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# Morphology and detachment mechanism of weathering crusts of porous limestone in the urban environment of Budapest

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A fine-grained and a medium-grained oolitic limestone of Miocene age were studied on ashlars of monuments in Budapest. The studied buildings are located in a polluted urban environment. The surface alteration is characterized by the presence of white (thin and thick) and black (laminar and framboidal) weathering crusts. Flaking, scaling and blistering are common crust detachment forms. Crust detachment is followed by rapid surface loss in the form of granular disintegration or of secondary crusts stabilizing the stone surface. Non-destructive *in situ* mechanical tests such as Schmidt hammer rebound and Duroscope clearly document the presence of thin and thick weathering crusts, and the degradation of underlying fine- and medium-grained limestone. Crust formation is associated with a textural change, since precipitation of pore-occluding calcite and gypsum and reduction of porosity in the crust zone has been recorded. Crust detachment is attributed to the crystallization pressure of air pollution-related gypsum, to freeze/thaw cycles, and to differences in mechanical properties of crust and host rock.

Key words: porous limestone, monuments, air pollution, decay, weathering crust, gypsum

#### Introduction

Monuments located in a polluted urban environment show severe signs of soiling and decay. One of the most striking features visible in the cities is the blackening of façades. Air pollution has often been considered to be one of the dominant factors controlling the damage to limestone monuments. Although pollution levels have dropped in most European cities, traffic-related pollutants are still common. Diesel engines in particular can still produce large quantities of smoke, and additionally airborne pollutants, and thus gaseous emissions, can still

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be major contributing factors to urban air pollution (Bonazza et al. 2004). The rate of deterioration depends upon several factors including pollution fluxes, environmental setting (meteorological and micro-climatic conditions), and rock properties. When various stone types were compared it became clear that carbonates, such as limestone and marble, show especially intense soiling and blackening in an urban environment (e.g. Kieslinger 1949; Amoroso and Fassina 1983). The most common form of weathering on limestone is the development of gypsum crusts, as has been known from many cities (Amoroso and Fassina 1983; Antill and Viles 1999; Lefèvre and Ausset 2002; Fassina et al. 2002; Smith et al. 2002, 2003; Török 2002; Thornbush and Viles 2004; Török and Rozgonyi 2004; Smith and Viles 2006: Török et al. 2007). Crust formation is mostly attributed to the transformation of calcium carbonate into calcium sulfate. It has also been noted at an early stage that industrialization and urbanization had caused a significant increase of air pollutant concentration in the atmosphere. In recent years gypsum crust formation has been modeled under laboratory conditions (Rodriguez-Navarro and Sebastian 1996; Ausset et al. 1999; Primerano et al. 2000; Cultrone et al. 2004). Different limestone test blocks exposed to the same pollution regime have also been analyzed to understand the role of pollutants and limestone fabric in weathering ("scale problem", cf. Smith 1996). In addition it has been emphasized that surface properties play a key role in pollution entrapment and various types of crust formation (Amoroso and Fassina 1983; Zappia et al. 1998; Grossi et al. 2003). Nevertheless, most previous studies have largely focused on the description of processes and decay products of limestone; fewer studies are available analyzing the mineralogical and related physical changes that are triggered by pollution fluxes. The few examples describing the physical changes deal with limestone (Christaras 1991; Török 2002, 2003; Török et al. 2004), marble (Christaras 1996), granite (Irfan and Dearman 1978; Kahraman 2001) and rhyolite tuff (Topal and Sözmen 2003; Török et al. 2005). The present study classifies weathering features of porous limestone monuments based on their morphology, and also provides data on the mineralogical composition of these forms. A new insight into the problem is the combination of these results with physical changes caused by weathering. For this purpose various types of porous limestone were studied in the urban environment of Budapest. The paper summarizes these results, emphasizing the cause and effect relationships by comparing mineralogical analyses and strength parameters obtained by Schmidt hammer and Duroscope.

