

Article

## Verde Macael: A Serpentinite Wrongly Referred to as a Marble

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**Abstract:** Serpentinites are used in both exterior and interior locations, but not all serpentinites are equal: they vary in physical-mechanical behavior and are not all suitable for similar uses. The serpentinites most commonly used worldwide come from India, Pakistan or Egypt. Spain has traditionally quarried two ultramafic massifs, one in Galicia (Verde Pirineos) and one in Andalucía (Verde Macael). Some of these quarries were small family-run businesses. In both cases, these rocks are commercially available as “green marble.” These serpentinites commonly have a high degree of carbonation, but the process does not always take place with the same intensity. Carbonate can act as a cementing agent of the other phases, increasing the mechanical strength parameters. As a result, an improvement in the strength conditions is achieved, but a misinterpretation of the suitability of the rock may occur because a perception among users that “green marble” is similar to geologically defined marble. This may lead to inappropriate applications as an ornamental stone. At a time of economic crisis in Europe, the natural stone sector is encouraged to invest in research to identify the best quality products that can compete profitably with those currently being imported from other countries. This paper provides a comparison of properties of the Verde Macael serpentinite with a true marble in the hope of contributing to improving the natural stone industrial sector.

**Keywords:** Serpentinite; Verde Macael; natural stone; carbonate replacement; mechanical behavior

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## 1. Introduction

Serpentinites are rocks made up primarily of minerals from the serpentine group (hydrated magnesium silicate): antigorite, lizardite and chrysotile. They can also contain other accessory minerals such as talc, chlorite, magnetite, carbonates and sulfides, as well as remnants of the precursor mineralogy such as pyroxene and olivine. They derive from the weathering of ultramafic rocks due to circulating fluids. These rocks have been widely used in construction, mainly for their aesthetic characteristics. They are found as structural (*i.e.*, columns) and decorative features (*i.e.*, plates, paving, cabochons, ornaments, *etc.*), both indoors and outdoors.

Serpentinites have a complex petrogenesis, with multiple geochemical and tectonic factors that influence their evolution and hence their subsequent behavior when used as building stone [1,2]. Depending on the degree of serpentinization and the main mineralogical transformation, serpentinites may show different textures that will be translated into different mechanical and physical properties, and therefore different performances upon emplacement [3] (and references therein). After serpentinization has taken place, other processes can affect the rocks, further changing their chemical and physical signatures. If the changes are related to the carbonation of serpentinites, the primary minerals are replaced by others with high volatile contents, dramatically transforming the behavior of the rocks when used in construction [4].

Outdoors, serpentinites are highly susceptible to weathering. The most frequent pathology in this case is a loss of material along frequent rupture discontinuities existing in the slabs. Other important pathologies are crusting, peeling of the surface, granular disintegration, flaking and cracking due to the effect of water flow, capillarity absorption, and dissolution processes that sometimes involve acid attack in polluted environments [5], as well as the replacement of the original mineralogy by carbonates [1].

Currently, the most commonly used serpentinites for construction come from India, Pakistan, Guatemala and Egypt, mainly because of the high volume of exploitable rock, but also because of the cheaper mining prices in those countries. Serpentinites are commercialized under the common name of “green marble” because, in many cases, they have undergone a full transformation into carbonates. Rajasthan Green and Emerald Green, from Pakistan and India, or even some varieties of Verde Macael, from Spain, have been completely transformed into carbonates [1]. As a consequence, there is no specific norm for serpentinites in Europe, and in the natural stone industry, the same norms are followed as for marble. However, their mineralogical, geochemical and physical properties do not correspond to marble, and, if carbonation has not completely affected the rock, an entirely different kind of behavior is to be expected when it is used as a construction material.

Serpentinite has been used throughout the history of civilizations to build decorative and structural elements. It is found in historical and modern buildings. Egypt has important examples in Islamic constructions [6], and in Greece and Rome, it is also possible to find impressive examples. Since

medieval times, many palaces, churches and cathedrals around Europe have used serpentinite as an important part of the whole construction (*i.e.*, Massimo Palace and Basilica di Santa Maria Maggiore in Rome, the cathedral in Florence [7,8], San Marcos in Venice, Saint Peter's Square and the church in the Vatican, the Saint Sophie cathedral in Istanbul, and Westminster Abbey [9]). Some cities in the United States of America have used serpentinites in both historical and modern buildings (e.g., Philadelphia) [5].

