Title:

Determination of anisotropy to enhance the durability of natural stone. Proposal and validation of a new anisotropy index

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Abstract:

Anisotropy is a petrophysical property of natural stone and other construction materials that determines their quality and resistance to decay caused by a variety of agents, such as water. A study was conducted on nine types of stone widely used in Spain's built heritage, using six previously defined anisotropy indices. This paper proposes a new index (dMm) related to the two indices, dM and dm, that best reflect anisotropy in materials. These indices can be used to determine the degree of anisotropy, which helps explain the differential decay observed in stone materials quarried in the very same bed and used to build the same structure.

The conclusion reached is that anisotropy should be determined in the natural stone used both to restore the architectural heritage and in new construction, for the appropriate choice of material quality ensures greater resistance to decay and, therefore, increased durability. Materials with the lowest possible anisotropy should be selected, as this property governs their hydraulic behaviour: the lower the anisotropy in a material, the better its behaviour in relation to water and the longer its durability.

1- Introduction

All kinds of natural stone used in construction have certain properties that vary according to the direction in which they are measured: in other words, they are anisotropic. A variety of causes of anisotropy in rock can be identified, including crystal and grain orientation, the accumulation of minerals, stratification and lamination, stress orientation and the number and orientation of pores and cracks. Anisotropy, then, which depends on composition, texture and structure (Takemura *et al* 2003, Guéguen and Schubnel 2003), is directly related to the origin of rocks and any subsequent alteration.

Every type of rock (granite, limestone, slate, marble) has its own level of anisotropy. Ascertaining anisotropy in natural stone and how it develops is of utmost importance, for this information is essential to determining: a) how it should be quarried and most efficiently processed; b) the quality of the stone products and their suitability for use as ashlar or ornamental elements; and c) its placement on buildings. This is why the use of natural stone in construction is influenced by its anisotropy index. Likewise, the directions of anisotropy have a bearing on stone material decay and consequently on its quality and durability (Fort *et al* 2008, Fernandez-Revuelta *et al* 2008, Sheremeti-Kabasti and Snethlage 2000). These issues must be considered when planning restoration work on the architectural heritage, or when a certain type of natural stone is to be used for structural, aesthetic or insulation purposes in new construction, whether of buildings or civil works.

One of the most suitable techniques for determining anisotropy in materials is ultrasonic measurement. Ultrasonic waves propagate inside the stone at different velocities, depending on the direction. Ultrasonic techniques are used to study the physical and mechanical properties of natural stone and other construction materials, to characterize the materials (Zezza 1990, Fort *et al* 2002, Prikryl *et al* 2007, Del Rio *et al* 2006, Buj and Gisbert 2007, Sheremeti-Kabasti and Snethlage 2000, Fort *et al* 2007, García del Cura *et al* 2008, Fort *et al* 2010a); evaluate and quantify their decay (Sousa *et al* 2005, Weiss *et al* 2000, Yavuz *et al* 2006, Esbert 2007); assess the effectiveness of consolidants (Sebastian *et al* 1999, Myrin and Malaga 2006); determine the compatibility between original and replacement materials, especially when they are adjacent (Varas *et al* 2008); and establish the origin of materials used in historic buildings (Dreesen and Duser 2004, Gomez-Heras and Fort 2004). The effect of anisotropy on resistance to deformation and to mechanical failure in stone has also been widely studied (Prikryl *et al* 2007, Vasconcelos *et al* 2008).

Benson *et al* 2005 used the propagation velocity of ultrasonic waves to evaluate the effect of the pore system on anisotropy in different types of sandstone. The anisotropy of natural stone governs its hydraulic properties, especially properties with a strong directional component, such as capillary water absorption and permeability (Hammecker *et al* 1993, Zhang and Sanderson 1995, King 2002, Cueto *et al* 2009, Fort *et al* 2010b).

Anisotropy of wave propagation essentially means that wave velocity depends on direction; for geologic materials, such preferred directions are either a result of the way the stone was laid or determined by external conditions.

A number of analysis and testing methods and conditions are in place to determine rock anisotropy using ultrasound. The shape of the specimens may vary, for example: in addition to the more common cylindrical or parallelepiped specimens, polyhedra or spheres have also been used to measure the propagation velocity of ultrasound waves in the largest possible number of directions, although this renders the study of anisotropy in stone materials more complex.

This study aimed is to compare the anisotropy indices in several types of rock that are representative of natural stone as a whole to determine the advantages and drawbacks of each. The effect of the degree of anisotropy (anisotropic classes) on the hydraulic behaviour of the materials was also analyzed, and a new anisotropy index (dMm) is proposed and validated.

2- Methods

In this study, nine types of natural stone traditionally used in the architectural heritage of Madrid, Spain were selected from the three major groups of ornamental rock (granite, slate and marble). Four types of granite (Alpedrete, Zarzalejo, Colmenar Viejo and Cadalso de los Vidrios), three carbonate rocks (dolostone from Redueña and Torrelaguna and limestone from Colmenar de Oreja) and two types of slate (Bernardos and Sierra Negra) were chosen (Figure 1). With the exception of the Redueña and Torrelaguna facilities, which closed down in the mid-twentieth century, the quarries supplying this building stone are still operating, although some more intensely than others.

The petrographic analysis of the stone materials was performed on an Olympus BX51 polarized optical microscope fitted with an Olympus DP12 digital camera. The granite samples were stained with cobaltinitrite to distinguish the potassium feldspars from the plagioclases, and the carbonate rocks were partially stained with alizarin red to distinguish the calcitic from the dolomitic components.

The mineral phases were determined by XRD of the powder fraction from the entire sample on a Philips Analytical PW 1752 diffractometer operated at 40 kV and 30 mA, equipped with a copper K-alpha anode tube, graphite monochromator and PC-ADP Diffraction software. Measurements were taken in the 2 to 65° range at a step size of 0.02°, and 2°/min in continuous mode.

