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Influence of natural carbonation process in serpentinites used as construction and building materials

R. Navarro [a,*], D. Pereira [a], A. Gimeno [b], S. Del Barrio [b]

[a] Geology Department, Science Faculty, University of Salamanca, Plaza Merced s/n, Salamanca 37008, Spain
[b] Spanish Geological Survey, c/La Calera 1, Tres Cantos, Madrid 28760, Spain

HIGHLIGHTS

- Carbonation affects the properties of serpentinites when used as dimension stone.
- This process is observed from small to large scale in relatively close outcrops.
- It is recommended a full characterization before its use to avoid misuse problems.

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ABSTRACT

Serpentinites are rocks commonly used in construction. They have a very complex origin and multiple factors can influence their behaviour once placed in a building. These include the carbonation process that occurs naturally in these rocks. This process causes transformations of the serpentinite minerals (hydrated magnesium silicates) into carbonates, which affect their properties as building material. In the south of Spain, serpentinites have traditionally been used from two areas: Sierra Nevada (Granada), starting from the thirteenth century and the Sierra de los Filabres (Almeria). More recently, the latter are extracted together with the "White Macael" marble. They show variable carbonation state which is reflected in different properties even between very close outcrops. Their mineralogy, geochemical composition, and main technological characteristics were determined in accordance with the different UNE-EN Standards for use in the building industry. According to the results obtained, the process of carbonation in serpentinites affects their properties for construction and building materials. This process produces increased bulk density and decreased water accessible porosity, water absorption under atmospheric pressure, and water absorption coefficient by capillarity. Carbonation also leads to increased velocity of ultrasonic p-waves. The uniaxial compressive strength displays low values compared with the other samples. The values obtained for flexural strength are similar, or slightly lower, compared to the non-carbonated serpentinite. Knoop microhardness increases with carbonation. The results of our study can improve efficiency, productivity, and competitiveness in the use of serpentinites in the building industry.

1. Introduction

Serpentinites are rocks that have been used for construction and ornamentation throughout the history of civilization. Currently, this natural stone is used widely and sought mainly for its aesthetic features. Hundreds of varieties are exploited and marketed worldwide. These rocks have a very complex origin [1] and there are many aspects that can influence their use as dimension stone, such as the degree of serpentinization and/or carbonation [2–6]. Despite their widespread use, serpentinites are rocks that lack specific regulations in Europe [5] and their properties are determined with the same test as marbles. Due to the high degree of carbonation, many of the traded varieties are included in the technical datasheets of marble, although their mineralogical, geochemical, physical and mechanical properties do not correspond to marble. This can lead to the misuse of the material [7–9].

The carbonation processes of these rocks are being studied at present due to the great utility they have in the field of geological storage of CO_2, together with other related rocks such as peridotites [10–15]. This is due to the natural carbonation reactions that occur [14], either during serpentinization or hydrothermal alteration [16]. However, there is very little research into how this process can affect their properties when is used as dimension

* Corresponding author.
E-mail address: rafanavarro74@gmail.com (R. Navarro).

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stone. Previous works compared serpentinites formed with a high percentage of serpentine (mainly lizardite) from Cabo Ortegal in Galicia, Spain (variety Verde Pirineos) [5], with serpentinites that feature a high percentage of carbonates from Almería, Spain (variety Verde Macael) and India (variety Rajasthan Green). The authors observed that the most carbonated varieties, in which several generations of carbonates are distinguished, present greater problems of alteration. These are especially due to the presence of large numbers of calcite filled veins and fractures that sometimes lose filling material. Other authors [8] generically describe all the transformation processes that a rock can undergo to become a serpentinite, and how these transformations influence its behaviour as dimension stone, especially the carbonation processes. Other work [9] compared the main parameters (mineralogy, chemical composition, physical properties, etc.) of a series of commercial serpentinites from the area of Moeche (Galicia) (variety Verde Pirineos), with others from India (Rajasthan Green), according to the criteria established in the American standard ASTM C-97, which is the regulation that refers specifically to serpentinite. In previous works, two varieties of serpentinites from the "La Milagrosa" and "La Carrasca" quarries, located respectively in Lubrín and Albanchez (Almería, Southeastern Spain) were studied in detail [4]. The first was a serpentinite *s.s.* while the second presented a greater mineralogical variety, with higher carbonate content. The conclusion was that although a correlation between carbonation and physical and mechanical properties cannot be clearly established, their characteristics are very different from marble. They also indicate the high variability in the properties between serpentinites of different outcrops due to the complex origin of these rocks and stress the importance of an adequate characterization.