In the Iberian Peninsula, there are significant bodies of ultramafic rocks located in the south (Sierra de Ronda, Sierra Nevada and Sierra de los Filabres in Spain) and in the northwest (the Cabo Ortegal complex in Spain and the Bragança-Morais complex in Portugal). These bodies have different origins and show different textures and mineralogy since they have been affected by different serpentinitization processes and subsequent ones. Serpentinites from Cabo Ortegal were commercialized under the name of Verde Pirineos. Serpentinites from the south of Spain are still being quarried and, in general, they are known as Verde Macael, named after the most important village where the extraction takes place. Many problems have been documented related to the behavior of Verde Pirineos as a construction material, mainly in architectonic heritage [3]. In order to determine appropriate use of this rock, as well as of any rock type, it is necessary to know both its mineral composition and its physical and mechanical properties. For this, standard testing procedures are used. The ability to perform the same test at different laboratories following the same guidelines with similar equipment and following the same process is the key to achieving comparable results.

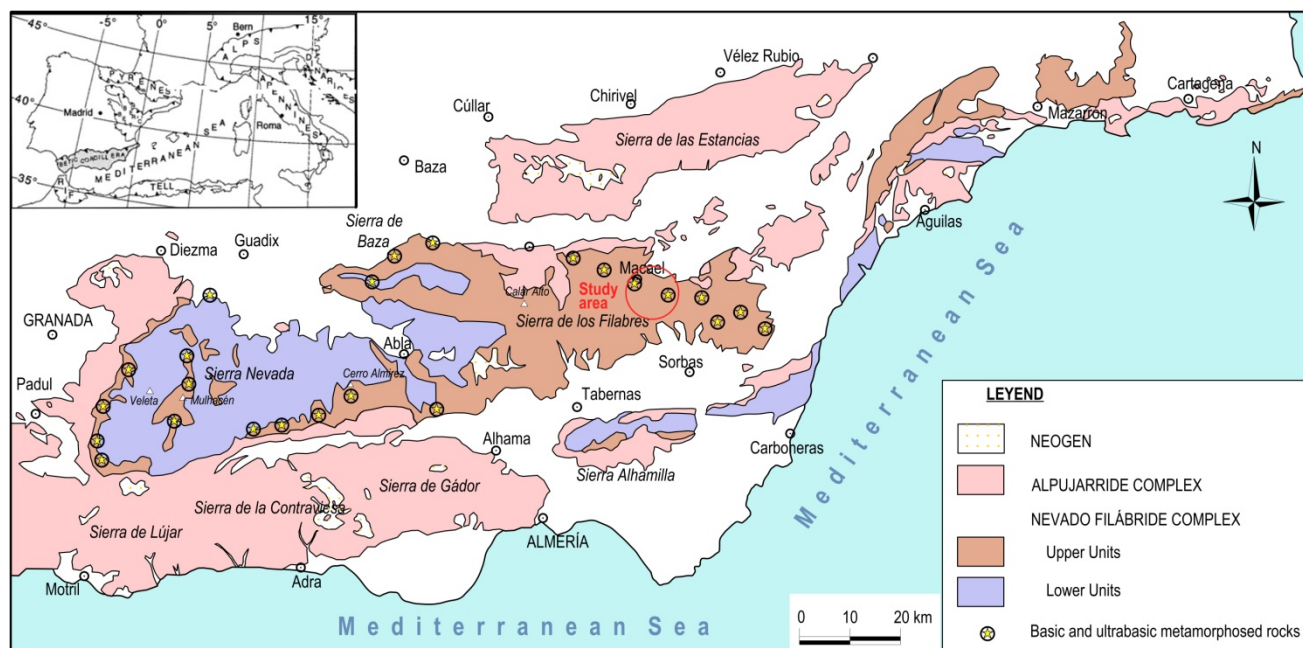
Here, we offer a comparative study of samples of serpentinite from Almería (Verde Macael), with different content of carbonates, and a sample of pure marble from the same region (Blanco Macael) in order to pinpoint the main differences that should be emphasized for their commercialization as a construction material. From the results, the suitability of the stone for specific construction uses can be assessed.