Several blocks of these materials were taken from each of the quarries, and cubic specimens measuring 50±5 mm on each side were cut from them. Specimen length was measured in the three orthogonal directions with a Mitutoyo digital calliper with a precision of ±0,01 mm. Four measurements were taken in each of the three directions, and averaged.

In each of these specimens, the ultrasound propagation velocity was measured in direct transmit/receive mode (transducers facing each other on opposite sides of the cubic specimen), and in the three spatial directions (xyz). Measurements were taken after the specimens were dried in an oven at 65 °C to a constant weight (when the difference between two consecutive measurements, taken 24 hours apart, was $\Delta \leq 1 \%$).

CNS Electronics PUNDIT test equipment, with a precision of ±0,1 µs, was used for the ultrasonic measurements. The system included transducers with a frequency of 1 MHz and a flat, round contact surface (11.82 mm in diameter). A water and carboxymethylcellulose paste (Sichozell Kleister brand, by Henkel) was used to improve and increase the contact and bond between the stone surface and the transducer.

Material anisotropy can be determined by measuring the propagation velocity of ultrasound or P waves (Vp) in different directions in space. In the present study, the anisotropy indices were found from the ultrasound propagation velocities measured in the materials under analysis. The indices proposed by Birch 1961, Guydader and Denis 1986, Schön 1996, and Ruedrich and Siegesmund 2007 were used, as shown below.

$$k = ((V_{pmax} - V_{pmin}) / V_{pmean}) \cdot 100 \quad [1] \text{ Birch 1961}$$

$$A_v \text{ (anisotropy coefficient) or AS} = ((V_{pmax} - V_{pmin}) / V_{pmin}) \cdot 100 \quad [2] \text{ Schon 1996}$$

$$ARS = ((V_{pmax} - V_{pmin}) / V_{pmax}) \cdot 100 \quad [3] \text{ Ruedrich and Siegesmund 2007}$$

$$AD = V_{pmin} / V_{pmax} \quad [4] \text{ Schon 1996, Benavente et al 2007}$$

$$dM\% = [1 - (2V_{pmin} / (V_{pmean} + V_{max}))] \cdot 100 \quad [5] \text{ Guydader and Denis 1986}$$

$$dm \% = [(2 \times (V_{pmax} - V_{pmean}) / (V_{pmean} + V_{max}))] \cdot 100 \quad [6] \text{ Guydader and Denis 1986}$$

The three velocities shown in the anisotropy calculations refer to the velocities found for the three orthogonal directions in space: V_{pmax} is the mean maximum velocity obtained in any one of those directions, V_{pmin} is the mean minimum velocity and V_{pmean} is the intermediate value. All authors include V_{pmax} and V_{pmin} in their anisotropy calculations, i.e., two of the three axes in space, while others include V_{pmean} to take all three axes, rather than just two, into consideration.

This latter was the criterion applied by Guyader and Denis 1986 in their definition of anisotropy: in total anisotropy (dM%), they took the three spatial axes into consideration, while in relative anisotropy (dm%) they used only two, excluding the axis for which the smallest V_p value was found. These two indices furnish complementary information, as each indicates the direction in space in which the structural, textural or mineralogical orientation that determines anisotropy in a material is concentrated. Therefore, these indices (dM and dm) provide more information than the others listed above.

The data were then statistically processed using cluster analysis (k-means clustering as per Khan and Ahmad 2004) to establish groups or classes of anisotropy for each type of material. The number of classes was established such that each group comprised a minimum of eight specimens, to ensure that the values of the stone hydraulic and durability parameters determined were representative (Fort *et al* 2008, Fernandez-Revuelta *et al* 2008, López-Arce *et al* 2010).

Nearly 800 (787) specimens were analyzed, and over 300 (329) selected for hydraulic characterization testing in accordance with the Spanish and European (UNE-EN) standards in effect for natural stone (AENOR 1999, 2002): Bulk density and effective porosity (UNE-EN 1936:1999), Water absorption at atmospheric pressure (UNE-EN 13755:2002) and Capillary absorption of water (UNE-EN 1925:1999). These were regarded to be the most suitable tests for determining the physical and hydraulic properties of the stone materials.

Two specimens of each type of stone were selected for the capillary water uptake test (UNE EN 1925:1999), according to the following criteria: the specimens had different degrees of anisotropy, so that one would show simultaneously high, and the other simultaneously low, values in both anisotropy indices. The test was performed in the directions defined by the maximum and minimum ultrasound propagation velocity, which would be parallel and perpendicular to the discontinuity planes causing the anisotropy ($V_p \parallel$ and $V_p \perp$).

3- Materials: petrological and mineralogical characteristics

The quarries from which the materials analyzed were extracted are located in the region of Madrid, except for the Bernardos slate quarry in the province of Segovia, located 120 km north-northwest of Spain's capital (Figure 1).

Granitic rocks

While the widespread use of granite for building in Madrid dates back to the twelfth century, it underwent a boom beginning in the sixteenth century, when the Royal Monastery of El Escorial was built and the capital of the Kingdom of Spain was moved to Madrid in 1561. This type of stone was used for many of the city's most emblematic buildings and monuments (such as the Royal Palace), and even for recent construction (the Nuevos Ministerios government complex and the Bank of Spain enlargement, to name two). The Sierra de Guadarrama Mountains are dotted with quarries, encompassing batholiths of peraluminous granites ranging from monzogranites to leucogranites (González Casado *et al* 1996, Villaseca *et al* 1998).