#### Methods

Ashlars of 19th and early 20th century's monuments were chosen and comparative analyzes were performed by non-destructive on-site strength tests and by laboratory analyzes. The studied buildings included the House of Parliament, the Citadella fortress, Mathias Church, the Central building of Budapest University of Technology and Economics, College of Fine Arts and Museum of Fine Arts (Fig. 1). The weathering features were described partly by using the nomenclature of Smith et al. (1992) and Fitzner et al. (1995) and partly by describing new weathering forms. At selected ashlars the strength properties of stone surfaces were tested by Schmidt hammer and Duroscope. On each tested block 10 measurements were made. These tests provide information on mechanical properties of rocks, giving a rebound value which correlates approximately with the strength properties of the rock. The water absorption of weathering crust and host rocks were detected by using a Karsten tube.



#### Fig. 1

The concentration of settling dust in Budapest with the location of studied buildings, insert map shows Hungary. 1. Central building of Budapest University of Technology and Economics; 2. the Citadella fortress; 3. Mathias Church; 4. House of Parliament; 5. College of Fine Arts; 6. Museum of Fine Arts

Small samples of weathering crusts (19 samples) and host rocks (4 samples) were taken form the buildings for laboratory analyses. In the laboratory, samples were analyzed by using X-ray Diffraction (XRD) and differential thermoanalysis (DTA) for the determination of mineralogical composition. The XRD analyzes were carried out using a Phillips Diffractometer (PW 1130 generator, PW 1050 goniometer, Cu anode and monochromator). The powdered samples (size fraction less than 63 microns) were analyzed at 40 kV, 20 mA. For the data collection and data evaluation a PCD-APD software package was used. Derivatograph analyzes (thermal analyzes) were carried out to determine the clay composition, gypsum and organic matter content of the samples. The samples analyzed by XRD were used in this test. The test apparatus was a MOM Derivatograph. 400–600 mg of powdered samples was heated at 10 °C/min, with the analyses carried out between 20–1000 °C. The thermic gravimetry sensitivity was 100–200 mg. Small samples were studied using binocular microscopy, and

thin sections were also prepared from samples to allow comparison of textural and mineralogical differences between the altered surface and host rock.

# Properties of limestone

The studied limestone ashlars were made of a soft and porous limestone of Miocene age. This shallow marine limestone has a yellowish-white colour when it is freshly quarried. According to XRD analyzes its main mineral is calcite (92–97%) but minor amounts of quartz and sand-sized lithic clasts are also found. Lithological analyzes have shown that several fabric types occur. The most common one is characterized by the presence of well to moderately rounded micro-oncoids of 0.2–1.0 mm in diameter (its commercial name is ooidal limestone of Sóskút). The ooids are surrounded by circumgranular calcite cement. Beside ooids, other textural elements such as gastropods, bivalves and foraminifera occur. Porosity is generally very high, and mainly related to intergranular pores, which are between 0.1 to 1 mm in diameter. Intragranular pores in the foraminifera or within the ooids also occur. The fabric of this limestone shows some variety in the size of ooids and in the amount of other particles, but is mainly classified as ooid grainstone.

Within the several textural varieties two predominant ones were found: a finegrained and medium-grained one. The first type, the fine grained oolitic limestone, contains very small ooids of 0.1–0.2 mm in diameter. Besides sparitic calcite minor micrite is also present (ooid packstone). The pore system is characterized by micropores. Most pores are intergranular and hence ensure a high effective porosity (37%) (Pápay and Török 2006). The second type is characterized by the presence of gastropods (of the genus *Cerithium*) and usually contains coarser ooids (nearly 1 mm), well to moderately rounded calcitic ooids and micro-



Micro-fabric of finegrained porous limestone (left) and medium-grained oolitic-microoncolitic limestone (right). The finegrained type is characterised by mostly wackestones and packstones while the medium-grained one is a typical oolitic grainstone