## 2. Location and Geological Setting of the Studied Samples

The serpentinites studied are part of the Nevado-Filábride Complex (NFC), which is the inferior complex of the Betic cordillera internal zone, belonging to the Alborán domain (Figure 1) [10]. One peculiarity of the NFC is the existence of basic or ultrabasic lenticular bodies of variable dimensions that may have been partially or totally transformed into eclogites, amphibolites or serpentinites [11] (and references therein) intercalated within marble and schist sequences.

The samples studied came from quarries located in Macael (Almería, south of Spain, Figure 2). Five samples were collected from La Milagrosa quarry (Verde Macael 1, Table 1) (Longitude: 2°01'55.2" W; Latitude: 37°12'57.6" N) and seven samples were collected from La Carrasca quarry (Verde Macael 2, Table 2) (Longitude: 2°07'51.6" W; Latitude: 37°15'21.6" N). Blanco Macael white marble was from the Cerro Arispe-Azules quarry (Longitude: 2°18'18.6" W; Latitude: 37°19'26.7" N).

At Macael, the marble and calc-schist sequence in the upper part of NFC is made up of white calcitic marble, serpentinites, pale mica-schists, grey marbles and banded marbles [12]. The serpentinites studied here are located in discontinuous and irregular, highly deformed bodies, intercalated with thick beds of pale mica-schists and small, metric layers made up of white marble, located in the main fracturing areas.

**Figure 1.** Geological setting of the Betic Massif. Modified from [12].**Figure 2.** Location of the quarries of our samples: 1, La Milagrosa quarry; 2, La Carrasca quarry; 3, Cerro Arispe-Azules quarry.

### 3. Methodology

Two blocks of serpentinite were collected from La Milagrosa and La Carrasca quarries. They were large enough to proceed with all the analytical assays. The specimens are grouped under the names of

Verde Macael 1 and Verde Macael 2, respectively. The sample from Blanco Macael is part of another investigation being carried out at the Spanish Geological Survey laboratories.

The mineralogical characterization of the samples studied was performed using X-ray diffraction and a Leica DM2500P (Leica Microsystems GmbH, Wetzlar, Germany) petrographic microscope, with a digital camera and the corresponding image treatment software. The equipment was a Panalytical X'pert-Pro diffractometer with a Cu X-ray tube ( $K\alpha$  of Cu,  $\lambda = 1.5405 \text{ \AA}$ ), graphite monochromator and automatic slit, voltage = 40 kV and intensity = 40 mA. To identify the crystalline phases, we used the software XPert High-Score (Version 2004) (PANalytical B.V., Almelo, The Netherlands) and data were managed with the PDF-2 database (International Centre for Diffraction Data, Newtown Square, PA, USA). The semiquantitative relations between minerals were determined according to the method of the reflecting power [13].

Physical and mechanical testing was accomplished following the European Standards (UNE normative) for natural stone. The testing was done at the Spanish Geological Survey laboratories, which are recognized by the National Quality Agency (Entidad Nacional de Acreditación, ENAC) as required by UNE-EN-ISO 17025:2005 [14] for laboratory testing.

Open porosity ( $P_0$ ) and apparent density ( $\rho$ ) were determined following norm UNE-EN 1936:2007 [15]. Specimens were dried in an oven ( $70^\circ \pm 5^\circ$ ) for a minimum of 48 h until a constant weight was obtained. Open porosity ( $P_0$ ) was analyzed using a vacuum chamber to reduce pressure to  $2.0 \pm 0.7 \text{ kPa}$ , extracting the air present within the open pores of the dried specimens. Then, the samples were placed in demineralized water at  $20^\circ \pm 5^\circ$ , documenting weight before ( $md$ ) and after the immersion ( $mh$ ). After drying the immersed specimens superficially, they were weighed again ( $ms$ ). Apparent density ( $\rho$ ) was calculated with the expression:

$$\rho_b = \frac{md}{ms - mh} \times 1000 \quad (1)$$

Open porosity ( $P_0$ ) is the ratio between the open pores volume and the apparent volume of the specimen:

$$P_0 = \frac{ms - md}{ms - mh} \times 100 \quad (2)$$

Water absorption under atmospheric pressure was determined following norm UNE-E 13755:2008 [16]. Testing was done on six cubic specimens ( $5 \text{ cm} \times 5 \text{ cm}$ ) after being dried in an oven at  $95^\circ$  for 72 h and immersion until constant weight was achieved.

