This is a post-print of a paper published in the Bulletin of Engineering Geology and the Environment, in 2019. The paper was submitted on 20 June 2018, and this is the submitted version, as requested by the Publisher Policy.

Effect of UV radiation on chromatic parameters in serpentinites used as dimension stones.

Rafael Navarro
Lidia Catarino
Dolores Pereira
Francisco Paulo de Sá Campos Gil

https://doi.org/10.1007/s10064-019-01469-3
Received: 20 June 2018 /Accepted: 9 January 2019 /Published online: 19 January 2019

Bulletin of Engineering Geology and the Environment

EFFECT OF UV RADIATION ON CHROMATIC PARAMETERS IN SERPENTINITES USED AS DIMENSION STONE

--Manuscript Draft--

Manuscript Number:	
Full Title:	EFFECT OF UV RADIATION ON CHROMATIC PARAMETERS IN SERPENTINITES USED AS DIMENSION STONE
Article Type:	Original Article
Corresponding Author:	Rafael Navarro Domínguez, Ph.D. Universidad de Salamanca Salamanca, Castilla y León SPAIN
Corresponding Author Secondary Information:	
Corresponding Author's Institution:	Universidad de Salamanca
Corresponding Author's Secondary Institution:	
First Author:	Rafael Navarro Domínguez, Ph.D.
First Author Secondary Information:	
Order of Authors:	Rafael Navarro Domínguez, Ph.D.
	Dolores Pereira Gomez, Full Professor
	Lidia Catarino, Assistant Professor
	Francisco Paulo de Sá Campos Gil, Auxiliary Professor
Order of Authors Secondary Information:	

Funding Information:		
	Geosciences Centre of the University of Coimbra (UID/Multi/00073/2013)	Dra. Lidia Catarino
	Fundação para a Ciência e a Tecnologia	Dra. Lidia Catarino
	Spanish Science and Technology Ministry (BTE2003–04812)	Dra. Dolores Pereira Gomez
	Spanish Science and Technology Ministry (CGL2004–03048)	Dra. Dolores Pereira Gomez
	Spanish Science and Technology Ministry (CGL2005-03048/BTE)	Dra. Dolores Pereira Gomez
	Spanish Science and Technology Ministry (CGL2006–05128/BTE)	Dra. Dolores Pereira Gomez
	Spanish Science and Technology Ministry (CGL2010-18579/BTE)	Dra. Dolores Pereira Gomez
	Spanish Geological Survey (IGME) (CONSTRUROCK)	Dr. Rafael Navarro Domínguez
	General Foundation of the University of Salamanca (Doctor TCUE 2015)	Dr. Rafael Navarro Domínguez

Abstract: Colour is an important parameter that must be taken into account if aesthetic homogeneity in both construction and restoration work is to be achieved. In serpentinites, the different degrees of serpentinization or carbonation can influence their final colour. Sunlight, especially ultraviolet (UV) radiation, is a common agent that can decay properties such as hue, lightness, and chroma, and can have a very significant effect on the ageing process of dimension stones. Testing the effects of UV radiation is fundamental for predicting how materials may behave upon exposure. In this work, the surface alteration of materials due to UV radiation by means of arc-xenon lamps was carried out to determine the degree of variation in colour parameters in serpentinites used as dimension stones. It was observed that the effect of UV radiation

Powered by Editorial Manager® and ProduXion Manager® from Aries Systems Corporation

	on the materials studied was low, and that the samples tended to darken and become yellow. Gloss was the parameter that varied the most, which decreased in all cases analysed. Associations were observed between the loss of gloss and a decrease in some parameters such as open porosity, water absorption at atmospheric pressure and water absorption by capillarity (direct), bulk density and uniaxial compression strength (reverse). The mineralogical composition of the stone was the main factor that influenced the loss of gloss. This research highlights the importance of the detailed study of colour variation in dimension stones due to exposure to UV radiation. The incorrect assessment of the damage that could be caused by this agent can lead to severe aesthetic destruction that may result in expensive legal actions.
Suggested Reviewers:	Eduardo Sebastian, Ph.D. Full Professor, Universidad de Granada rolando@ugr.es He is an expertise in the field of dimension stone for more than 30 years, being internationally recognized. He is the author and collaborator of numerous scientific papers published in the most prestigious international journals, as well as editor and author of several books on dimension stones applied to heritage
	Sabina Kramar, Ph.D. Researcher, Slovenian National Building and Civil Engineering Institute sabina.kramar@zag.si She is an expert in the characterization of building materials and their relationship with the environment. Currently participates in several European projects in relation to this topic. She is the author and collaborator of a large number of scientific papers in international journals, as well as book chapters related to dimension stones.
	Giovanna Antonella Dino, Ph.D. Research Assistant, Università degli Studi di Torino giovanna.dino@unito.it She is an Environmental Engineer, responsible of Mineral Dressing and Sampling Laboratory at the University of Torino. She is involved in EU, national and local projects about Enviroment, dimension stones and mining activities, their exploitation and application in historical and rural local buildings. She has numerous papers in international journals about the relation between the enviroment and the mining activities, speciallly concerning to dimensión stones