Alpedrete granite: This granitoid is classified as a medium-grained subidiomorphic, equigranular monzogranite containing quartz (crystal size 2-3 mm and 55-60 % vol.), plagioclase (1-3 mm and 20-23 % vol.), potassium feldspar (microcline; 2-4 mm and 7-10 % vol.) and biotite (1-2 mm and 10-15 % vol.). Its accessory minerals are cordierite, apatite, zircon and monazite. The plagioclase

is idiomorphic and zonal and exhibits albite twinning, while the potassium feldspar is subidiomorphic-allotriomorphic and has a poikilotopic texture and perthitic exsolutions. The quartz is globular and markedly heterometric, with fluid inclusions (Figure 2a)

Zarzalejo granite: Subidiomorphic, equigranular monzogranite, coarse-grained and grey in colour. It contains quartz (2-5 mm y 60 % vol.), plagioclase (3-4 mm and 15-18 % vol.), potassium feldspar (4-6 mm and 7-10 % vol.) and biotite (2-3 mm y 15 % vol.). The most significant accessory minerals are apatite, zircon, opaque minerals and monazite. The plagioclase has heterometric crystals, complex zoning, a low level of seritization and a tendency to form polycrystalline aggregates. The potassium feldspar is large in size, with a poikilotopic texture and an abundance of perthitic exsolutions. The quartz varies in shape from subidiomorphic to allotriomorphic, and fluid microinclusions are present. The biotite is very abundant and is slightly chloritized, forming polycrystalline aggregates.

This stone is currently marketed under the name Blanco Rafaela (Rafaela White) or Gris Escorial (Escorial Grey) (Figure 2b).

Cadalso de los Vidrios granite: This is a medium-grained, equigranular leucogranite with a uniform composition, comprising quartz (2-4 mm and 50-55 % vol.), plagioclase (1-3 mm and 20-25 % vol.), potassium feldspar (2-4 mm and 20-25 % vol.), and in a smaller proportion, biotite (1 mm and 5 % vol.). Accessory minerals are zircon, apatite, monazite, sphene, ilmenite, allanite and some opaque minerals. The biotite, present in very small quantities, is somewhat chloritized, forming polycrystalline aggregates. The plagioclase ranges from idiomorphic to subidiomorphic and is slightly seritized and zonal, with polysynthetic twinning (albite). The potassium feldspar is subidiomorphic and albitized, with poikilotopic textures and Carlsbad twinning. The quartz is subidiomorphic with fluid inclusions. This granite is sold under the name Blanco Cristal (Crystal White) (Figure 2c).

Colmenar Viejo granite: This is a medium-coarse-grained, heterogranular, leucocratic monzogranite with potassium feldspar (5-6 mm and 35-30 % vol.), quartz (2-4 mm and 30-40 % vol.), plagioclase (2-5 mm and 20-28 % vol.), and to a lesser degree, biotite (1-2 mm and 7-10 % vol.) in its composition. Present as accessory minerals are cordierite, muscovite, apatite, zircon and opaque minerals. The cordierite is altered to sericite. The potassium feldspars, large in size, vary from idiomorphic to subidiomorphic and appear as porphyries, with Carlsbad twinning, a poikilotopic texture and perthitic exsolutions. The subidiomorphic quartz forms polycrystalline aggregates (Figure 2d).

Carbonate rocks

The oldest carbonate rocks used in Madrid are known as Redueña and Torrelaguna stones, from the Upper Cretaceous period. These type of stones were used extensively from the fourteenth to the eighteenth centuries, and were quarried until the mid-twentieth century. Another stone that has been widely quarried since the eighteenth century is Colmenar stone or limestone, used in most of the monuments in the central part of the Iberian Peninsula (Dapena *et al* 1989, Fort *et al* 1996).

Redueña stone: This is a dolostone with a massive internal structure formed by rhombic dolomite crystals. Its textural variations are reflected in the domain of two crystal sizes; compacted areas with dark, dirty and altered (opaque cores) microcrystals (15-20 μm) alternate with less compacted areas with clear, clean and zonal mesocrystals (60-150 μm). This latter area is characterized by incipient de-dolomitization (transformation of dolomite to calcite) and high porosity (20-40 %). The pores exhibit fairly regular morphology and range in size from 100-200 μm to 7 mm (Fort *et al* 2008) (Figure 2e).

Torrelaguna stone: This is a massive dolostone formed by rhombic dolomite crystals. It is occasionally banded because remnants of its original depositional texture (stromatolitic structures) are preserved. The crystals are microcrystalline, equigranular (<50 μm) and dark, with few mottled cores. Porosity is very high (>40%), and poikilotopic and blocky mosaic cements predominate. Incipient de-dolomitization can be observed. The presence of localized iron oxyhydroxide deposits gives this stone its reddish colour (Fort *et al* 2008) (Figure 2f).

Colmenar stone: This is an Upper Miocene limestone from the Madrid sedimentary basin. It is classified as a lacustrine biomicrite/biosparite formed by a bioclast skeleton (40 % characeae, ostracods and gastropods) and a paste where the micritic matrix (20-30 %) alternates with sparitic cement (30-40 %). The micrite is a cryptocrystalline mass, calcitic in composition and dark-coloured. The sparitic cement is a drusy mosaic material. The existence of geopetal fill areas (matrix/cement) is indicative of significant phreatic-vadose conditions in the sedimentary medium (Figure 2g).

Metamorphic rocks:

Bernardos slates: These slates are extracted from quarries located in the Schist-Greywacke Complex dating from the Precambrian/Lower Cambrian period in the Central Iberian Zone of the Iberian Peninsula (Alonso *et al* 2005).

In construction, they have been mainly and widely used as roofing shingles. They can be found on many emblematic buildings in Madrid dating from the sixteenth century onward (Fernández-Revuelta *et al* 2008), as well as elsewhere on and beyond the Iberian Peninsula.

These slates have a visibly uniform texture, although polarized optical microscopy reveals small differences in their particle size distribution, related to their mineral composition (quartz 70-55 μm and phyllosilicates <65 μm). The quartz and a small amount of plagioclase (albite) appear both in aggregates and forming fine bands 0.5 to 1 mm thick, alongside the phyllosilicates, the most predominant group of minerals: biotite, muscovite, chlorite and clinochlore. Apatite, tourmaline, zircon and rutile are accessory minerals (Figure 2h).