Compressive strength was measured as established by norm UNE-EN1926:2007 [17]. Cubic specimens ( $5 \text{ cm} \times 5 \text{ cm}$ ) were dried in an oven at  $70^\circ \pm 5^\circ$  for a minimum of 48 h, until a constant weight was reached. Then, the samples were broken and the compressive strength was calculated with the equation:

$$R = \frac{P \times 1000}{A} \quad (3)$$

where  $R$  is compressive strength in MPa,  $P$  is the breaking load in kN, and  $A$  is the specimen area in  $\text{mm}^2$ .

Sound speed propagation was measured following the UNE-EN 14579:2005 norm [18], using a CNS-FARNEL PUNDIT PLUS digital ultrasonic meter. The specimens were placed between two

frequency transducers. One of the transducers emits a pulse which, after passing through the specimen, is detected at the other end and converted into an electrical signal, allowing the timing of the impulse propagation to be determined. Measurements were taken for each spatial direction (X, Y and Z). To calculate the textural anisotropy of the rock, we used the equation proposed by Guyader and Denis [19].

## 4. Results

### 4.1. Textures and Mineralogy

Table 1 shows that the main mineral phase in the studied serpentinites is antigorite, followed by magnetite and/or carbonates (*i.e.*, magnesite and calcite). Samples from La Carrasca quarry (Verde Macael 2) show a greater mineralogical heterogeneity than Verde Macael 1. Besides antigorite as the main mineral phase, Verde Macael 2 also contains a high amount of talc and carbonates in several samples, with some chlorite and pyroxene.

**Table 1.** Mineralogical composition of Macael serpentinites by X-ray diffraction.

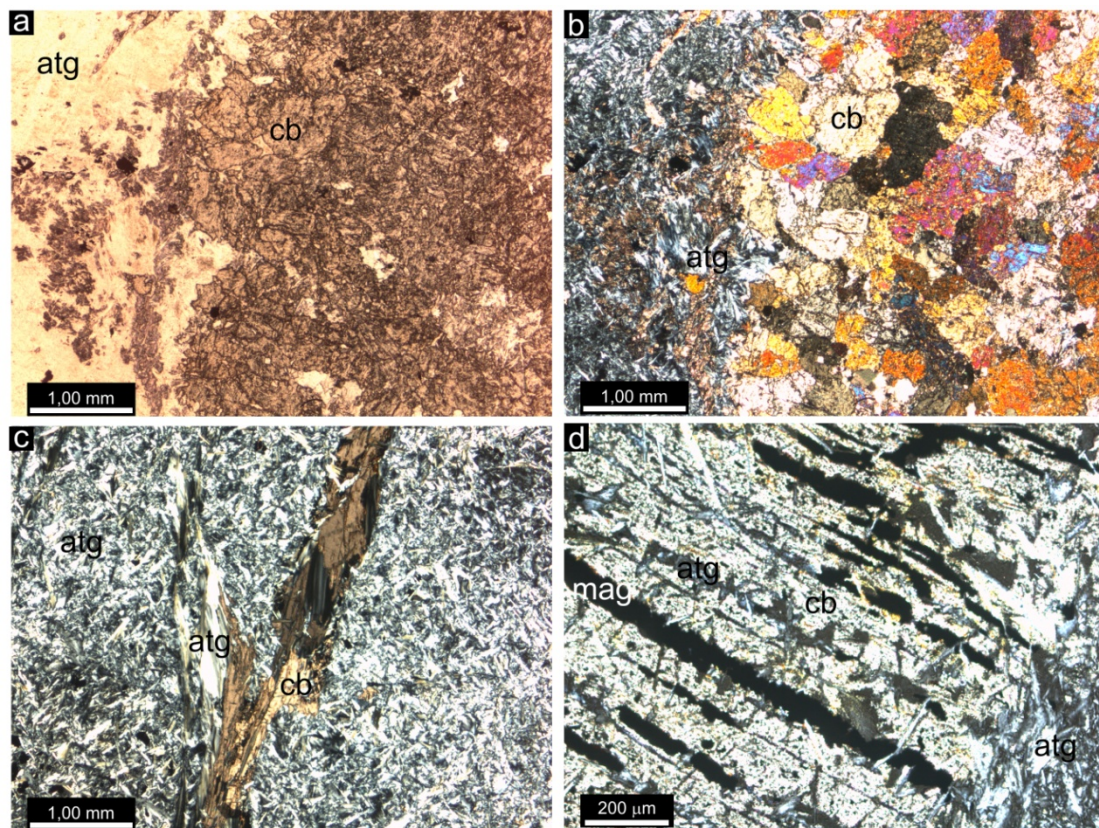
Sample	Main	Subordinate	Accessories and trace
Verde Macael 1			
MI-101	Antigorite (89%)	Magnetite (11%)	
MI-201	Antigorite (88%)	Magnetite (12%)	
MI-301	Antigorite (89%)	Magnetite (11%)	
MI-401	Antigorite (89%)	Magnetite (11%)	Calcite (<1%)
MI-501	Antigorite (89%)	Magnetite (11%)	Calcite (<1%)
Verde Macael 2			
CA-101	Talc (45%)	Calcite (25%), Dolomite (22%)	
CA-201	Calcite (65%)	Augite (35%)	
CA-202	Chlorite (70%)	Calcite (30%)	Talc (<1%)
CA-301	Antigorite (87%)	Magnetite (13%)	Magnesite (<1%)
CA-401	Antigorite (82%)	Magnetite (15%)	Magnesite (<1%)
CA-402	Antigorite (87%)	Magnetite (8.5%)	Magnesite (4.5%)
CA-501	Antigorite (100%)		Calcite (<1%)

