Biodeterioration of marble in an underwater environment

Beatriz Cámara a,⁎, Mónica Álvarez de Buergo a, Manuel Bethencourt b, Tomás Fernández-Montblanc b, Mauro F. La Russa c, Michela Ricca c, Rafael Fort a

a Dept. of Geomaterials, Instituto de Geociencias (IGEO, CSIC-UCM), José Antonio Novais 12, 28040 Madrid, Spain
b Dept. of Material Science, Metallurgical Engineering and Inorganic Chemistry, Campus de Excelencia Internacional del Mar (CEI-MAR), Universidad de Cádiz, Avda. República Saharaï s/n, 11510 Puerto Real, Cádiz, Spain
c Dept. of Biology, Ecology and Earth Sciences (DiBEST), Università della Calabria, Via Pietro Bucci, 87036 Arcavacata di Rende, Cosenza, Italy

HIGHLIGHTS

• The biodeterioration of Macael and Carrara marbles was examined in a mid-term underwater test.
• Three conditions were used to simulate those of stones on the seabed.
• Calcareous deposits and microboring patterns were the main biodecay effects.
• Buried and water exposures derived into the least and the greatest biocolonization.
• Understanding biodeterioration helps to protect underwater cultural heritage.

GRAPHICAL ABSTRACT

This study examines the deterioration of geomaterials used throughout history that today may be found lying on the ocean floor. Submerged archaeological sites including cargoes from shipwrecks or ancient city ruins have been a topic of interest from a perspective of in situ musealization, as a way of making underwater cultural heritage accessible to the public. In an experimental study conducted at an underwater archaeological site in the Bay of Cádiz (SW Spain), we subjected two types of marble (Carrara and Macael) to three conditions to which submerged archaeological objects are often exposed: full exposure to the water column, natural processes of burial and unearthing, or permanent burial. After an 18-month study period, the factor found to mostly affect these materials was their biological colonization. This factor was assessed by estimating total surface biocover and the rate of surface biocolonization, and also through the identification of skeletons and associated alteration forms by light microscopy, and scanning electron microscopy (SEM). Biofouling and bioerosion were the main causes of biodeterioration and dependent on the position of the marble specimens in the seawater. The response of both materials was similar, though dolomite crystals in the Carrara marble acted as a protective barrier against actively penetrating microorganisms. These investigations have allowed the study of tracers left by epilithic encrusting organisms and endolithic bioeroders on marbles intentionally exposed to seawater, providing new insights to the understanding of the biodeterioration processes occurring in cultural heritage stones, with significant implications when they are part of underwater archaeological remains.

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1. Introduction

With changing environmental conditions, stone materials undergo transformation in a new scenario. This transformation, or decay process, starts at the surface in direct contact with the environment. Weathering of stone materials, besides causing surface aesthetic changes, will also affect their functional characteristics (bulk properties). Decay is mainly determined by the type of material and the prevailing conditions of the new environment (Warscheid and Braams, 2000; Doehe and Price, 2010). Materials deteriorate differently when exposed to subaerial or underwater conditions, since distinct environmental factors are involved in each case. In a marine environment, the decay of archaeological or engineering materials is still poorly understood, as several agents interact simultaneously and different types of deterioration processes take place (Pearson, 1987; Fernández-Montblanc et al., 2014a; Winton, 2015). Among all factors, we should highlight the important role played by hydrodynamic variables, hydrochemical features, sedimentary dynamics and biological communities. Several processes including physical (abrasion), chemical (corrosion) and biological occur as a consequence of interaction of the different factors with the stone. One of the main causes of the decay of stone materials in an underwater environment is biodeterioration (Cris ci et al., 2010; Davide et al., 2010; La Russa et al., 2013, 2015) in the form of biofouling and bioerosion phenomena. In the biofouling phenomenon, defined as the undesirable accumulation of microorganisms and organisms on surfaces of submerged materials bacteria, yeast and diatoms (microfouling), macroalgae, barnacles, tube worms, bryozoans, molluscs and mussels (macrofouling) are involved (Faj et al., 2011). The establishment of these fouling communities occurs in the sequential phases of biochemical conditioning of the surface, adhesion of bacteria, yeast and diatoms, unicellular eukaryotic colonization and pluricellular eukaryotic fouling (Wahl, 1989; Faj et al., 2011). The second type of biodeterioration, bioerosion, not immediately visible to the naked eye, is defined as the removal of stone material through the actions, mechanical and/or chemical, of endolithic microorganisms (cyanobacteria, algae, heterotrophic bacteria, fungi, foraminifers as microbioerosion) and organisms (clionid sponges, polychaetes and lithophagine bivalves as macrobioerosion) (Davide et al., 2010; Tribollet et al., 2011). Microbioeroders actively penetrate the rock interior (euendoliths) and leave their fingerprints via biochemical dissolution of the materials (Golubic et al., 1981; Davide et al., 2010; Tribollet et al., 2011; Ricci et al., 2013) involving mechanisms of calcium pumps, respiratory carbonic acid and/or enzymes (T ribollet et al., 2011 and references therein). However, the most damaging actions have been attributed to macrobioeroders, capable of excavating cavities and tunnels and causing irreversible loss of material in archaeological artefacts (Davide et al., 2010; La Russa et al., 2013).