Sierra Negra slates: These slates were extracted from small quarries in the Sierra de Ayllón Mountains, in the easternmost part of the Sistema Central mountain range. They are Silurian in age, folded during the Variscan orogeny and raised during the Alpine orogeny. The use of these materials as roofing shingles and blocks gave rise to a traditional type of architecture in the region, known as "black architecture" or the architecture of the "black villages". This slate is clayey in texture, with a fairly heterogeneous composition and slight foliation. Under a polarized light optical microscope, it is very dark, with irregular, wavy banding. Its mineral composition and grain size vary widely. The average grain size is around 35 μm , and quartz, muscovite, chlorite and some biotite can be detected. Chlinoclore, a phyllosilicate is likewise present, either isolated or in aggregates. Its crystals are not oriented and range in size from 35 to 190 μm (Figure 2i). This material is also characterized by long parallel cracks, indicative of the plane of foliation.

4- Petrophysical characteristics

Table 1 shows the mean values of the petrophysical characteristics determined for the nine varieties of natural stone analyzed. The granites from the westernmost quarries of the Autonomous Community of Madrid, Zarzalejo and Cadalso exhibited the greatest porosity and lowest Vp, while the granites with the highest Vp values and lowest porosity were found in the northern part of the region, Colmenar Viejo and Alpedrete. Of the carbonate rocks, the Cretaceous dolostones (Redueña and Torrelaguna) had the highest porosity and lowest Vp values, while the Miocene limestones (Colmenar de Oreja) had the highest Vp and lowest porosity.

The Sierra Negra slates exhibited moderate to high porosity (>8 %) and a moderate to low Vp value (3 455 m/s), while porosity was very low (0.4 %) and the Vp very high (5 694 m/s) in the Bernardos slates.

The sorptivity of these materials varied with their porosity: the higher the porosity, the greater the capacity to absorb water, although in no case in the stone studied did the absorbed water exceed 6 % on average. The Redueña and Torrelaguna dolostones and the Sierra Negra slate were the stones with the highest water sorptivity, while Bernardos slate had the smallest value of all the varieties analyzed, followed by the granites (mean water absorption values of <0,7 %).

Table 2 shows the mean, maximum and minimum values for each anisotropy index determined and for each variety of stone, disregarding the atypical values. The anisotropy indices revealed differences and similarities among the rocks. The K, As, ARS and dM indices were very similar (Table 2) for the stones analyzed. The dM index exhibited the lowest values and the As the

highest, while the ARS results were the closest to the dM values. The farther these index values were from 0, the more anisotropic the materials. As the AD index ranges from 0 to 1, with 1 indicating isotropic material, its values logically differed from all the others, although its variation was proportional to, albeit in the opposite direction from, the variations recorded in the other indices, and depended on the type of stone.

The correlation coefficient among these indices was generally over 0.95 for all the materials, except for the dm index, where it ranged from 0.1 to 0.65. This was because this index does not take the velocity in the direction of maximum anisotropy (V_{pmin}) into consideration, but only the maximum and mean velocities (V_{pmax} and V_{pmean}).

The K, As, ARS and dM indices were higher for the Zarzalejo and Cadalso than for the Alpedrete and Colmenar granites, while the AD index was lower for the former than for the latter group. The Zarzalejo and Cadalso granites were consequently more anisotropic than the Alpedrete and Colmenar Viejo materials.

Of the carbonate rocks, the two dolostones were slightly more anisotropic than the limestones: the K, As, ARS and dM indices for the former were slightly higher, and the AD index lower, than for the limestones.

The anisotropy index values were very high for the slates, especially for the Sierra Negra material, because of their pronounced schistosity. This was the most anisotropic of all the varieties analyzed.

The carbonate rocks and granites had similar anisotropy indices, with values well below the findings for the slates. The Colmenar limestone, together with the Alpedrete and Colmenar Viejo granites, were the most isotropic varieties.

5- Classes of anisotropy

While any of the indices described in Section 2 above may be used to determine anisotropy classes, the indices proposed by Guydader and Denis (1986) were chosen because, unlike the others, they distinguish between two-dimensional (dm) and three-dimensional (dM) anisotropy, which affects the hydraulic behaviour and decay of these materials. These indices provide additional information for the definition of anisotropy classes, inasmuch as they indicate the spatial direction in which the structural, textural or mineralogical orientation that determines anisotropy is concentrated.

The relative (dm%) and total (dM%) anisotropy values for each of the specimens made of the nine types of stone analyzed are shown in the charts in Figure 3. Cluster analysis identified five classes of anisotropy (I-V, respectively the lowest to the highest) in the dolostones and granites from Cadalso de los Vidrios and Colmenar Viejo, and only three classes (I-III) in the Colmenar limestone, the granites from Alpedrete and Zarzalejo and the two varieties of slate.

Table 3 shows the mean relative (dm) and total (dM) anisotropy values for each of the classes identified, the percentage of specimens in each class, and the Vp values (V_{pmax} , V_{pmean} , V_{pmin}). The new dMm index ($dMm: dM+dm$), one of the parameters that may provide the best measure of the degree of material anisotropy, is also shown. The higher the dMm index, the more anisotropic the rock.

High total anisotropy index values (dM%), in conjunction with low values for relative anisotropy (dm%), were an indication that anisotropy in the stone was more intense in one of the three spatial directions. This may have been due to a preferential structural, textural or compositional orientation, slight in the case of the Zarzalejo and Cadalso granites, the Colmenar limestone and the Torrelaguna dolomite, and very pronounced in both types of slate. By contrast, when dM% and dm% values were similarly high, no single direction predominated in maximum anisotropy. In this case, the entire variety is altered equally in all three directions. The Alpedrete and Colmenar Viejo granites and the Redueña dolostone are a case in point.

The anisotropy indices for the Alpedrete and Colmenar Viejo granites were lowest, and while the dM and, especially, the dMm values tended to increase from the lower (class I) to the higher (classes III and V) classes, the dm values did not. Their anisotropy values were low and their mean Vp values were fairly high (Alpedrete, 4 600 m/s and Colmenar Viejo, 5 200 m/s). The dM index was also much greater in the higher classes for the Zarzalejo and Cadalso de los Vidrios granites, with values of over 16. Their mean Vp values were 3 300 and 3 600 m/s, respectively.