Observations on the same samples were made under the microscope, detecting other minor phases that were not detected by X-ray diffraction (*i.e.*, pyroxenes in Verde Macael 1, although highly transformed into carbonate). Carbonates are found both replacing the original phases (Figure 3a,b) and filling shears and fractures (Figure 3c). Opaque minerals and acicular amphiboles are also found as accessory minerals. The most common texture is an interpenetrated pseudomorphic mesh [20], with fractures filled mainly by carbonates, but also by serpentine. The opaques tend to concentrate parallel to the fractures. Some pyroxene crystals have been completely transformed into pseudomorphic serpentine and carbonate, leaving some magnetite grains within the mineral cleavage (Figure 3d).

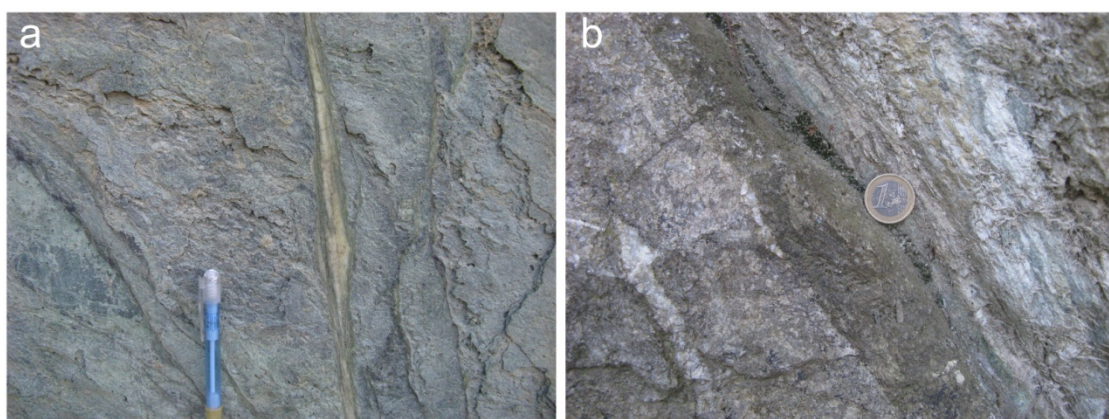
The microscopic observations support the field observations (Figure 4a,b), where centimeter-sized fractures filled by serpentine and carbonate, or only by carbonate, can be found.