Powered by Editorial Manager® and ProduXion Manager® from Aries Systems Corporation

Manuscript
Click here to download Manuscript EFFECT OF UV RADIATION V.3.docx
Click here to view linked References

EFFECT OF UV RADIATION ON CHROMATIC PARAMETERS IN SERPENTINITES USED AS DIMENSION STONE

Navarro, R.[1a*]; Catarino, L.[2].; Pereira, D.[1b]; Gil, F.P.S.C.[3]

1 Geology Department, Science Faculty, University of Salamanca, Plaza Merced s/n, Salamanca 37008, Spain; ORCID ID: a: 0000-0002-0181-1193; b: 0000-0003-3893-4813

2 Earth Sciences Department, Geosciences Centre, University of Coimbra. Rua Sílvio Lima – Polo II 3030-790 Coimbra, Portugal; ORCID ID: 0000-0002-1476-7486

3. Dep. Physics, Centre for Physics of the University of Coimbra (CFisUC), and Molecular Chemical-Physics Unit, University of Coimbra. Rua Larga P-3004 516 Coimbra, Portugal; ORCID ID: 0000-0001-7546-0288

*Corresponding author:
E-mail address: rafanavarro74@gmail.com (R. Navarro)
Phone number: 0034 669 35 49 01

Abstract

Colour is an important parameter that must be taken into account if aesthetic homogeneity in both construction and restoration work is to be achieved. In serpentinites, the different degrees of serpentinization or carbonation can influence their final colour. Sunlight, especially ultraviolet (UV) radiation, is a common agent that can decay properties such as hue, lightness, and chroma, and can have a very significant effect on the ageing process of dimension stones. Testing the effects of UV radiation is fundamental for predicting how materials may behave upon exposure. In this work, the surface alteration of materials due to UV radiation by means of arc-xenon lamps was carried out to determine the degree of variation in colour parameters in serpentinites used as dimension stones. It was observed that the effect of UV radiation on the materials studied was low, and that the samples tended to darken and become yellow. Gloss was the parameter that varied the most, which decreased in all cases analysed. Associations were observed between the loss of gloss and a decrease in some parameters such as open porosity, water absorption at atmospheric pressure and water absorption by capillarity (direct), bulk density and uniaxial compression strength (reverse). The mineralogical composition of the stone was the main factor that influenced the loss of gloss. This research highlights the importance of the detailed study of colour variation in dimension stones due to exposure to UV radiation. The incorrect assessment of the damage that could be caused by this agent can lead to severe aesthetic destruction that may result in expensive legal actions.

Keywords: Chroma, colour, dimension stone, gloss, lightness, serpentinite.

1. Introduction

The chromatic characteristics of building materials depend on several factors, including mineralogical composition, the presence of small amounts of iron or manganese oxides or sulphides, such as pyrite (Bams and Dewaele 2007), the textural aspects of rock (Esbert et al. 1997, Erdogan 2000) and properties such as porosity (Erdogan 2000) or finish (Benavente et al. 2003, Simonot and Elias 2003, Sanmartín et al. 2011). In dimension stones, colour homogeneity is one of the most important characteristics considered, depending on the application and use. This is especially important outdoors where these stones have been used to construct historic and more recent buildings, since one of its main functions is decorative. This is also a common problem in large structures, where the material used could come from more than one quarry (Sousa and Gonçalves 2012).

Polished finishes are more popular than other types of finishes, such as rustic, honed and bush hammered, because at this stage the surface becomes bright and the material reaches its maximum definition in terms of the textural aspects that enhance its chromatic qualities (Careddu and Marras 2013). Prolonged exposure to sunlight, the main source of ultraviolet radiation (UV), together with increases and decreases in temperature or humidity, can cause serious problems of decay. These include scaling (Sáez-Pérez and Rodríguez-Gordillo 2008), biodeterioration due to the presence of black fungi and cyanobacteria (Sterflinger 2011), soiling due to the combination with atmospheric particles from pollution (Urosevic et al. 2012) and variations in colour due to oxidation of some minerals, such as pyrite or chromite (Winkler 1997, Meierding 2005). Many white marbles are prone to discolouration, turning into more creamy or yellowish tones; grey limestones tend to increase in lightness and whitening due to solar action; and some brown-hued travertines tend to bleach due to prolonged exposure to sunlight (Winkler 1997; Harrell et al. 2007). Serpentinites, on the other hand, tend to lose stone fragments and form yellowish and reddish crusts due to the possible alteration of relict olivine, rich in iron, and the transformation of chromite to limonite (Meierding 2005). Although polishing accentuates the chromatic characteristics of the materials and gives a high degree of protection against damage caused by climate changes, it also highlights defects in the stone (variations of tones and textures, veins, etc.) (Ozcelik et al. 2012). The polished surface of a dimension stone tends to become opaque, and the gloss value decreases as a result of varying weather conditions, such as rain, wind, snow, temperature variation (Winkler 1979) or friction due to use (Blanchard 2012).