In the carbonate rocks and the Colmenar limestones, the dM values rose in the higher classes. This trend was also observed in the dolostones, which had the highest anisotropy indices (dM and dm). The Colmenar limestone was the variety with the highest mean Vp, nearly 6 000 m/s, while the values for the stone from Torrelaguna and Redueña were 3 800 and 2 700 m/s, respectively.

The Bernardos and Sierra Negra slates showed the highest dM and dMm anisotropy values of all of the materials studied and the relative anisotropy value, dm, was also very high for the Sierra Negra slates. While these materials are highly anisotropic in general, because of its greater schistosity, the slate from Sierra Negra exhibited the highest anisotropy (dM 76.6 % and dm 27.7 %). Anisotropy, which rose progressively from class I to class III, was mirrored in a slightly downward trend in the Vp. This same trend was observed in the Bernardos slates, but with smaller anisotropy indices (dM 35.7 % and dm 5.3 %). The Bernardos material had a higher ultrasound propagation velocity (5 700 m/s) than the other slates.

Mean ultrasound velocity is a parameter widely used to determine the quality of building stone. Vp declined with rising anisotropy only in the most anisotropic varieties (slates). In all other varieties, no significant trend was perceptible. This was because taking the values of the three directions measured in each specimen into consideration narrowed the differences defined by the plane of anisotropy, which is characterized by a decline in Vpmin. Since determining the mean Vp proved to be scanty significant, finding the mean Vpmin value was a more meaningful measure of material quality, as in the case of the granites from Zarzalejo and Cadalso as well as the dolostones. In the more isotropic rocks (Colmenar limestone, Alpedrete and Colmenar Viejo granites), no significant variations were observed in the Vp value. Consequently, anisotropy indices that take Vpmin into consideration and relate it to the other directions (Vpmax and Vpmean) furnish more information about rock behaviour. The distribution of the specimens in each anisotropy class was also indicative of stone quality. Specimens made from less anisotropic materials with a greater ultrasound propagation velocity were consistently more suitable, for they were less severely or at least more uniformly altered. Fort *et al* 2008 reported that increased anisotropy in rocks may be due to alteration, which entails a rise in porosity. It may also be caused by the heterogeneity of the stone with different diagenetic processes (cementation or dolomitization/de-dolomitization), as in the case of carbonate rocks. Quality proved to be highest in the Colmenar limestone, as most of its specimens were in classes I and II, with lower dM, dm and dMm index values than the Redueña and Torrelaguna dolostones. Redueña exhibited the least desirable behaviour.

The Alpedrete and Colmenar Viejo granites had a larger number of specimens in the less anisotropic classes, while the Zarzalejo and Cadalso de los Vidrios materials had more specimens in the higher classes. In both slates more than fifty percent of the specimens analyzed were in class II.

This explains how the anisotropy class affects material alteration. Alteration would be faster, more intense and more uneven in Redueña than in Torrelaguna dolostone, where the process would be more uniform; the Alpedrete granite, like the Colmenar limestone, would behave more uniformly than the Zarzalejo and Cadalso granites, primarily because the anisotropy classes of the latter two are characterized by higher indices than the Alpedrete and Colmenar varieties.

6- Stone hydraulic behaviour and anisotropy

The heterogeneity in composition, texture and structure of construction materials underlies the differences in their rate of decay. Positioning materials along their planes of anisotropy may favour these processes by facilitating water ingress. Water absorption in natural building stone may, then, be influenced by its degree of anisotropy. Water tends to be absorbed differently depending on the preferential directions of anisotropy, which are characterized primarily by the position and distribution of the pores in the rock along preferential planes. In principle, water absorption at atmospheric pressure should not be affected by the degree of anisotropy of the materials, for in

that test water penetrates inward into the rock in all directions, because the specimen is completely immersed in water. Nonetheless, the planes where more pores are concentrated may tend to fill more quickly than in other directions, in which water is taken up more slowly. This may explain the change in slope of the absorption and desorption curves (amount of water absorbed/evaporated over time) in some of these materials, as more porous planes facilitate the movement of water.

Water absorption and porosity accessible to water

These two parameters are directly related: the more porous a rock, the more water it can absorb. Similarly, materials with a high level of porosity accessible to water show the lowest ultrasound propagation velocity. In this study, the dolostones and Sierra Negra slates absorbed more water than any of the other materials.

Both types of dolostones (Redueña and Torrelaguna) exhibited a high degree of porosity accessible to water, very likely as a result of their diagenetic dolomitization. The specimens in the highest anisotropy classes had the highest porosity, and consequently the greatest sorptivity.

These high porosity values were also observed in the slates, more visibly in the Sierra Negra than in the Bernardos material, due to the greater schistosity in the former.

In the Colmenar limestone, whose average porosity was 3.8 % (Table 1), the value declined as the anisotropy index rose (dM%) (Tables 3 and 4). All the granitic rocks followed a similar pattern: as their anisotropy rose (dM%), their porosity dipped, except in the Colmenar Viejo granite, whose value rose from 0.72 % to 0.81 % (Table 4) between class I and class V.

In short, in the less anisotropic rocks, no significant trends were observed in the average porosity and water absorption values with rising anisotropy, whereas in the more anisotropic rocks (dolostones and slates), porosity and water absorption increased with anisotropy.

Capillary water uptake

Capillary water uptake is conditioned by the pore distribution in the stone and tends to rise in systems with a predominance of capillary pores, which favour water ingress when absorption takes place along the planes of anisotropy.

Table 5 shows the water absorption coefficients for the classes with the highest and lowest anisotropy in each type of rock. As a rule, the most anisotropic rocks had the highest capillary coefficient. Hence, the Zarzalejo and Cadalso de los Vidrios stones, the most porous granites with the lowest propagation velocity (Table 1) exhibited the highest anisotropy indices (Table 3) and absorption coefficients (Table 5). The same pattern was observed in the carbonate rocks: the Redueña stone had very high absorption coefficient values, most likely because it had the highest porosity accessible to water and a significant percentage of capillary pores (Fort *et al* 2008). This same relationship was observed in the slates, where the coefficient for the Sierra Negra material was much higher than the value for Bernardos stone.

