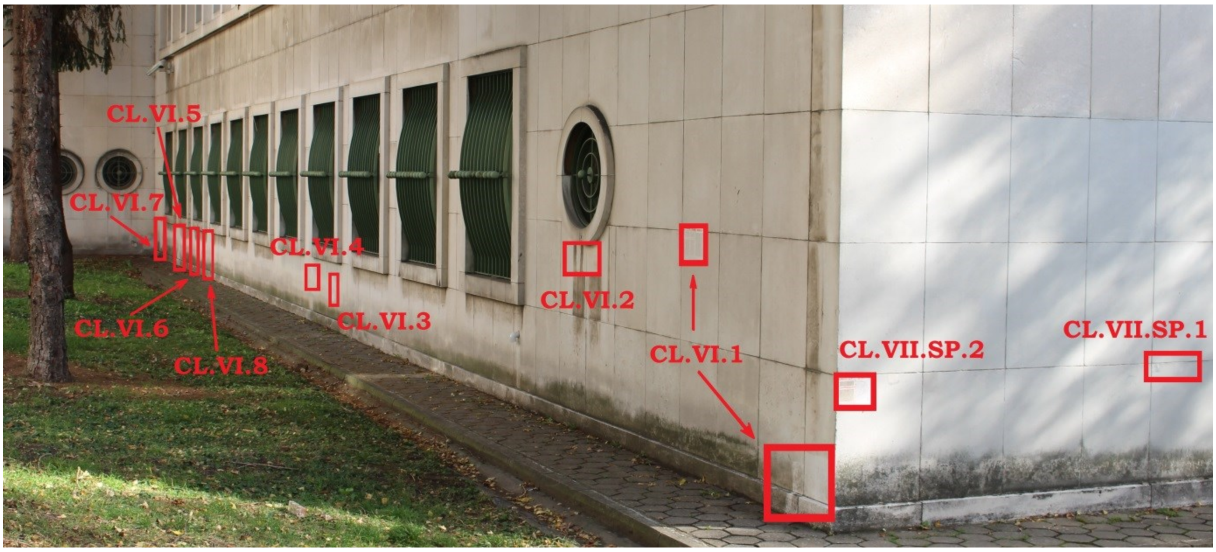


Figure 2: In situ test panel

The risk of damage was monitored during the study by continuous monitoring of surfaces and particular details under the microscope and spectrophotometer, before and after the cleaning trials, to assure that cleaning techniques and products do not jeopardize stone surface. Finally, on the areas where it was assessed not possible to remove all surface contaminants without notable risk of surface damage which would increase surface porosity and thus expose the stone surface to intensified weathering, the decision was taken not to seek a 100% clean stone, but rather to ensure that the majority of the contamination will be removed. Also, following the good practice and conservation principles the decision was made to preserve patina as genuine stone protection layer wherever possible.

After the cleaning, it was revealed which parts of the façade stone panels and stone pillars possess visible cracks and missing parts, as well as the extent of defects which require repair. The most compatible and effective repair materials were chosen in laboratory based on the results of spectrophotometry and drilling resistance tests (DRILL method two weeks after application). In the next step the selected ones were evaluated and confirmed in situ.

3.3 Stone protection – selection of materials and methods

Products for long-term stone protection have been tested under laboratory conditions. For in situ application and testing, products have been selected based on the results obtained in the laboratory, if they have met the criterion of compatibility with the substrate [11]. Effectiveness of the selected products in situ has been tested on previously cleared areas.

Protective agents of different functionality were selected to respond to the various degradation phenomena identified in the diagnostic phase of the study. The following functionalities were needed: anti-graffiti protection and self-cleaning. Anti-graffiti layer was required in lower zones of the building to allow future graffiti to be easily removed from the protected surface by steam or water. Self-cleaning to be applied on entire façade surface to extend the durability of the cleaning treatment effects by protecting surfaces from the deposition of environmental organic pollutants over the years.

In addition to examining the individual effects of anti-graffiti protection and the self-cleaning agents on the original stone, the interplay between them was also studied in real environmental conditions.

Various potential materials were tested on laboratory test panel to assess hydrophilicity/hydrophobicity on the stone substrate (contact angle measurement, Figure 3), graffiti removal and self-cleaning efficiency.