In the case of serpentinites, this fact is even more relevant, since their characteristics present large variations in colour, texture and mineralogy, from one outcrop to the next, due to differing degrees of serpentinization or carbonation (Pereira et al. 2007; 2013; Navarro et al. 2013; 2018). Therefore, colour is an important parameter that must be measured and controlled for achieving a certain degree of homogeneity in new construction and restoration works and in the replacement of damaged fragments; thus avoiding undesirable visual effects (Fig.1). Additionally, possible legal actions can be prevented in

cases where potential colour changes in already placed stones have not been previously discussed with the client. The standard specification for serpentine dimension stone, ASTM C1526-02 (2002), indicates that an alteration in the colour and finish of the serpentine may occur as a result of exposure to changing weather conditions, which should be taken into account in the evaluation of the suitability of serpentinite for a specific emplacement and weather context. The alteration of serpentinite, due to exposure to the weather, can result in whitish mineral deposits on the surface of the stone, discolouration, and loss of the polished surface.

Fig. 1. Unattractive visual appearance of serpentinite slabs with different chromatic characteristics placed on the exterior the Granada Exhibition and Convention Centre

UV radiation caused by sunlight is a common decay agent of the colour properties of materials (hue, lightness, and chroma), and can have a very significant effect on the ageing process of building materials (Careddu and Marras 2013; Sáez-Perez and Rodriguez-Gordillo 2008). Therefore, testing the effects of UV radiation is fundamental for predicting how materials may behave upon exposure. The surface alteration of serpentinite caused by exposure to UV radiation was carried out using arc-xenon lamps to determine the degree to which the colour parameters change after such exposure. The xenon-arc radiation source, in combination with a filter system, was designed to modify the distribution of the radiation produced to match the spectral distribution in the visible and ultraviolet region of solar radiation.

Although colour measurement is a basic technique used in the field of conservation and restoration of dimension stone (Prieto et al. 2010), no method has been proposed for examining colour changes in the context of this study. Most methods involve paints and coatings, and there are different ISO and UNE standards that regulate their use. In addition, there are more studies on colour change produced after the application of protective coatings in restoration projects (e.g. Poli et al. 2006, Becherini et al. 2017, Carmona-Quiroga et al. 2018) or after the application of cleaning treatments (e.g. Grossi et al. 2007; Carmona-Quiroga et al. 2017; Pozo-Antonio et al. 2017; 2018). Moreover, there is a lack of publications that measure colour changes in stones. Only Careddu and Marras (2013) measure the effects of solar UV radiation on the gloss of marble and limestone samples. In this work, they use a similar method to the one used here, but instead filter radiation so that it mimics radiation coming through a glass window. Also, they apply 28 cycles of radiation in a 24-hour period.

The objective of this work was to determine variation in the chromatic characteristics of several types of polished serpentinites, extracted from the south of Spain, which occurs when UV radiation with specific wavelengths continuously strikes the surface of the stones. This study is particularly relevant because serpentinites from Granada (South-eastern Spain) are included as candidates as a Global Heritage Stone Resource (Cooper 2015, Marker 2015) and are found in many Spanish heritage buildings (Navarro et al. 2015). Likewise, the serpentinites from Macael (Almería, South-eastern Spain) are included, together with White Macael marble, as candidates as Global Heritage Stone Province, due to their frequent use in both recently built and historical buildings (Navarro 2016).

2. Materials and Methodology

2.1 Materials

The research material comes from two different areas in Andalusia (Southern Spain) and both have been previously described in detailed (Navarro et al. 2018 and references therein). The quarries where the stones were sampled are located in Guejar Sierra (Granada) and in the Macael area (Almeria) (South-eastern Spain) (Fig. 2). The quarry in Granada, "Barranco de San Juan", ceased its activity during the middle of the 20th century. The four quarries in the Macael area are called "Virgen del Rosario", "C.E. Verde Almeria", "La Carrasca" and "La Milagrosa". The stone from the quarry in Granada is known as "Verde Granada" (VG) and was used in the construction of historic buildings such as the Alhambra, the Granada cathedral and the Royal Palace in Madrid (Navarro et al. 2015). In the Macael area, the stones are called "Verde Macael" (VM) from the quarry "Virgen del Rosario", "Verde Almería" from the quarry "C.E. Verde Almeria", and samples CA and MI were taken from the quarries "La Carrasca" and "La Milagrosa", respectively. Both of these quarries have since been abandoned and there is no information about the commercial names of the stones extracted from them.

Geologically, these serpentinites are placed in the Nevado-Filábride Complex (Internal Zone of the Betic Cordillera) (Fig. 2), which can be subdivided into lower and upper units (Martín-Algarra et al. 2004). The serpentinites quarries are located in the upper unit. From the bottom to the top, this upper unit is composed of metaconglomerates, light micaschists and quartzites, dark schists and marble levels, with metamorphosed lenses of basic and ultrabasic rocks, totally or partially transformed to eclogites, amphibolites, and serpentinites interspersed throughout the sequence. The age of this unit is from Palaeozoic to Triassic (Gómez-Pugnaire et al. 2000; Puga et al. 2002).