The absorption capillary coefficient was generally observed to be higher in the more anisotropic classes of each type of stone. The differences between the values of this coefficient for the most and least anisotropic classes were slight, except in the case of the Alpedrete granite, where the coefficient rose from $1.52 \text{ g}\cdot\text{m}^2\cdot\text{s}^{-0.5}$ in class I to 3.13 in class III, and the Torrelaguna dolostone, which varied from 8.03 to $45.05 \text{ g}\cdot\text{m}^2\cdot\text{s}^{-0.5}$ (Table 5). Moreover, when capillary water was taken up in a direction perpendicular to the planes of weakness or anisotropy, the absorption coefficient was observed to be lower than when it was measured in a direction parallel to that plane. In other words, the absorption coefficients were higher when measured parallel to the direction of the lowest anisotropy in the stone (e.g., the Sierra Negra slate). These increases were more significant in the varieties in which anisotropy was generated by planar phenomena, such as cracking or open schistosity (Sierra Negra slate), or stromatolite growth (Torrelaguna stone). The dm% index was usually high in these varieties, such as in the Redueña stone.

In a nutshell, the capillary absorption coefficient tends to rise with rising porosity and anisotropy (Tables 3, 4 and 5), especially when water ingress is influenced by the presence of planar structures that facilitate suction and circulation inside the rock.

Conclusions

The dM and dm indices are regarded to be best suited to determining the degree of anisotropy in rocks, and a new index, dMm , is proposed that provides an overall measure of anisotropy with which to establish stone quality.

As expected, the Bernardos and Sierra Negra slates were the varieties of natural stone with the highest degree of anisotropy, with class III dMm values of 48.3 % and 119.0 %, respectively. On the grounds of anisotropy, ultrasound propagation velocity and lower open porosity, the Bernardos slate was of higher quality than the Sierra Negra material. Furthermore, according to the values obtained, this variety was very heterogeneous, with higher standard deviations. The Bernardos slate water absorption coefficient was also very low, although it rose slightly when suction was along the plane of cleavage. These coefficients were much lower than in the Sierra Negra slates, which were not suitable for use in construction requiring high quality materials.

Colmenar limestone was the highest quality carbonate rock studied, with fairly low anisotropy (dMm : 3.4 %). Its V_p values were the highest of all the natural stone varieties analyzed in this study. As the most isotropic stone, it can be laid with no regard to orientation. By contrast, the stone from Torrelaguna and Redueña had high anisotropy indices in classes IV and V. The class IV and V dMm values were respectively 13.3 % and 20.2 % for the Redueña variety, and 16.3 % and 18.5 % for the Torrelaguna stone. The materials in these classes are not recommended for use, as their anisotropy would have a very direct and adverse effect on decay. Of the two, resistance to decay would be higher in the less porous Torrelaguna dolostone than in the Redueña material. When limestone is the material chosen to restore the architectural heritage, use of the Torrelaguna variety is recommended whenever possible, providing it is compatible with the original stone. The stones should be laid with the plane of greatest anisotropy or lowest V_p parallel to the direction in which the capillary water is absorbed (which is usually vertical and ascending), so as to minimize decay induced by water ingress.

Of the granites, in turn, the Alpedrete variety had the lowest anisotropy. This and the Colmenar Viejo stones were the highest quality granites, with high ultrasound propagation velocities and low effective porosity. Moreover, no significant variations were observed in the anisotropy from one class to another, with greatest uniformity found for the Alpedrete stone. In the other two varieties (Zarzalejo and Cadalso de los Vidrios), anisotropy was higher and the V_p lower, especially in the former. In this variety, only class I materials should be used, whose dMm index was 12.6 %. The Cadalso granite was of higher quality than the Zarzalejo stone.

Mean ultrasound propagation velocity is not enough to determine the quality of the natural stone; the minimum propagation velocity (V_{pmin}) is more representative, as it has a more direct effect on stone decay.

Construction material anisotropy determines resistance to the agents inducing decay. The way these materials are positioned may contribute to decay by facilitating water ingress. Capillary water uptake is also influenced by the direction that stone is laid: absorption is greater in more anisotropic, more porous varieties and in the direction parallel to the plane of anisotropy, where water is absorbed more quickly and with a higher coefficient (direction of the lowest ultrasound speed).

This explains the differential alteration of materials from the same quarry and forming part of the same structure, as these members and ashlar would often have been laid according to no apparent structural criteria or regard for their anisotropy. The least suitable arrangement would facilitate water ingress, in larger volumes in certain directions, expediting decay in those directions. Therefore, higher quality can be attained by choosing stone varieties with low anisotropy indices (classes I and II) and refraining from using materials in higher classes to build the same wall, for this may induce differential decay. Further to these criteria, the Alpedrete and Colmenar Viejo granites afford the highest quality because of their uniform anisotropy. Of the carbonate rocks, the Colmenar limestone offers the highest quality because of its petrophysical properties, as well as its very low, uniform anisotropic indices. The Torrelaguna dolostones would ensure better performance than the other variety as they absorb less water, and their anisotropic indices are more uniform (48 % of the test pieces are in class I). Bernardos slates would behave more satisfactorily than the Sierra Negra variety.

The determination of anisotropy in a material is a simple, repetitive process that entails very low costs after the initial investment in equipment. It can provide medium- to long-term savings by delaying decay and lengthening the durability of materials and, therefore, buildings or infrastructures. This is a preventive conservation method, in line with today's trends, as opposed to interventional or corrective conservation, which is always costlier and more invasive.