Fig. 2

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oncoids of 0.2–0.6 mm in size, as well as a few fragmented bioclasts of tens of millimeters size (ooid grainstone with few bioclasts) (Fig. 2). Typical pore sizes are smaller and are in the order of 0.01–0.02 mm (Török et al. 2004). The porosity is less than that of the fine-grained type. The most common sedimentary feature of this second limestone type is cross bedding, which is often visible on ashlars. The studied porous limestone types are similar in many aspects to some other porous limestone, such as the British Great Oolite (Monks Park Limestone; Bell 1993), various Jurassic limestone formations of Oxford (Viles 1993), the porous limestone of Cairo (Fitzner et al. 2002) or the French Jaumont Limestone (Ausset et al. 1999), but are very often much softer, lighter and especially the fine-grained types are more porous (Török 2003). The differences in fabric are also reflected in the physical properties. The quarries of the oolitic limestone still exist but almost all were operated during the second half of the 19th century, when most of the public buildings were built in Budapest. The only active quarry is found approx. 30 km to the west of Budapest in the village of Sóskút.

## Environment

Budapest is characterized by a continental climate. The annual mean temperature is 10.8 °C with 2000 hours of sunshine. Winter frosts are common, and the number of annual freeze-thaw cycles is 78 on average. Relative humidity shows diurnal and annual changes with lower values in the morning and increased values in the evening, and an annual maximum of nearly 90% during winter. The main gaseous pollutants are  $NO_x$  and  $SO_2$  but air quality has been improved with respect to  $SO_2$  in the past decade. Nevertheless the  $SO_2$  concentration is still double that of Paris or London (Török 2003). After a period of decline in the amount of settling dust a recent increase has been recorded. The main sources of  $SO_2$ ,  $NO_x$  and settling dust are attributed to urban traffic. The distribution of pollutants is uneven, since the city center, where street canyons and multi-story buildings are found, experiences nearly 50% of the pollution.

# Weathering features

Weathering crusts are by far the most common weathering forms. Crusts can be classified according to their colors and morphology. Dark-colored crusts are divided into laminar black crust, framboidal black crust and dust crusts. White weathering crusts are grouped into thin and thick crusts. Mechanical breakdown and crust removal are also very common.

# Dark-colored crusts

Three dark-colored crust types were identified on porous limestone façades; laminar black crust, framboidal black crusts and dust crust.

Laminar black crusts are found on vertical walls and surfaces that are sheltered from rain-wash (Figs 3 and 4). This type of crust is very common on other limestone types; it is also called "dark-colored crust tracing the surface" (Fitzner et al. 1995). Similar black crusts have been described from Venice (Amoroso and Fassina 1983; Maravelaki-Kalaitzaki and Biscontin 1999), from Oxford (Antil and



#### Fig. 3

Weathering features: a) laminar black crust (Mathias Church); b) framboidal black crust (Mathias Church); c) grey dust crust (Central building of Budapest Technical University); d) white crust (Citadella); e) severely damaged wall with scaling white crust and multiple flaking and blistering black crust (Citadella); f) rounding of edges due to granular disintegration (Citadella)

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Fig. 4 Variations in crust development. a) black crust on sheltered wall now partly shows scaling and flaking (Citadella); b) scaling white crust on rain and wind exposed wall (Citadella); c) surface blackening (arrows) on partly sheltered parts of a relief (Trinity Monument, Castle Hill)

Viles 1999; Thornbush and Viles 2004), from Paris (Lefèvre and Ausset 2002), and from other cities. Crust removal processes include blistering and multiple flaking (Fig. 4).

Framboidal black crust evolves on protected parts of walls, generally below cornices or ornaments (Fig. 3). Similar black crust morphology is also known as dendritic black crust (Camuffo 1995; Maravelaki-Kalaitzaki and Biscontin 1999) or as ropey ('bubble-shaped') crust (Antill and Viles 1999). Large surfaces are also covered by framboidal crusts, especially on sheltered ashlars and surfaces which are not exposed to direct rain wash. Framboidal black crusts are the thickest dark-colored decay features, with a maximum thickness of approximately 2 cm. Below the crust a thin cemented calcitic zone is observed (Fig. 5). The framboidal black crusts are relatively stable and crust detachment is less common than with laminar black crusts.



