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Causes and forms of decay of stuccos and concretes from the Roman city of Baelo Claudia (Southern Spain)

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Abstract

The stuccos and concretes from the Roman city of Baelo Claudia, a relatively small location based on the fishing industry (near present-day Tarifa, South Spain), have been studied. As a consequence of this study the nature and composition of those materials and the causes of their deterioration have been established. The main cause of the stucco and concrete decay is biodeterioration.

1 Introduction

The city of Baelo Claudia, situated in the southwest of the province of Cadiz, was built on the Bolonia cove at the end of the 2nd century BC and soon became a city with a certain power within the Roman Baetica. In the 1st century AD the city reached the peak of its development. By the end of the 2nd century AD, its economic decline had begun, probably as a result of the economic and social crises taking place in the Empire during that century. The abandon of the city and the earthquakes that it suffered initiated the long process of its decadence and deterioration.

In the city of Baelo Claudia the capitals and walls were stuccoed. The Roman architects and craftsmen used different artificial building materials in order to protect the stone from external aggression (marine environment, strong winds, etc.). These materials, called stuccos, had not only a decorative mission but also one of protecting the stone surface. Some stuccos were polychromed[1].

The aims of this work were to know the chemical and mineralogical composition of the different stuccos existing in the city of Baelo Claudia, the biological communities of algae and lichens colonizing them, and to determine the principal causes of their decay.

2 Materials and methods

Samples of stuccos and other artificial materials (concretes) were taken in different areas of the archaeological site of Baelo Claudia. A fragment of each sample was submitted to a process of complete grinding until reaching a particle size smaller than 88 μm. The following tests were carried out on ground samples: Mineralogical X-ray diffraction analysis (XRD), determination of the contents of water-soluble salts, and quantitative determination of anions and cations of soluble salts by ion chromatography. Different pieces of the same samples were used for thin sections (studied by optical microscopy), and study by SEM/EDX.

To characterize the biological communities on the stucco surface, the main colonizing organisms were identified. For algal and cyanobacterial identification, enrichment cultures were made in parafilm-sealed Petri dishes with BG_{11} or BBM solid medium. Lichens were identified in the field and thereafter confirmed in the laboratory.

3 Results

Mineralogical analysis by X-ray diffraction

Table I shows the results obtained in the analysis of some stuccos and concretes by XRD. This analysis leads us to the conclusion that the prevailing mineralogical component in all the samples is $CaCO_3$ in the form of calcite. The samples of stuccos 1, 3 and 4 contained (together with calcite) α-SiO_2 (quartz), although in a relatively small amount; traces of $CaMg(CO_3)_2$ (dolomite) were detected in samples 3 and 4. From these data it can be concluded that the binder of these stuccos is lime with aggregates most probably of a limestone nature (basically calcite and dolomite in a very small proportion).

In the samples of stuccos 6, 7 and 8 there was a greater amount of dolomite. As in the previously mentioned stuccos the binding material was lime, but the aggregates were calcite and dolomite.

The samples of concrete 17 and 18 had very similar mineralogical compositions. Together with calcite there was quartz, felspars, mica and clinochlore.

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Table I. Mineralogical composition of stuccos and concretes by XRD

S	Quartz	Calcite	Dolomite	Felsp	Clinochlore	Mica
1	+	+ + +	--	--	--	--
3	+	+ + +	o	--	--	--
4	+ +	+ + +	+	--	--	--
6	+ +	+ + +	+ +	--	--	--
7	+ +	+ + +	+ + +	o	o	--
8	o	+ + +	+ +	--	--	--
17	+ +	+ + +	o	+	+	+
18	+ +	+ + +	o	+ +	+	+

+ + + abundant, + + moderate, + little, o traces

Water Soluble Salts Contents. Quantification of Inorganic Ions by Ions Chromatography

Table II shows the contents of water soluble salts as well as the concentrations of inorganic anions and cations present in the salts. The stuccos and concretes had low contents of water soluble salts (in all cases less than 1 % wt). The anion present in greatest concentrations in all the samples was Cl^-, except for sample 8 in which was SO_4^{2-}. The predominant cation in all the samples was Ca^{2+} . The contents of Na^+, Mg^{2+} and K^+ were relatively small.