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Table 1. Mean values of ultrasound velocity transmission (Vp), bulk density, open porosity and water absorption at atmospheric pressure

	STONE	Vp (m·s ⁻¹)	Density (Kg·m ⁻³)	Open porosity (%)	Water absorption (%)
Granites	Alpedrete	4601±204	2669±17	0.8±0.1	0.3±0.0
	Zarzalejo	3296±198	2662±21	1.6±0.1	0.6±0.0
	Cadalso	3575±178	2599±15	1.2±0.2	0.5±0.1
	Colmenar Viejo	5190±207	2610±22	0.7±0.1	0.3±0.2
Carbonate rocks	Redueña	2753±314	2349±92	16.2±3.4	5.6±1.4
	Torrelaguna	3788±278	2527±38	10.0±1.4	3.3±0.6
	Colmenar	5941±111	2579±30	3.8±1.2	0.8±0.4
Slates	Bernardos	5694±183	2751±7	0.4±0.1	0.2±0.0
	Sierra Negra	3455±461	2598±42	8.2±1.1	2.6±0.4

Table 2. Anisotropic indices of the nine stones analysed: mean, maximum and minimum values

Stones	Anisotropy Indices	K	As	ARS	AD	dM%	dm%
Alpedrete granite	Mean	6.87	7.18	6.66	0.93	5.78	1.90
	Max-Min	11.00-2.15	11.76-2.12	10.52-2.14	0.98-0.89	9.38-1.72	8.81-0.02
Zarzalejo granite	Mean	14.86	16.41	13.97	0.86	12.65	3.08
	Max-Min	22.32-6.61	25.47-6.88	20.30-6.44	0.94-0.80	18.20-5.70	13.09-0.15
Cadalso granite	Mean	17.90	20.28	16.656	0.833	15.339	3.173
	Max-Min	28.98-7.38	34.62-7.68	25.72-7.13	0.93-0.74	23.91-4.65	13.70-0.07
Colmenar Viejo granite	Mean	4.58	4.73	4.45	0.96	3.48	2.05
	Max-Min	15.72-0.57	17.05-0.58	14.57-0.57	0.99-0.50	11.25-0.33	8.00-0.02
Redueña dolostone	Mean	7.69	8.11	7.32	0.93	5.58	3.85
	Max-Min	16.31-0.41	17.80-0.41	15.11-0.41	1.00-0.85	14.09-0.24	13.39-0.02
Torrelaguna dolostone	Mean	7.47	7.88	7.13	0.93	5.61	3.33
	Max-Min	18.02-0.69	20.02-0.69	16.68-0.69	0.99-0.81	14.25-0.42	11.84-0.13
Colmenar limestone	Mean	3.50	3.80	3.62	0.96	3.05	1.19
	Max-Min	7.13-0.09	7.33-0.09	6.83-0.09	1.00-0.93	5.23-0.08	5.95-0.01
Bernardos slate	Mean	41.22	55.97	35.45	0.65	33.59	5.80
	Max-Min	64.77-33.24	108.40-41.49	52.02-29.32	0.71-0.48	50.27-26.21	44.83-1.23
Sierra Negra slate	Mean	123.15	440.14	79.63	0.20	76.73	29.23
	Max-Min	168.92-84.57	997.83-167.98	90.89-62.68	0.37-0.09	88.58-58.88	76.33-2.40

Table 3. Anisotropy indices and ultrasound transmission velocity by cluster (velocities in $\text{m}\cdot\text{s}^{-1}$). The % column represents the percentage of samples analysed in each stone variety, and V_p is the mean value obtained from from $V_{p_{\max}}$, $V_{p_{\text{mean}}}$ and $V_{p_{\min}}$

		Cluster	dM%	dm%	dMn	%	$V_{p_{\max}}$	$V_{p_{\text{mean}}}$	$V_{p_{\min}}$	V_p
Granitic Rocks	Alpedrete	I	4.69	0.84	5.52	40	4783±112	4744±113	4539±96	4689±100
		II	4.93	3.67	8.61	30	4739±161	4568±163	4425±186	4577±167
		III	7.58	1.49	9.07	30	4816±94	4745±114	4408±95	4660±91
	Zarzalejo	I	10.81	1.54	12.35	35	3374±208	3322±195	2985±175	3227±189
		II	9.72	6.56	16.28	23	3411±209	3196±215	2982±188	3196±197
		III	16.02	2.65	18.67	42	3656±159	3561±156	3030±136	3416±146
	Cadalso de los Vidrios	I	8.50	4.14	12.64	20	3631±200	3550±224	3006±157	3623±194
		II	14.07	0.79	14.19	14	4042±228	3897±233	3380±167	3582±205
		III	14.48	3.29	17.77	29	3698±118	3593±152	3195±216	3580±187
		IV	20.89	1.58	22.48	22	3914±85	3783±122	3077±124	3567±129
		V	17.84	6.24	22.67	15	3841±139	3723±136	3227±375	3542±155
	Colmenar Viejo	I	1.75	0.95	2.70	36	5360±100	5219±131	5005±242	5153±182
		II	2.66	2.45	5.11	20	5394±113	5236±139	5053±186	5163±223
		III	4.85	1.10	5.95	21	5276±84	5081±75	4970±99	5272±104
		IV	3.44	4.91	8.35	11	5416±125	5262±165	5013±236	5204±228
		V	7.82	3.22	11.03	12	5336±100	5153±139	4939±238	5248±188
Carbonate Rocks	Torrelaguna dolostone	I	2.69	1.64	4.34	47	3858±290	3795±291	3724±294	3792±290
		II	4.71	5.26	9.97	14	3667±196	3508±179	3419±195	3531±189
		III	9.44	2.15	11.59	19	3970±348	3887±355	3557±317	3805±339
		IV	7.87	8.39	16.26	11	3954±216	3627±160	3494±167	3692±178
		V	13.39	5.09	18.48	9	4102±370	3907±317	3470±294	3826±320
	Redueña dolostone	I	1.94	1.57	3.51	33	2860±332	2815±326	2783±329	2820±329
		II	5.65	2.09	7.74	19	2890±315	2840±307	2715±311	2815±310
		III	5.52	7.07	12.59	20	2854±278	2661±271	2606±272	2707±272
		IV	9.43	3.65	13.33	19	2732±315	2618±316	2385±287	2578±304
		V	9.52	10.7	20.23	9	2879±259	2564±205	2448±174	2630±208
	Colmenar limestone	I	1.48	0.51	1.99	42	5954±121	5881±136	2573±31	5939±121
		II	3.42	1.68	5.11	36	5967±99	5812±117	2586±31	5949±97
		III	5.21	0.49	5.69	22	6008±139	5708±116	2590±27	5918±131
Slates	Bernardos	I	33.06	3.81	36.87	32	6567±105	6315±92	4271±283	5717±120
		II	31.62	6.21	37.83	55	6649±79	6248±73	4385±283	5761±89
		III	42.39	5.90	48.29	13	6660±110	6278±82	3727±252	5555±105
	Sierra Negra	I	67.88	21.2	89.09	22	5121±641	4114±305	1487±264	3574±372
		II	77.01	27.9	104.9	50	5363±453	4053±420	1079±151	3498±302
		III	84.89	34.1	119.0	28	5494±182	3930±596	712±114	3379±252