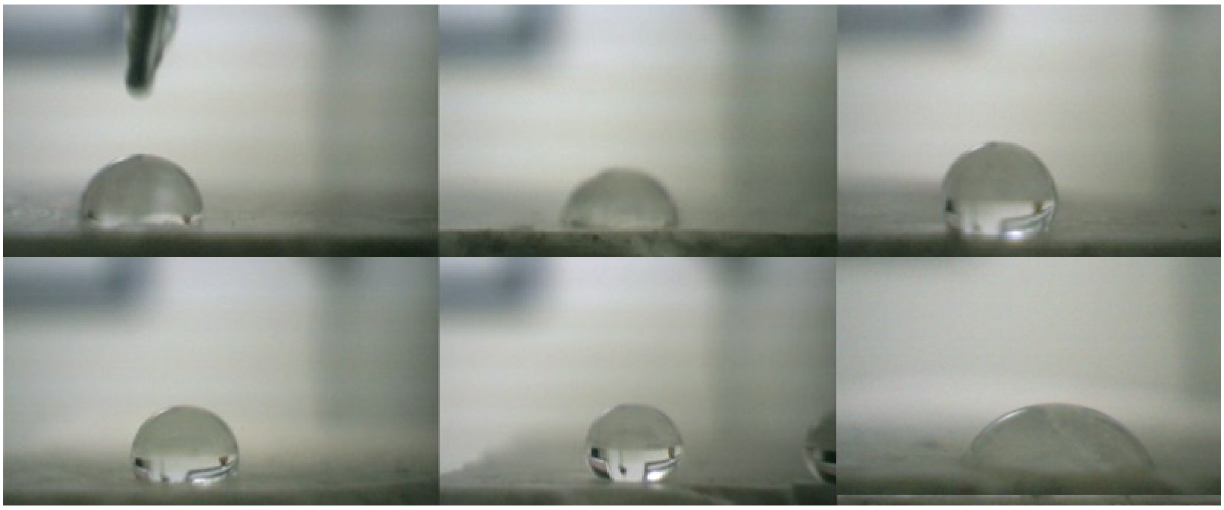


Figure 3: Contact angle measurements on the laboratory test panel

For the self-cleaning a photocatalytic material has been identified as potentially compatible with the substrate based on relevant scientific sources [12,13]. This material is a water suspension with photocatalytically active component encapsulated in hydrophilic nano particles. Due to the action of solar radiation it activates and breaks down atmospheric pollutants to inorganic (harmless) compounds. At the same time the surface becomes hydrophilic, which allows the degraded impurities to be washed away due to the effect of atmospheric precipitation and winds. Compatibility of this material with the stone substrate has been demonstrated by measuring colour parameters (spectrophotometric measurements) and contact angle values.

4 MONITORING

4.1 From laboratory to large scale stone cleaning and protection

The specific challenge for laboratory team was to scale up the research results from laboratory and in situ testing to large scale cleaning and protection of the Banovina Palace.

Figure 4: In situ control

From laboratory testing and establishment of conservation approach to inviting contractors to perform the stone façade cleaning and protection, some adjustments of the established methodology were required together with transfer of know-how. This process took into account the following important issues:

- steam / water washing –adjustment of parameters to large professional equipment and drain infrastructure available at the sight;
- what additional measures will be taken if in-depth stains persist;
- planned budget.

On the other hand, it was highly relevant to ensure that the contractor will fully comply with the work specification, even if that implies some changes of their standard practice. With certain refinements to comply with above-mentioned limits, the cleaning included: water/steam spraying with alkaline hot water/steam (pH 8; 120 ºC; 30 bars), dry cleansing paste, graffiti remover spray, and an addition of biodegradable and environmentally friendly dirt-solver as an alternative method for stubborn stains removal. For repair and protection established methodology remained unchanged.

4.2 In situ control

The involvement of the Laboratory on the sight in continual scientific monitoring of cleaning and protection works at Banovina Palace, from May to November 2019, Figure 5, was beneficial from multiple points of view. The most important, it proved that no significant visual damage (change of colour or surface roughness) was caused by the treatments. What is even more important, the regular presence of the Laboratory team at the building enabled solving unexpected issues promptly. Finally, regular monitoring and reporting after each cleaning stage with portable digital microscope and mobile spectrophotometer, together with regular XRF, FTIR and microbiological measurements, enabled some procedures to be repeated if necessary, and ensured that each phase (cleaning, repair and protection) is documented and approved when the satisfying results are achieved, Figure 5.

a) before b) after

Figure 5: Cleaning and protection - before (a) and after (b)

5 CONCLUSION

The main objective of the presented project was to establish methodology for stone façade cleaning, repair and protection at both laboratory and large scale. The key challenge was not only to scale up laboratory findings, but to make sure that the maximum amount of dirt deposits will be removed, while the stone surfaces will not be jeopardized and opened more vulnerable to future environmental exposure. The case presented certifies that the tasks put in from of the Laboratory team were delivered in optimal quality and in time, and that the challenges were answered competently and in the highest scientific-based manner.

The collaborative interdisciplinary capacity of the selected approach to conservation of 20^{th} architectural heritage emphasised the benefits of scientific support in all stages. It was evaluated as extremely effective by all relevant parties involved: the Provincial Government of Vojvodina which funded the works, the Institute for Protection of Cultural Monuments of the City of Novi Sad, the contractor company and the Laboratory. From this point forward it is established as a model to follow in cleaning and protection works on façades of the Banovina Residence, the other building of this complex, which are scheduled for summer/autumn 2020.

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