THE EFFECT OF URBAN POLLUTION ON CONSTRUCTION MATERIALS: STONE AND BRICK

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1. INTRODUCTION

The deteriorating effects of urban pollution are examined in this work, taking as an example the materials used in the construction of the Sacred Heart Church in Granada, Spain. The building is neogothic-neomudéjar in style and was built a century ago. Despite its relatively recent erection, the church is intensely deteriorated. It is located on the Gran Via, which has some of the heaviest traffic in the city, which, in conjunction with urban heating emissions, have caused intense blackening of all the buildings on this street. As with the church, the effects of the pollution are not only aesthetic, but are likely causing serious damage to the construction materials. Most of the buildings are modernist and comprise an interesting urban grouping representative of the architectural style at the end of the XIXth century and the beginning of the XXth in Granada. The study of the church is intended to be extrapolable to the other buildings in the area that are undoubtedly in similar circumstances.

2. CONSERVATION STATE

This study was carried out in conjunction with the preparations for conservation procedures, which were easily seen to be necessary from visual observation. The first aim, given the imminence of the conservation steps, was to determine the true state of conservation of the building materials. The entire façade of the church was covered by a dark layer of dirt. The surface of the bricks walls was clearly dusty but showed no other signs of weathering. The stone towers and the Neogothic ornaments carved in stone were much more alarming in appearance. The mortar joints between the ashlar were missing in many points, while higher plants and lichens were also growing in some zones. In addition, the stone showed various processes of weathering: black crusts, blistering, flaking, spallling, and even the loss of large blocks of stone, with evident danger for passers-by in this, one of the most central and busiest zones of the city.

3. MATERIALS AND METHODS

The church was built mainly of brick, with stone in the towers and upper ornamental areas having neogothic motifs. There are two types of brick: one with a rough surface, most likely shaped by hand, found in the less visible parts of the façade, while the other type of brick is smoother and was used in the exterior walls that face the street. The mortars joining the stone ashlar were also sampled, as well as other mortars used in the plastered interior of the towers. The black crusts and the powdery material inside the afore-mentioned blisters were also analysed.

X-ray diffraction (powder method) was used to determine the mineralogy of the samples. The samples were ground in an agate mortar to obtain a particle size of 50 μm and were then examined with a Philips PW 1710 Diffractometer with an automatic slit window (from the Department of Mineralogy and Petrology, Universidad de Granada). Thin sections were also prepared for study by petrographic microscopy. The samples were analysed by SEM, as well, using
a Zeiss DMS 950 microscope with a QX 2000 Microanalysis Link (from the Centro de Instrumentación Científica, Universidad de Granada).

Figure 1. Cross section of a black crust engulfing dark particles of pollutants: a) plane-polarized light; b) crossed polars.

4. RESULTS

XRD analysis has confirmed that the rocks are almost exclusively comprised of calcite, although in some samples there were traces of quartz and in the most weathered samples there were also small amounts of gypsum. In the quarry samples, however, gypsum is never found. Under
the petrographic microscope it can be seen that it is a biocalcarenite, principally formed of fossil remains of calcareous organisms (equinoderm, lamelibranchia, foraminifers, coralline algae, etc.). This rock is the so-called Santa Pudia stone which has been studied in many articles (Sebastian et al., 1991; Rodriguez-Navarro & Sebastian, 1991; Sebastián et al., 1992) and has been commonly used in many historic buildings in Granada (the Cathedral, to name one). In the most weathered areas of the rock, sheetlike or needlelike gypsum crystals can be seen growing in the pores and in the fissures, perpendicular to the fissure walls. This process causes great mechanical pressure, thus increasing the width and length of the fissures. This type of deterioration is common in many other historic buildings in urban areas.

XRD revealed that the black crusts are primarily formed of gypsum, which is also the powdery mineral found mixed with residue from the original rock inside the blisters. SEM and light microscopy were used to observe the crusts and the most weathered areas. The crusts are formed of intertwined sheetlike gypsum crystals engulfing subspherical particles of pollutants (Figures 1, 2 and 3).

Figure 2. Scanning electron micrograph of initial growth of needlelike gypsum crystals on the rock surface.
Figure 3. Scanning electron micrographs of particles of pollutants: a) with a morphology typical of oil combustion; b) with a morphology typical of coal combustion.