Biodeterioration will differ depending on a variety of environmental conditions in seawater (chemical composition of the water column and its sediments, light regime, nutrient availability, waves and currents) (Bethencourt et al., 2014). Besides being conditioned by these factors, biodeterioration will also vary widely in relation to the intrinsic features of the materials (mineralogical composition and textural features e.g., porosity or crystal distribution) (Pearson, 1987; Aloise et al., 2013).

According to its good aesthetic and physical properties, white marble has been one of the most widely used materials throughout history for both structural (e.g., buildings) and decoration or artistic purposes (e.g., sculptures) (Gorgoni et al., 1998; Ricca et al., 2015). To preserve this extraordinarily rich cultural heritage, interest grows in trying to understand how marble decays in each environment, and especially in marine environments, which so far have been the least well characterized. This is because marble has been much exposed to this environment as a result of its use in both structural (e.g., buildings) and decorative or artistic purposes 

Their stone cargo lying on the seabed has provided valuable information on transport routes and embarkation points, on the marble trade, and on quarries and monument construction (Bel trame et al., 2012).

Within this context, it needs to be determined to what extent these underwater biodecay processes could affect the durability of stone materials, especially when these are underwater archaeological remains. According to the main principles of the UNESCO Convention on the Protection of the Underwater Cultural Heritage 2001, underwater cultural heritage is defined as all traces of human existence having a cultural, historical or archaeological character that have been partially or totally submerged, periodically or continuously, for at least 100 years (Maareveld et al., 2013). In this scenario, in situ prevention and protection measures were described as priority options for the conservation of underwater cultural heritage. These measures are considered to protect an archaeological site fully or in part in the marine environment. In addition, any protection or prevention measure taken should be monitored, as the protection conferred or deterioration rates underwater will vary in time with changes in the environmental marine conditions of the archaeological sites (Gregory et al., 2012; Fernández-Montblanc et al., 2014a; Winton, 2015).

The purpose of this study was to determine the deterioration of two types of marbles when exposed to seawater by estimating surface biocover (overall and of major groups of epilithic encrusting organisms) and rate of surface biocolonization. In addition, polarized light (PLM) and electron microscopy (SEM) were used to identify encrusting organisms by their calcareous skeletons and to investigate biodecay forms (epilithic and endolithic) derived from the interaction of biological components with the substrate in an attempt to understand biodeterioration processes occurring in the marine environment.

2. Study area

The archaeological site selected for our underwater exposure experiment was Bajo del Chapitel in the Bay of Cádiz (southwest coast of the Iberian Peninsula) (listed as Assets of Cultural Interest and recognized as Archaeological Zone in the General Catalogue of the Andalusian Historical Heritage). Currently, this archaeological site is being monitored in the framework of the ARQUEOMONITOR project. The archaeological remains of the S.M.I. (Sa Majesté Impériale/His Imperial Majesty, in this specific case, in honour of Napoleon) Bucintre and Roman/Punic battleships (Fig. 1A–B) (Bethencourt et al., 2014) were found scattered on a rocky-sandy seabed at a depth of 11 m. The S.M.I. Bucintre of the French-Spanish fleet was sunk in the Battle of Trafalgar (1805) during a violent storm that struck the coast of Cádiz.

The archaeological site Bucintre shows seasonal water temperature oscillations from 13 to 14 °C in February–March to 24 °C in September, small variations in salinity (36.0–36.2 psu, practical salinity unit), well oxygenated water (8 mg/L), slight seasonal and depth pH variations (~8.2) and redox potential fluctuations of the water column (50–200 mV). Sediments at the Bucintre site can be classified as moderately-sorted coarse quartz sand (D50 = 1.095 mm). The site also features a high current velocity of 0.35 m s~1 (Bethencourt et al., 2014) and wave heights ≤ 1 m for 50% of the year. During extreme storm events, offshore wave heights > 4 m from the WSW have been recorded (Fernández-Montblanc et al., 2016).

3. Experimental

3.1. Materials

The materials used for the experiment were Italian Carrara and Spanish Macael marbles. There are records of the uninterrupted use of both these marbles in cultural heritage from Roman times to the present (Malesani et al., 2003; Sáez-Pérez and Rodríguez-Gordillo, 2008). In addition, their presence has been reported in several underwater
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