Table 4. Density, open porosity and water absorption at atmospheric pressure of each of the clusters of the anisotropy classes

	Cluster	Density (Kg.m ⁻³)	Porosity %	Water absorption %
Alpedrete granite	I	2670±20	0.80±0.11	0.29±0.05
	II	2668±12	0.88±0.14	0.31±0.03
	III	2667±18	0.77±0.08	0.28±0.04
Zarzalejo granite	I	2656±19	1.68±0.08	0.58±0.03
	II	2660±15	1.68±0.09	0.58±0.03
	III	2669±25	1.58±0.12	0.54±0.04
Cadalso de los Vidrios granite	I	2599±19	1.34±0.24	0.49±0.08
	II	----	-----	-----
	III	2602±18	1.26±0.23	0.47±0.08
	IV	2595±70	1.13±0.19	0.41± 0.07
	V	2598±13	1.28±0.27	0.48±0.09
Colmenar Viejo granite	I	2613±23	0.72±0.12	0.36±0.02
	II	2604±15	0.74±0.19	0.41±0.29
	III	2625±30	0.73±0.13	0.32±0.06
	IV	2600±11	0.70±0.16	0.28±0.06
	V	2606±28	0.81±0.19	0.33±0.07
Torrelaguna dolostone	I	2531±49	9.89±1.70	3.31±0.67
	II	2537±30	9.57±1.08	3.15±0.40
	III	2517±23	10.46±1.20	3.49±0.51
	IV	---	---	---
	V	2510±35	10.76±1.00	3.62±0.44
Redueña dolostone	I	2365±88	15.8±3.20	5.44±1.31
	II	2369±70	15.0±1.40	5.11±0.42
	III	2397±47	14.5±2.00	4.81±0.77
	IV	2293±11	18.4±4.30	6.67±1.72
	V	2350±76	16.2±2.70	5.38±1.15
Colmenar limestone	I	2572±30	4.07±1.45	0.92±0.52
	II	2586±31	3.76±0.98	0.78±0.17
	III	2590±27	3.29±1.20	0.62±0.17
Bernardos slate	I	2754±50	0.42±0.05	0.16±0.01
	II	2748±40	0.39±0.03	0.14±0.01
	III	2751±11	0.51±0.06	0.19±0.01
Sierra Negra slate	I	2598±38	7.12±0.43	2.23±0.09
	II	2618±30	8.77±0.99	2.81±0.35
	III	2575±49	8.49±0.93	2.66±0.30

Table 5. Capillarity coefficient coefficient ($\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-0.5}$) of the stone varieties according to the anisotropic class, anisotropic indices and orientation of the material during the capillarity test

	Anisotropy Class	dM (%)	dm (%)	Orientation	Capillarity coefficient
Alpedrete granite	I	2.10	0.91	$\perp$ =	1.523 1.639
	III	9.38	2.55	$\perp$ =	3.128 3.983
Zarzalejo granite	I	9.31	1.90	$\perp$ =	4.238 4.580
	III	17.45	7.15	$\perp$ =	4.648 4.796
Cadalso granite	II	7.90	0.77	$\perp$ =	3.502 4.706
	V	22.60	8.40	$\perp$ =	3.635 3.695
Colmenar Viejo granite	I	1.14	0.70	$\perp$ =	0.969 1.437
	V	7.38	3.84	$\perp$ =	1.054 1.058
Torrelaguna dolostone	I	1.92	1.47	$\perp$ =	8.029 27.831
	IV	11.30	9.50	$\perp$ =	45.053 52.025
Redueña dolostone	I	0.66	0.14	$\perp$ =	86.228 86.187
	V	11.34	14.46	$\perp$ =	77.821 88.093
Colmenar limestone	I	0.08	0.01	$\perp$ =	1.429 1.544
	II	5.21	2.01	$\perp$ =	1.488 3.589
Bernardos slate	I	29.59	1.23	$\perp$ =	0.171 0.227
	III	48.29	8.06	$\perp$ =	0.191 0.280
Sierra Negra slate	I	60.68	31.30	$\perp$ =	5.987 16.760
	III	88.40	26.17	$\perp$ =	5.331 22.839

1. Alpedrete monzogranite, 2. Zarzalejo monzogranite, 3. Cadalso leucogranite, 4. Colmenar Viejo monzogranite, 5. Redueña dolostone, 6. Torrelaguna dolostone, 7. Colmenar limestone, 8. Bernardos slate, 9. Sierra Negra slate.

Figure 2. Micrographs at the polarized optical microscope of thin sections from the nine stone materials studied: a) Alpedrete granite, b) Zarzalejo granite, c) Cadalso granite, d) Colmenar Viejo granite, e) Torrelaguna dolostone, f) Redueña dolostone, g) Caliza de Colmenar limestone, h) Bernardos slate, i) Sierra Negra slate

Figure 3. Anisotropic classes established in each stone type by clustering, and according to their anisotropy indices dM and dm .