Petrographic analysis by optical microscopy (thin sections)

Petrographic analysis of the stuccos studied, performed by thin section optical microscopy using transmitted light, confirmed the results obtained by XRD. The aggregates were mainly limestones, above all calcite - although dolomite was also present. Dendritic quartz was found in much smaller amounts.

The limestone aggregates presented exfoliation planes, indicating that they come from crushing of calcareous rocks or marbles. It was also observed that some aggregate particles had undergone corrosion processes by their contact in the binder, giving rise to further recrystallization which could facilitate adhesion with the newly formed materials. The size of the aggregates varied, with a diameter ranging between 0.9 and 0.2 mm.

Most of the walls and capitals of Baelo Claudia have distinct layers of stucco covering the supporting stone (Figure 1). Thin section analysis of these stucco layers showed that their main difference is in aggregate/binder ratio and aggregate size. The innermost stuccos, that is those closest to the supporting stone, had coarser aggregates and higher aggregate/binder ratio (of the order of 3-4/1). The outermost stuccos had very fine aggregates and much higher proportions of binder, with aggregate/binder ratios of the order of 1/4.

SEM-EDX analysis

The microstructural analysis performed by SEM/EDX showed the practically total absence of salt deposits on the stucco surfaces. This confirms the low contents in water soluble salts found in these materials. Crystals of NaCl, KCl and/or $CaCl_2$ were found sporadically.

This microstructural study also revealed that these stuccos had a surface finish that has been lost in some areas. This finish was seen morphologically as a very smooth surface. Microanalysis of such a surface indicated that it is covered by a thin film whose main (if not exclusive) element is Si. It is not known if this surface finish is Roman in origin or has been applied later.

In some of the stuccos studied, both the limestone and silica aggregates of the stucco showed obvious signs of corrosion, as can be observed in Figure 2.

Biological colonization

The stuccos and mortars supported a considerable biological colonization. The sunny and dry surfaces of highly exposed stuccos had a mainly lichen covering composed of xerophilous and nitrophilous species, well adapted to resist dryness and high insolation. This community was dominated by lichens with crustose thallus: *Verrucaria viridula* (Schrader) Ach., *Caloplaca lactea* (Massal.) Zahlbr. and *Lecania turicensis* (Hepp.) Müll. Arg. In addition, when the surface was sloping or horizontal it was possible to find typically nitrophilic species with longer epilithic thallus: *Lecanora albescens* (Hoffm.) Branth. & Rostr., *Caloplaca flavescens* (Hudson) Laundon, *Caloplaca teicholyta* (Ach.) Steiner and even the foliose thalli of *Xanthoria parietina* (L.) Th. Fr. In these stuccos, algae and cyanobacteria were restricted to cryptoendolithic growth (inside the stone, in the existing pores) where a green layer visible to the naked-eye developed, composed mainly of cyanobacteria. In this community the cyanobacteria *Aphanocapsa* sp., *Plectonema boryanum* Gom., *Phormidium fragile* (Menegh.) Gom., *Phormidium autumnale* (Ag.) Gom., *Chroococcidiopsis* sp., *Synechococcus elongatus* (Näg.) Näg., *Nostoc* sp., and the algae *Muriella terrestris* Boye-Pet and *Chlorosarcinopsis* sp. were frequent.

In contrast, the surfaces of sheltered stuccos, shadowed and damp, were dominated by communities of algae and cyanobacteria, whilst lichens were restricted mainly to some water-running tracks

(*Collema* sp.) and niches protected from the rain (*Lepraria lesdainni* (Hue) Harris). Algae were relatively abundant, with *Choricystis minor* (Skuja) Fott, *Stichococcus bacillaris* Näg., *Muriella terrestris*, *Bracteacoccus* sp. and *Navicula mutica* Kütz., together with the cyanobacteria *Calothrix braunii* Bornet et Flah., *Gloeothece* sp., *Chroococcidiopsis* sp., *Tolypothrix byssoidea* Hassall. and several species of the genera *Phormidium* and *Nostoc*.