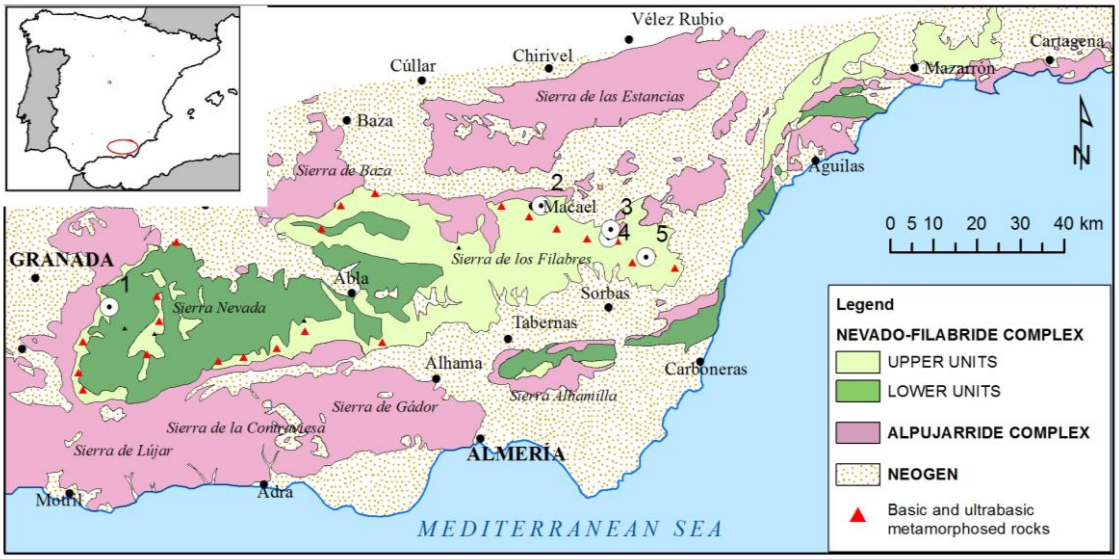

Fig. 2. Location of the quarries sampled: 1. Barranco de San Juan (VG); 2. Virgen del Rosario (VM); 3. C.E. Verde Almería (VA); 4. La Carrasca (CA). 5. La Milagrosa (MI) (Modified from Vera 2004)

Two polished tiles with the dimensions of 120 x 60 x 25 mm were studied from each serpentinite sample extracted. Two samples were selected from the stone type Verde Granada (VG) due to their differences in hue, light green (VG1) and dark green (VG2). Two samples with different carbonate contents (VM1 and VM2) of the stone type "Verde Macael" were studied, as well as the stone type Verde Almería (VA) and the two samples taken from La Carrasca (CA) and La Milagrosa (MI). The samples VG1, VG2 and VA were prepared at the Los Carriles Marble Handicraft Workshop in Macael, VM1 and VM2 were polished by the company "Mármoles Naturales, SL" and samples CA and MI were polished at the laboratory of the Spanish Geological Survey in Madrid.

Fig. 3 (a-g) shows the appearance of the different polished samples before testing. Fig. 3h shows schematically the location of the measured points.