**Figure 3.** Macael serpentinites under the microscope. (a) Verde Macael 2: antigorite replacement by carbonates; (b) the same as in (a) but under crossed nicols; (c) Verde Macael 1: interpenetrated texture with the replacement of antigorite by carbonates in a fracture. Crossed nicols; (d) Verde Macael 1: pyroxene being replaced by antigorite and carbonates, leaving magnetite in the cleavage. Crossed nicols.



**Figure 4.** (a) Veins of carbonate cutting the serpentinite; (b) *In situ* replacement of the serpentinite phases by carbonates (La Carrasca quarry).



#### 4.2. Physical and Mechanical Parameters

Table 2 shows the averages for the main testing done on the Macael serpentinites. A sample of Blanco Macael, a white marble quarried in the same area, has been introduced for comparison.

**Table 2.** Values of some of the most important parameters in the samples tested.

Sample	Bulk Specific Gravity (g/cm <sup>3</sup> )	Open Porosity (%)	Absorption (% weight)	Compressive Strength (MPa)
Verde Macael 1	2.68	0.7	0.30	252
Verde Macael 2	2.90	1.4	0.40	279
Blanco Macael	2.72	0.2	0.07–0.08 <sup>1</sup>	125

Note: <sup>1</sup> Source [21].

Verde Macael 1 shows lower density values than Verde Macael 2. This result can be explained in terms of the different mineralogy in the samples selected. The latter contain higher amount of carbonates (density for calcite = 2.71 g/cm<sup>3</sup>; density for dolomite = 2.85 g/cm<sup>3</sup>; density for magnesite = 3.00 g/cm<sup>3</sup>) and magnetite (density = 5.15 g/cm<sup>3</sup>). The former are made up mainly by antigorite (density = 2.54 g/cm<sup>3</sup>) and a little magnetite. Blanco Macael has a density corresponding to its mineralogy, almost pure calcite.

Both serpentinites show a higher value for open porosity than the marble. However, Verde Macael 2 has a higher value than Verde Macael 1. Weathering processes involving the transformation of the porous system (*i.e.*, the crystallization of salts and carbonates in shears and fractures) can lead to strong stresses on the rock walls [22]. This effect, together with other factors such as the orientation of the buildings, water content, frost-resistance capacity cycles, drastic changes in temperature [5], is translated into the initiation and propagation of fissures, leading to scaling and sanding that can end up with the complete destruction of the natural stone if no remediation measures are taken. Some of these processes can be observed at quarry faces where, depending on the orientation, the rock is more or less deteriorated (Figure 5).

**Figure 5.** Scaling in serpentinites at the La Carrasca quarry front. In this case, the exposure faces north, where marked changes in temperature and insulation take place.



A large difference is also observed in the water absorption values, being much lower for the marble than for the serpentinites. This is a very important result because water absorption is related to many rock pathologies observed in buildings. However, it should be noted that this parameter was obtained



from the literature. Tests on our own samples of Blanco Macael are yet to be performed to draw conclusive results concerning this comparison.

The compressive strength values in serpentinites are higher than in marble. Verde Macael 2 has a higher carbonate content than Verde Macael 1. However, the compressive strength values are higher than for Verde Macael 1. In this case, carbonates act as a cement between the other mineral phases, thereby increasing strength. Transformation takes place due to replacement of the original phases by carbonates, but also to the direct precipitation of carbonate in shears and fractures. As a result, the mechanical properties of the rock are improved directly [1].

#### 4.3. Sound Wave Speed Propagation

Sound wave speed propagation is a measurement of the anisotropy and compaction of the rock [23]. Lower velocities indicate greater fracture development. Table 3 shows the results for the serpentinites and the white marble studied.

The  $V_p$  values for Macael serpentinites are at the lower limit in comparison with other studies in bulk rock samples ( $V_p = 6.54\text{--}6.69$  km/s [24];  $V_p = 5.80\text{--}8.93$  km/s [25]) or for individual antigorite crystals ( $V_p = 5.6\text{--}8.9$  km/s [25]). The anisotropy coefficient ( $\Delta M\%$ ) for Verde Macael 1 is very low ( $<1.0\%$ ), while it is higher for Verde Macael 2 (samples with a higher amount of carbonates) and Blanco Macael. The latter can be explained by the strong anisotropy of the calcite “c” axis [26]. Anisotropy values fall with the degree of serpentinization of the rock, becoming higher if the serpentinites are mainly made up of antigorite instead of other serpentine polymorphs [27].