Fig. 5

Binocular microscopic image of oolitic limestone with black framboidal crust on the top. Note the irregular surface of the black crust and the thin calcite cemented white zone below

Grey dust forms an approximately millimeter-thick, or in some cases a centimeter-thick, unconsolidated layer on the stone surface that can be removed by sweeping (Fig. 3). It is found primarily on sheltered and dry stone surfaces in the

city center. The gray dust layer is very rich in organic carbon (8.1%) and in other minerals (59%; mostly quartz). The average gypsum content is also relatively high (28% – Török 2002).

The main mineral of black crust is gypsum, while calcite and accessory minerals such as clay and quartz provide a small contribution to the composition of the crust. The highest amount of gypsum was found in framboidal black crusts (Fig. 6).



Fig. 6

Average mineralogical composition of black framboidal weathering crust in percentage (others include quartz, feldspar and clay minerals)

# Light-colored crusts

The light-colored crusts are found mostly on exposed façades, where the stone surface is regularly washed by rainwater. Light-colored crusts are divided into two major types according to their thickness: thin and thick. The thick white crust has a thickness of a few millimeters up to 2 centimeters. Under the microscope one can see that the crust incorporates a large part of the stone substrate. Pores are occluded by micritic calcite within the crust zone. The surface of the crust is smooth and contour scaling is the most common crust removal form of thick white crust.

Thin white crust has a thickness of 1 mm or less. It is common on exposed vertical to subvertical ashlars of fine-grained oolitic limestone. Although the crust surface is relatively smooth it often shows some surface irregularities. It can have a pale grayish color, which is due to small organic carbon inclusions in the crust zone. Flaking and blistering are the most common detachment forms of such crusts (Török 2005). After crust removal the surface can be temporarily stabilized by secondary crust, or can step back by granular disintegration. Thus multiple crusts (primary to tertiary) might develop, which provide a protective surface and slow down the deterioration (Smith et al. 2003).

## Mechanical properties

Non-destructive on-site tests (Schmidt hammer and Duroscope) have shown that the surface strength of the black crusts is higher than that of the host limestone (Fig. 7). The difference between the two values is in the order of 30 to 45%.

Schmidt hammer rebound values of white crusts are larger than that of the host rock. On the other hand, decreased Schmidt hammer rebound values were recorded in zones where the crust was detached (Fig. 8). The detected Schmidt hammer rebound difference between the host rock and white crust is greater than between the host rock and black crust.







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Average Schmidt hammer rebound value of white crust and the limestone substrate below (host rock)



Average Duroscope rebound value of white crust and the limestone substrate below (host rock)

Duroscope rebound values are very similar to the Schmidt hammer values, since the host rock shows lower numbers than the white crusts. On average, double rebound values were very commonly measured on porous limestone ashlars (Fig. 9).

Thus the crust formation leads to an increase in surface strength within the crust zone, and a concurrent decrease in strength below the crust at the host rock (Figs 7 to 9). This is in good agreement with the fact that pores are mostly occluded in the crust, and thus a secondary cementation of the crust is observed.

#### Discussions

Porous limestone is very sensitive to surface degradation. It has been shown above that various decay features are observed on limestone monuments in Budapest, and that a single ashlar can display different types of soiling and decay. The most common weathering forms such as black crusts, flaking, scaling and granular disintegration are also observed on porous limestone in other cities (Smith and Viles 2006). However the extent of each feature may show significant variations depending on the substrate, pollution and meteorological factors. In Oxford catastrophic decay of limestone ashlars generally appear in the form of black crusts and blistering black crusts (Viles 1993). In other cities such as Cairo, porous limestone rarely exhibits black crusts, but scaling, flaking and followed granular disintegration are common (Fitzner et al. 2002).