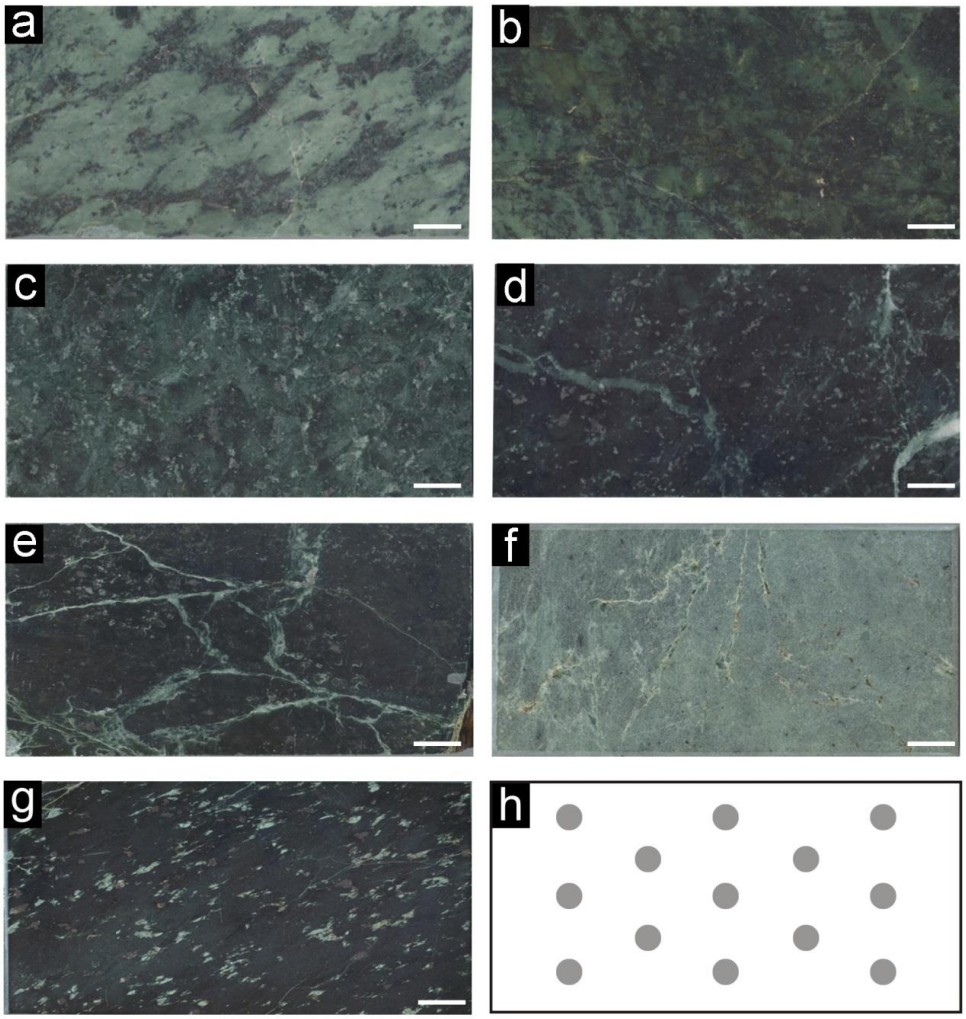

Fig. 3. Appearance of the polished samples before the test: a) Verde Granada 1 (VG1); b) Verde Granada 2 (VG2); c) Verde Macael 1 (VM1); d) Verde Macael 2 (VM2); e) Verde Almería (VA); f) La Carrasca (CA); g) La Milagrosa (MI); h) Location of the points measured. Scale bar = 1 cm.

The mineralogy of the samples studied is shown in Table 1 and the physical and mechanical properties of these serpentinites are shown in Table 2 (Navarro et al. 2013, 2018):

Sample	Main	Accessory	Secondary
VG1[+]	Atg	Ol, Px, Am, Chl, Cb, Crt, Mag, Py, Sp, Ilm, Hem	Cb, Tlc, Trm
VG2[+]	Atg	Ol, Px, Am, Chl, Cb, Crt, Mag, Py, Sp, Ilm, Hem	Cb, Tlc, Trm
VM1[+]	Cb	Atg, Px, Amp, Mag, Py, Ttn, Sp	Cb, Hmt
VM2[+]	Atg, Cb	Ol, Px, Chl, Crt, Mag, Ilm, Py, Sp	Cb, Ilm, Hem
VA[+]	Atg	Cb, Crt, Chl, Mag, Py, Ilm, Hem, Grt	Cb, Tlc, Hem
CA[+]	Px; Amp; Ep	Atg, Crt, Chl, Mag, Ilm, Chr, Hem	Cb, Tlc, Hem
MI[x]	Atg	Am, Crt, Chl, Mag, Ilm	Cb, Tlc, Hem

Table 1: Main mineral contents of the serpentinites studied ([+]Navarro et al. 2018; [x] Navarro et al. 2013).

Abbreviations: Atg: antigorite; Am: amphibole; Cb: carbonates; Chl: chlorite; Chr: chromite; Crt:

chrysotile; Ep: epidote; Grt: garnet; Hem: hematites; Ilm: ilmenite; Mag: magnetite; Ol: olivine; Px: pyroxene; Py: pyrite; Sp: sphalerite; Tlc: talc; Trm: tremolite; Ttn: titanite; (Abbreviations: Siivola and Schmid 2007)

Sample	BD (kg/m^3)	OP (%)	WAA (%)	WCC ($g/m^2s^{0.5}$)	V_p (km/s)	CS (MPa)	FS (MPa)	KM (MPa)
VG1[+]	2666 ± 27	0.53 ± 0.36	0.19 ± 0.12	0.66 ± 0.59	5.65 ± 0.48	331 ± 41	34 ± 7	2398 ± 1123
VG2[+]	2658 ± 9	0.92 ± 0.28	0.33 ± 0.07	1.17 ± 0.49	5.53 ± 0.44	361 ± 60	19 ± 9	3198 ± 1115
VM1[+]	2888 ± 59	0.27 ± 0.11	0.10 ± 0.03	0.26 ± 0.13	6.13 ± 0.43	139 ± 42	23 ± 10	1928 ± 721
VM2[+]	2704 ± 5	0.34 ± 0.07	0.16 ± 0.02	0.40 ± 0.09	6.02 ± 0.19	315 ± 43	14 ± 15	3235 ± 1432
VA[+]	2650 ± 11	1.37 ± 0.39	0.53 ± 0.11	2.19 ± 1.05	5.38 ± 0.45	246 ± 37	27 ± 12	1150 ± 319
CA[+]	2949 ± 256	1.31 ± 0.14	0.42 ± 0.08	*	5.03 ± 0.89	279 ± 84	22 ± 3	790 ± 106
MI[x]	2687 ± 17	0.73 ± 0.24	0.30 ± 0.08	*	5.76 ± 0.51	252 ± 34	*	1631 ± 583

Table 1: Main physical and mechanical parameters (mean and standard deviation) ([+]Navarro et al. 2018; [x] Navarro et al. 2013). Abbreviation: BS: Bulk density; OP: Open porosity; WAP: Water absorption at atmospheric pressure; WCC: Water absorption coefficient by capillarity; Vp: velocity of the ultrasonic p-waves; CS: Compressive strength; FS: Flexural strength; KM: Knoop microhardness. * Not tested.