4 Discussion

The state of preservation of the different stuccos and concretes studied was quite variable. Some materials were in an advanced state of deterioration with loss of interior cohesion and lack of bonding to the supporting stone, as in the samples 1 (Forum), BC-6, BC-7 (Capitol temples). Others, on the other hand, showed high cohesion and good bonding, as in the sample 3 (Isis temple), 17 (*garum* factory), 18 (*macellum*). The water soluble salts contents in the materials studied was always lower than 1 % wt. (Table II). This is a rather low content and seems to indicate that there is no serious problem caused by salt crystallization. The stuccos and concretes are located in an environment affected by sea spray, but in spite of this the contents of Cl^- and Na^+ were quite low. The contents of the rest of the ions were also lower than 0.15 %, except those of Ca^{2+} which were about 0.4-0.6 %. This Ca^{2+} may come from a small partial dissolution of the carbonates which are a part of both binder and aggregates.

No efflorescence was found on the surface of the studied materials. This does not prevent us from ruling out the possibility of some salt crystallization on the material surface.

In some stuccos it was observed that there had been a loss of binding material, since the aggregates stood out and the binder was scarce. This phenomenon is explained by the fact that on the stucco surface a process of partial solubilization can be produced by rain water, which sweeps away not only soluble salts but some lime and binder. This also accounts for the low amount of water soluble salts. Moreover, the coast of Cadiz is a geographical area with strong east winds that facilitate the separation of poorly bound particles from stuccos and walls by physical and mechanical effects.

One of the phenomena observed in the archaeological site of Baelo Claudia is the loss of bonding of the stuccos with the supporting stone. The innermost part of the stucco in contact with the stone is separated and totally disintegrated. The stone (calcarenite in many cases, limestone in others) is a highly porous material able to absorb a large amount of water. This water moves within the stone, trying to escape by evaporating through the stuccoed coating.

The high water-accessible porosities in the studied stuccos shows that all of the latter have high permeability; nevertheless, this liquid water can become stagnant and thus favour the partial disintegration and solubilization of the calcareous stone, leading to the disintegration phenomena described. When this process of decay begins it can be intensified by the development and growth of microorganisms in the already altered area. These provoke and enhance the total loss of bonding since they inhabit holes, cracks and fissures which form in the separating layers of stucco, leading to the decrease of its cohesion and consistence. The excretion of organic acids and the production of CO_2 as result of cell respiration are important factors in the weathering processes, favouring carbonate dissolution. Also extensive pitting is originated, especially in the lichen/stucco interface, obvious to the naked-eye in the sun-exposed stuccos supporting high lichen colonization.

Cyanobacteria and algae also have an important role. Due to the high porosity of the material it is easy to penetrate and to develop cryptoendolithic communities, which contribute to the disintegration of the stucco, facilitating the appearance of organisms such as bryophyta or higher plants, with the consequent structural damage.

The study carried out has proved that the main causes of decay of the stuccos and concretes is the biological attack from the proliferation and colonization of different organisms in these materials. They have been found on the surface of the different materials and at different depths, as well as in the interfacial areas between the supporting stone and lime stucco. It has been proved that those samples in which none of these microorganisms thrived had a much better internal cohesion and bonding than the strongly attacked materials.

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Table II. Water soluble salts content in stuccos and concretes (% wt)

Sample	Cl^-	NO_3^-	SO_4^{2-}	$C_2O_4^{2-}$	Na^+	K^+	NH_4^+	Mg^{2+}	Ca^{2+}
BC-1	0.04	--	0.02	0.05	0.02	0.01	--	0.01	0.49
BC-3	0.06	--	0.01	--	0.02	0.01	0.01	--	0.35
BC-4	0.06	--	0.01	--	0.02	0.01	0.01	0.01	0.51
BC-6	0.13	0.12	0.02	--	0.01	--	--	0.01	0.54
BC-7	0.07	0.01	0.05	--	0.04	0.01	0.01	0.03	0.60
BC-17	0.07	0.01	0.12	0.06	0.02	0.01	--	0.01	0.47
BC-18	0.06	--	0.02	--	0.02	--	--	0.02	0.20

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Figure 1. Deteriorated stucco of the Forum of Baelo Claudia.

Figure 2. Calcite crystal from sample taken in the stucco shown in Figure 1. Etching, biological dissolution and pitting is evident. Bar represents 10 μm.