**Table 3.** Sound waves speed ( $V_p$  in m/s). Blanco Macael data are from [23].

Sample	$V_p$ (X)	$V_p$ (Y)	$V_p$ (Z)	$V_p$ (mean)	$\Delta M\%$
Verde Macael 1	5.72	5.73	5.83	5.76	0.98
Verde Macael 2	4.75	5.26	5.07	5.03	7.92
Blanco Macael	5.89	5.76	5.06	5.57	13.10

## 5. Discussion

Some of the most common varieties of serpentinites employed in ancient times are known as Porfido verde serpentine, Porfido verde di Grecia, Porfido verde antico or Verde antico [6]. Many of these rocks have traditionally been referred to as “marbles.” However, in very few cases do these rocks show a transformation high enough for them to be termed pure marble. Natural stone companies have long been using this term, leading to inappropriate selection of serpentinites for specific construction uses, both in historical buildings and new constructions.

Several authors have concluded that carbonated serpentinites (*i.e.*, Verde Macael and Rajasthan Green [3]) have better physical and mechanical properties than those that are not carbonated (*i.e.*, Verde Pirineos, [3]). Those studies also report the different problems that carbonated serpentinites could have once emplaced on buildings, mainly because of the large amounts of veins and fractures filled by carbonates. Dissolution can affect these carbonated fractures and the complete slab may disintegrate due to fracturing. At the same time, these carbonated rocks are more vulnerable to acid attack by contaminated urban atmospheres [5]. Ismael and Hassan [2] also described high carbonate

contents in Egyptian serpentinites with better mechanical behavior, although also subject to the same physical and chemical deterioration. These authors mentioned other factors influencing the behavior of the natural stone in construction, such as the content in different serpentine phases (lizardite or antigorite), the grain size of the rock, and the distribution and frequency of anisotropies (*i.e.*, fractures affecting the serpentinites). The present study comparing serpentinites from different quarries at Macael has allowed the comparison of samples with a greater or lesser degree of carbonation, replacing the original mineralogy against samples with a minor carbonation process affecting the rock except for carbonate precipitation in fractures. Physical and mechanical studies of the rocks have shown that serpentinites with higher contents of carbonates afford higher values for density, porosity, absorption and compressive strength that are significantly greater than those of the white marble used for comparison. Verde Macael 1, with a higher content of serpentine group minerals and a lower amount of carbonates than Verde Macael 2, has a lower anisotropy index. Discontinuities such as shears and fractures also affect the degree of anisotropy, but the higher amount of carbonates of Verde Macael 2 is translated into a higher textural anisotropy, similar to the values of Blanco Macael, the pure marble. This is probably due to the different content of antigorite, as well as to carbonation, because the replacement of serpentine by carbonates is not a homogeneous process and some parts of the rock may be affected to a greater extent than others.

## 6. Conclusions

Although we cannot yet establish a clear correlation between the presence of carbonate and the different physical and mechanical parameters, the differences observed between the serpentinites studied and serpentinites from other works (*i.e.*, Verde Pirineos, Rajasthan Green or the Egyptian serpentinites) highlight the importance of a detailed mineralogical characterization of the natural stone. The diverse origin of serpentinites, affected by different metamorphic and tectonic events, produces rocks with different mineralogy and different physical-mechanical properties that can serve different construction purposes, either in interiors or exteriors. This highly appropriate procedure should be emphasized in today's very deteriorated industrial sector, helping to increase its quality and economic profit. A common mistake with serpentinites is their inclusion within marbles. Each quarry has its own particular characteristics that should be properly assessed to ensure success, rather than failure, of the operation. The procedures used here should be adopted by today's significantly reduced industrial sector to better define quality, suitability for specific uses, and maximization of the resource to secure economic profitability. This will help to overcome the mistakes that are commonly made by users due to the assumption that "green marble" is the same as marble.

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