2.2 Methodology

A Q-Lab climatic chamber at the Faculty of Science and Technology of the University of Coimbra (Portugal) (Q Sun Xenon Test Model Xe-3-HS) was used to simulate the possible effect that continuous exposure to UV Rays, with specific wavelengths, had on the chromatic parameters of polished serpentinite samples prepared for commercial use. The test conditions were carried out following the ISO 11341: 2004, which is standard that best fits. The temperature, humidity and radiation values were selected taking into account the climatic characteristics of the city of Granada (Spain). The annual average global solar radiation in the city, according to Sancho-Ávila et al. (2012) is 5.20 kW*h/m^2*day. The average relative humidity varies between 40% and 80% and the maximum average temperature is 34ºC (INM 2004). The weather station "Granada Base Aerea" (Latitude: 37° 8'13" N - Longitude: 3° 37' 53" W - Height: 687 masl) was the reference.

The samples were irradiated by the UV lamps during 4 hours, with an irradiance of 0.63 W / m^2 * per every wavelength (in nm), which is the result of the conversion of the average global radiation value in Granada. The ambient temperature was fixed at 40ºC, according to the ISO 11341 (2004) Standard and the relative humidity was fixed at 40%. After this treatment, the samples passed through a condensation step for 4 hours, at which time no UV radiation was applied. The ambient temperature was 40 °C and the relative humidity was 80%. The combination of these two steps constituted a cycle. The samples were exposed to 126 cycles and the total duration of the experiment was 1008 hours.

A Konica Minolta CM700d spectrophotometer from the Faculty of Science and Technology of the University of Coimbra (Portugal) was used to measure colour parameters in the specular component excluded mode (SCE). This decision was based on the work by Sanmartin (2012), who analysed the influence of the inclusion or exclusion of the specular component in the measurement mode on granites with different finishes (heterogeneous rocks from the point of view of colour). She found that the excluded specular component (SCE mode) magnifies the differences in colour produced by the different surface finishes, making this the preferred mode when analysing colour differences, as opposed to the specular component included mode (SCI). The illuminant mode used was 8º/D65, with a measuring window 8 mm in diameter (Medium Area View-MAV). The value 8º refers to the angle with respect to the position of the observer, while the illuminant D65 is a standard illuminant (daylight, colour temperature: 6504 K). The colour space used was based on the tristimulus values, L*a*b*, of the 1976 CIEL*a*b* colour system (Smith 1997) (Fig. 4). The data obtained were expressed by an alphanumeric code that represents the colour coordinates, that is to say the three axes of the three-dimensional CIEL*a*b* system, of the exact point that is being measured (Fig. 4). The vertical axis is L* (lightness) with a range of values from black (0) to white (100). The horizontal plane is defined by axes a* and b*. The a-axis varies between red (+60) and green (-60) and the b-axis varies between yellow (+60) and blue (-60).

The colour of the sample (chroma) is defined by the chromatic coordinates, C *, whose value is determined according to the expression (CIE 2004) (1):

$$C^* = \sqrt{(a^*)^2 + (b^*)^2} \qquad (1)$$

The hue angle (h_{ab}) is defined as (2):

$$h_{a^*b^*} = \arctan(b^*/a^*) \qquad (2)$$

The total colour difference is the Euclidean distance between two points within it. It is denoted as ΔE and its equation is (CIE 2004) (3):

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \qquad (3)$$

The differences in hue ΔH* is calculated according to the following formula (4):

$$\Delta H^* = \sqrt{(\Delta a^*)^2 + (\Delta b^*)^2 - (\Delta C^*)^2} \qquad (4)$$

Hue is positive if the hue angle h of the sample is greater than the reference angle and negative if the hue angle of the sample is smaller.

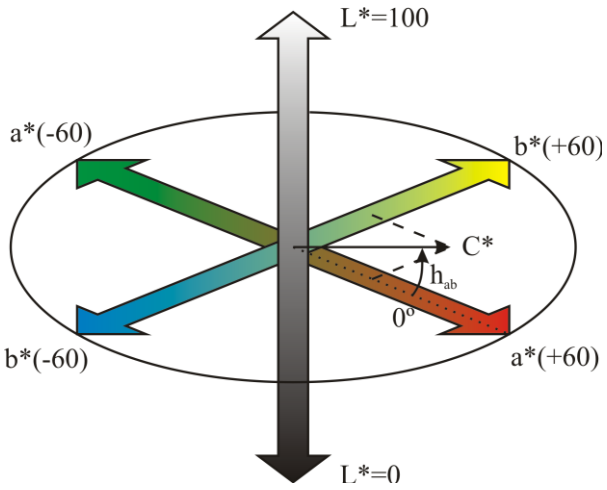

Fig. 4. Representation of the 1976 CIEL*a*b* space (modified from Smith 1997).