The serpentinites from Sierra Nevada have been used in historic building since the thirteenth century, in monuments as important as the Alhambra, the Cathedral in Granada, the Monastery of San Lorenzo de El Escorial, the Royal Palace and the Convent of the Salesas Reales in Madrid (Fig. 1a). Given their importance as dimension stone for historical use [17], they have been proposed as candidates for Global Heritage Stone Resource designation [18]. The serpentinites of the Sierra de los Filabres, have been included in the candidacy as Global Heritage Stone Province of the "Comarca del Mármol" [19] due to their recent use in some significant heritage buildings such as the cathedral of Nuestra Señora de La Almudena in Madrid (Fig. 1b) and many civil buildings across Spain [20]. The "Global Heritage Stone" initiative is an IUGS and UNESCO project that seeks the recognition of those dimension stones that have importance throughout history. Full details of this initiative are given at http://globalheritagestone.com/.

In the present work several serpentinites from quarries located in Sierra Nevada and Sierra de los Filabres (Southeastern Spain) have been fully characterized. Some of them show a high carbonation degree. Their mineralogy, geochemical composition, and main technological characteristics were determined in accordance with the different UNE-EN Standards for use in the building industry. The variation of their properties according to the different degree of carbonation was also studied.

The objective of this study was to highlight how the carbonation process can affect the main properties as dimension stone of several traded serpentinites used as dimension stone with the similar geological background.

2. Location and geological context

The quarries were the samples were taken are located in Güejar-Sierra (Granada), in the Sierra Nevada National Park (Fig. 2a) and in Macael area (Almería), in the Sierra de los Filabres (Southern Spain) (Fig. 2b).

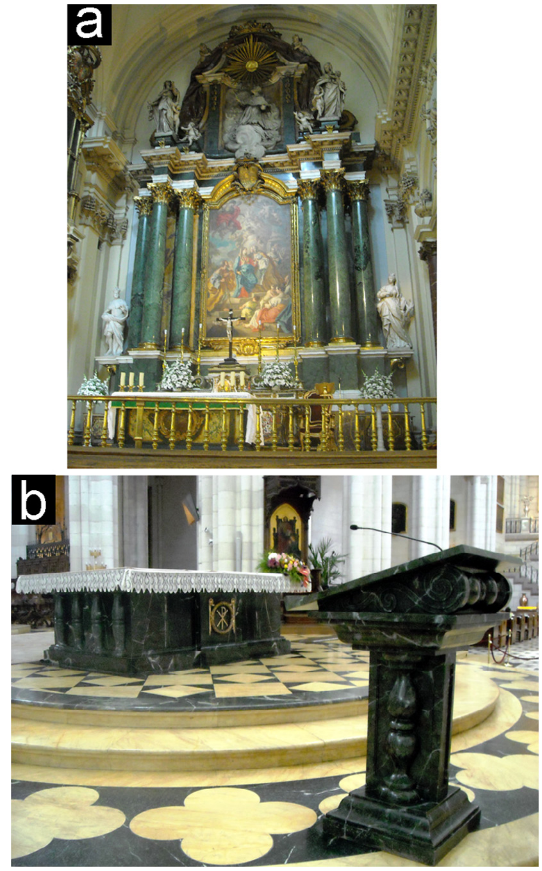

Fig. 1. Some examples of the use of serpentinites in heritage: a) main altar of the Convent of the Salesas Reales (Madrid); b) main altar of the cathedral of La Almudena (Madrid).

Geologically, they belong to the Nevado-Filábride Complex, which is the lowest metamorphic complex of the Internal Zones of the Betic Cordillera. This complex can be subdivided into upper and lower units [22]. The serpentinite quarries are located in the upper unit (Fig. 3). This unit is formed, from the bottom to the top, by metaconglomerates, micachists and quartzites of light colors, schist with garnets, and marble levels in the upper part. Throughout this sequence are interspersed metamorphosed igneous rock bodies. They are composed of stratoid levels of orthogneisses with tourmaline. The age of this unit is from Palaeozoic to Triassic [23,24].

A peculiarity of this unit is the intercalation of lenses with dimensions from metric to kilometric of basic and ultrabasic rocks, totally or partially transformed to eclogites, amphibolites and serpentinites. This is where the rocks here studied are located. It is possible to differentiate the following lithotypes [25–27]: a) *meta*-ultramafic rocks composed of serpentinites with diopside and Ti-clinohumite and metamorphosed ultramafic rocks. They come from metamorphized lherzolites and serpentinized secondary harzburgites; b) metadoleritic dikes that emerge as lenticular bodies of decimetric thickness partially rodingitizated and that have been transformed into metarodingites or eclogites during metamorphism [25]. They are composed of calcium-rich silicates and are embedded in the above serpentinites.

3. Materials and methods

All the samples to carry out the study come from quarries placed in Sierra Nevada and Sierra de los Filabres. Sierra Nevada