In comparison with other cities white crusts are more common in Budapest. The predominance of white crusts in Budapest may be an indication of the significant solubility of Hungarian porous limestone under the continental climate conditions and urban atmosphere of Budapest. White crusts appear to be more cemented than black ones. Schmidt hammer and Duroscope rebound values of white crusts are greater than those of the black crusts. The rock surface below the crust has even lower Schmidt hammer and Duroscope rebound values than those of the crust. By comparing these numbers to the ones obtained from quarry stones (Török et al. 2004, 2007), it has been proved that Schmidt hammer rebound values decrease in the following order: white crust, quarry stone, and rock surface below the white crust. Thus white crust formation strengthens the porous limestone surface (Török 2003).

The crust detachment is partly controlled by crystallization pressure and in parts also related to various competences of cemented crust and host rock. Ice and gypsum crystals accumulate below the non-porous crust and thus exert extra pressure on it by lifting it. The combination of freeze-thaw cycles and salts (in Budapest mostly gypsum) can lead to catastrophic decay.

In cities with a milder climate, such as Oxford, air pollution and moisture are the primary causes of crust detachment (Antill and Viles 1999; Smith and Viles 2006), while in Budapest the role of freeze-thaw cycles is very obvious, since lowlying ashlars, which often become wet, show catastrophic decay more often than the ashlars that are found 2 m or more above the ground level (Török et al. 2004). The high porosity of limestone allows the spread of decay from one block to another within a limestone façade. A similar feature was observed on limestone walls in Oxford as well (Smith and Viles 2006).

Compared to sandstone, porous limestone shows very similar decay forms. Smith et al. (2002) have documented that black crusts can also develop on sandstone, but catastrophic decay occurs mostly on isolated ashlars without spreading from one to the other. The major difference between the decay mechanism of sandstone and porous limestone is in the dissolution and re-precipitation process. The crust on limestone can form an almost uniform seal with minor or no porosity (Török 2003; Török et al. 2007), while in the sandstone case hardening is also a sign of porosity loss, although the pores in sandstone are rarely occluded entirely.

It is difficult to compare the changes in surface strength (e.g. Schmidt hammer rebound values) of porous limestone and sandstone, since only sparse data is available for the latter. Based on the measurements on porous limestone of Budapest it can also be supposed that case-hardened sandstone has higher Schmidt hammer rebound values than its host rock. Further studies are needed to clarify small-scale mechanical changes that are associated with weathering crust formation. The micro-drilling resistance values provide additional data to understand these changes (Török et al. 2007).

#### Conclusions

Limestone decay in a polluted urban environment is a complex process which leads to changes in mineralogical composition and mechanical properties of the stone. There is a marked difference between the surface strength of quarry rock, weathering crusts and host rock. Air pollution and freeze-thaw cycles are the main trigger mechanisms of limestone decay in Budapest. Airborne sulfurdioxide and water vapor provide the source for gypsum, which is a reaction product of gaseous pollutants and the carbonate surface under moist conditions. Weathering leads to the formation of secondary gypsum in the near-surface zone of the limestone ashlars. Gypsum crystals mostly accumulate within the black crust or below the crust within the porous substrate. Gypsum contributes to the removal of crust by exerting crystallization pressure if it is formed below the crust. Concurrently, when gypsum precipitates in the pores of limestone it can also serve as a pore occluding cement and thus it participates in the formation of a non-porous surface crust. Schmidt hammer tests have shown that a qualitative description of the weathering state is possible. However, some rebound values for samples assumed to be less decayed show a large scattering, indicating that the stone may be more degraded in depth than one would expect.

Thick white crusts have the highest Schmidt hammer rebound values. Compared to the host rock the difference in rebound can be in the order of 100%.

Duroscope rebound values show the same trend, i.e. the strength of the thick white crust is significantly higher than that of the host rock. White crusts seem to be more common in the porous limestone of Budapest than in other cities, probably because of the properties of the host Miocene limestone. Further studies are required to better understand why porous limestone and sandstone show very similar weathering forms and can undergo a catastrophic decay under various urban environmental conditions.

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