In addition, this spectrophotometer incorporates a reflectometer that allows the gloss value, expressed in GU (gloss units) at an angle of 8º, to be obtained. The gloss is defined as the ratio between the amount of light reflected from a surface and the amount of light incident on that surface (Cayless et al. 2011). This spectrophotometer measures the amount of light that is reflected when a ray of light hits the surface at a certain angle, which in this case was 8º. The values range from 0 (opacity) to 100 (maximum gloss) GU.

The mean values were obtained from a total of thirteen points, with the three measurements taken at each point (Fig. 3 h). The position of the spectrophotometer was fixed by a mask to measure always at the same points, and the values were measured at the start and at the end of the experiment.

3. Results

After carrying out 126 irradiation cycles, the colour parameters of the samples tested were measured and compared to the initial values recorded. Regarding the initial colour measurements, two groups with similar characteristics were established. It was observed that most of the samples presented values of L* that were around 23, ranging between 19.36 and 27.82. However, there were two samples, VG1 and CA, with a lighter colour, with L* values equal to 39.57 and 47.55, respectively, and somewhat higher values for parameters a* and b*. Table 3 lists the initial values and Table 4 the final values.

Sample	L*	a*	b*	C*	h_{ab} (º)	G^8 (GU)
VG1	39.57 ± 4.61	-6.70 ± 1.55	2.67 ± 1.58	7.21 ± 1.82	-21.75 ± 13.67	75.08 ± 7.69
VG2	23.33 ± 2.53	-3.09 ± 1.58	1.73 ± 1.60	3.54 ± 1.97	-29.33 ± 24.27	63.96 ± 6.43
VM1	27.82 ± 2.59	-5.66 ± 0.78	0.59 ± 0.50	5.69 ± 0.81	-5.94 ± 4.91	65.69 ± 5.72
VM2	22.19 ± 5.16	-3.69 ± 0.98	-0.21 ± 0.45	3.70 ± 0.97	3.27 ± 6.70	63.04 ± 8.47
VA	19.36 ± 3.45	-2.92 ± 1.01	-0.86 ± 0.55	3.04 ± 0.88	16.37 ± 13.98	62.65 ± 10.64
CA	47.55 ± 2.88	-6.11 ± 0.62	1.67 ± 1.18	6.33 ± 0.82	-15.26 ± 9.48	71.58 ± 6.84
MI	24.15 ± 3.37	-2.46 ± 0.52	-1.19 ± 0.78	2.73 ± 0.49	25.79 ± 16.56	80.08 ± 7.96

Table 3. Initial mean values and standard deviation of the main colour parameters.

Sample	L*	a*	b*	C*	h_{ab} (º)	G^8 (GU)
VG1	38.56 ± 4.46	-6.53 ± 1.38	2.57 ± 1.52	7.02 ± 1.68	-21.47 ± 12.13	70.85 ± 9.37
VG2	23.37 ± 2.45	-2.97 ± 1.57	1.79 ± 1.55	3.47 ± 2.05	-31.12 ± 16.81	60.15 ± 5.53
VM1	26.98 ± 2.45	-5.69 ± 0.71	0.81 ± 0.42	5.75 ± 0.74	-8.12 ± 3.63	63.35 ± 4.82
VM2	22.14 ± 5.30	-3.92 ± 0.78	0.06 ± 0.35	3.92 ± 0.80	-0.83 ± 4.59	62.58 ± 5.40
VA	19.48 ± 3.16	-2.67 ± 0.93	-0.61 ± 0.50	2.74 ± 0.83	12.97 ± 13.82	56.12 ± 9.86
CA	46.55 ± 3.06	-6.01 ± 0.62	1.69 ± 1.16	6.24 ± 0.82	-15.71 ± 9.58	70.96 ± 5.83
MI	23.83 ± 3.25	-1.88 ± 0.54	-1.04 ± 0.87	2.15 ± 0.64	28.85 ± 22.42	74.58 ± 7.46

Table 4. Final mean values and standard deviation of the main colour parameters after 126 cycles.

From these values, the variation of lightness (ΔL*), chromatic coordinates (Δa* and Δb*), Chroma (ΔC *), gloss (ΔG8) were calculated. The colour difference (ΔE) and the hue difference (ΔH) were also obtained (Table 5).