The joint mortars of the ashlers and the rendering mortars are 70% gypsum, 25% calcite and 5% quartz (with slight variations from sample to sample). Under the petrographic microscope their high porosity is very evident and is accentuated by processes of dissolution. The matrix comprises acicular gypsum crystals and includes unbaked fragments of the original gypsum. There are also nodules of micritic calcite.

Both types of bricks have a similar mineralogic composition: on average they contain 45% phyllosilicates, 25% quartz, 15-20% calcite, and 10-15% high-temperature phases (diopside and gehlenite). Some contain traces of gypsum. Paradoxically, analysis under the microscope revel as that the roughly finished bricks have finer-sized temper fragments and smaller amounts than the
other type. In both cases the matrix is quite birrefringent and scarcely vitreous. Relatively large micas grains are well preserved and there are also quartz crystals and schist fragments comprising the temper. Calcite is secondary or else shows signs of having been transformed by the baking, such as reaction rims or loss of texture.

The very few bricks that give the appearance of being weathered in general show a lower proportion of high-temperature phases under diffractometry. Under light microscopy, gypsum can be seen growing in the pores in the same way as occurs in the rock. Most of the bricks seem to be sound and to the naked eye reveal only a light coating of dust. Nevertheless, under the microscope it can be observed that just a few millimetres under the surface the bricks have gypsum growing in them, although in very small amounts. Interestingly, in some cases the gypsum is substituting the calcite grains; the calcite appears surrounded by a halo of gypsum, leaving a more or less unaltered nucleus (Figure 4).

![Figure 4. Carbonate grain with a rim transformed into gypsum, in a brick: a) plane-polarized light; b) crossed polars. (cc: calcite; gp: gypsum)](image-url)
5. DISCUSSION AND CONCLUSIONS

The materials used in the construction of the church were, broadly speaking, of quite good quality, which evidently makes it impossible to blame intrinsic factors to justify the high degree of weathering. The bricks were made with a primary material lacking problematic minerals, such as gypsum or pyrite, and were baked at a minimum temperature of 850°C (Maggetti, 1982), which guarantees acceptable durability. The Santa Pudia stone also tends to react well to weathering under normal conditions. However, the choice of a gypsum mortar for the joints was very unfortunate since it is easily soluble, thus causing two negative effects: on one hand, a loss of strength in the mortar itself and, on the other hand, the release of Ca²⁺ and SO₄²⁻ ions, which can crystallise in the empty spaces of other construction materials. Nevertheless, this cause is not enough to account for the tremendous deterioration in the rocks in some of the more affected zones.

The case of this church is likely similar to that of other buildings located in urban areas with high pollution. They may be relatively recent buildings, but they are as weathered as other, much older, buildings. This fact clearly indicates that, generally speaking, atmospheric pollution is the biggest problem of monuments in cities. Numerous works have demonstrated that the emission of SO₂ into the atmosphere, together with the catalyzing action of metal or carbon particles, give rise to the gypsum crusts that occur on many buildings and are the cause of the surface weathering of the materials in the buildings (Del Monte et al., 1981; Fassina & Stevan, 1992; Sabbioni, 1992; Zappia et al., 1994; Alonso et al., 1995; Sebastian Pardo & Rodríguez Navarro, 1996; Sabbioni, 1996).

This work has managed to identify one of the earliest stages in brick weathering, thereby providing more information about the mechanisms involved in this process. There has been much speculation on the origin of the gypsum in the black crusts. It is clear that the sulphate ion comes from urban atmospheric pollutants (Furlan & Girardet, 1992; Connor & Girardet, 1992; Sabbioni, 1996), but it is still unknown whether the Ca proceeds from pollutants as well or from the material itself. We have been able to photograph the initial stages of weathering and it is evident that the carbonate crystals forming the temper of the bricks are beginning to be substituted by gypsum, leaving a ringlike structure with a sulphated edge and an intact interior. This clearly demonstrates that at least in this case the calcium ion derives from the construction material itself.

The fact that gypsum crusts have been found in other non-carbonated materials (granites) does not exclude the possibility that, in some cases, the Ca may have its source in atmospheric pollutants. In any event, this study does establish the greater reactivity and sensitivity to SO₂ of the calcareous materials used in some monuments. We have noted that on the rocks thicker, more extensive crusts form with a greater potential for damage than on the bricks. In fact, in the bricks calcite crystals have been selectively attacked, leaving the silicate part intact.

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