Sample	ΔL*	Δa*	Δb*	ΔC*	Δh	ΔG^8 (GU)	ΔE_{ab}	ΔH
VG1	-1.02	0.17	-0.10	-0.19	0.28	-4.23	1.04	0.03
VG2	0.04	0.12	0.06	-0.07	-1.79	-3.81	0.14	0.11
VM1	-0.84	-0.03	0.22	0.05	-2.18	-2.35	0.87	-0.22
VM2	-0.04	-0.22	0.27	0.22	-4.10	-0.46	0.35	-0.27
VA	0.11	0.25	0.24	-0.30	-3.40	-6.54	0.37	-0.17
CA	-1.00	0.10	0.02	-0.09	-0.44	-0.62	1.01	-0.05
MI	-0.32	0.58	0.15	-0.59	3.06	-5.50	0.68	0.13

Table 5. Variation of the mean values after 126 cycles.

4. Discussion

The effect of the ageing process due to UV radiation on the materials studied was low. In the CIEL*a*b* colour space, variations in lightness and Chroma lower than 3 units are imperceptible to the human eye (Völz 2001). In this study, none of the samples exhibited variations greater than this value. However, all the samples, except for VA and VG2, tended to darken, meaning that the values obtained for lightness (L*) were lower. The values obtained for lightness (L*) in samples VA and VG2, on the other hand, varied only slightly. The chromatic coordinates a* and b* shifted towards yellow, and the Chroma (C*) and the hue (h) tended to decrease. The variation observed with respect to the other parameters, such as the total colour difference (ΔE_{ab}) and the difference in hue (ΔH), was also minimal. In some cases, especially with regard to the h_{ab} parameter, standard deviation was quite high, which may be the result of small differences in hue at the positions measured, owing to the presence of minerals with different hue characteristics. The variation of these parameters could be associated with the presence of some minerals, such as pyrite and iron oxides that, even in small quantities, can produce staining upon oxidation in wet conditions, modifying the colour characteristics (Bams and Dewaele, 2007). Other authors (Winkler 1997) have reported that sunlight can modify lightness due to bleaching and oxidation processes.

Gloss was the parameter that exhibited the most variation, as it had decrease in all of the samples. In the case of the VG1 sample, variation was -4.23 GU (5.64%), while in the darkest sample (VG2) it was -3.81

GU (5.95%). In the most carbonated samples, VM1 and VM2, the variation in gloss was of -2.35 GU (3.57%) and -0.46 GU (0.73%) respectively. In the VA variety, which had the largest decrease in lightness, the decrease in gloss was found to be -6.54 GU (10.44%). The samples of the variety CA varied only -0.62 GU (0.86%). Finally, in the MI samples, this variation was -5.50 GU (6.87%). Also, it was observed that the samples with greater mineralogical diversity had less gloss loss, which was less than 4%. This was the case of VM and CA, which contain other components such as carbonates, pyroxenes and amphiboles or antigorite. However, the samples which were comprised almost exclusively of antigorite (VG1 and VG2, VA and MI) suffered gloss losses ranging from 4% to 11%. It should be noted that the loss of gloss is caused by the erosion of a small amount of material on the surface of the stone, and that the loss of several hundred nanometers of surface material is sufficient enough to affect gloss. Furthermore, it is likely that the mechanism of erosion involves superficial chemical and mechanical processes (Wood et al. 2000), and different mineralogical compositions of the stone can lead to different results.

Some authors, such as Erdogan (2000), have related the value of gloss with physical parameters like porosity, the presence of fractures and intergranular contacts. Also, other authors such as Ozcelik et al. (2012) or Careddu and Marras (2013) relate variation in gloss in decay tests with certain textural characteristics, such as grain size, as well as other physical properties such as density, absorption and even compression strength. Only Careddu and Marras (2013) have assessed changes in gloss due to the action of UV rays on marbles and limestones used as dimension stones. In the present work, the relationship between different physico-mechanical parameters and variations in gloss has been established. The results obtained are shown in Fig. 5.

With respect to physical properties, there is a direct relationship between the loss of gloss and parameters such as open porosity, water absorption at atmospheric pressure and water absorption coefficient by capillarity. As the values for these parameters increases, so did gloss loss. However, the relationship between gloss and bulk density and ultrasonic velocity (VP) was the opposite. The loss of gloss decreased as the samples became denser. This was observed for all samples except for the CA variety, which showed different results with respect to water absorption at atmospheric pressure and water absorption coefficient by capillarity. In addition, the VM2 sample had different results with respect to bulk density. It is remarkable that the sample CA was the one that exhibited the most variation. This may be due to the fact that it has greater mineralogical diversity, and is a rock with incomplete serpentinization (Navarro et al., 2018), causing this stone to present behaviour that is different from serpentinites *sensu strict.*

With respect to the relationship between the loss of gloss and mechanical properties, an inverse relation was observed when gloss was compared to the compression strength of the sample. The loss of gloss decreased as the value of compressive strength increased. Based on this relationship, we were able to divide the samples into two groups. The first group included the samples with the highest carbonate content and the greatest mineralogical diversity such as VM1, VM2, and CA. These were the samples with the least amount of gloss loss (less than 4%). The second group included the samples comprised

almost exclusively of antigorite, such as VG1, VG2, VA and MI, and which had a gloss loss ranging from 4 to 11%.

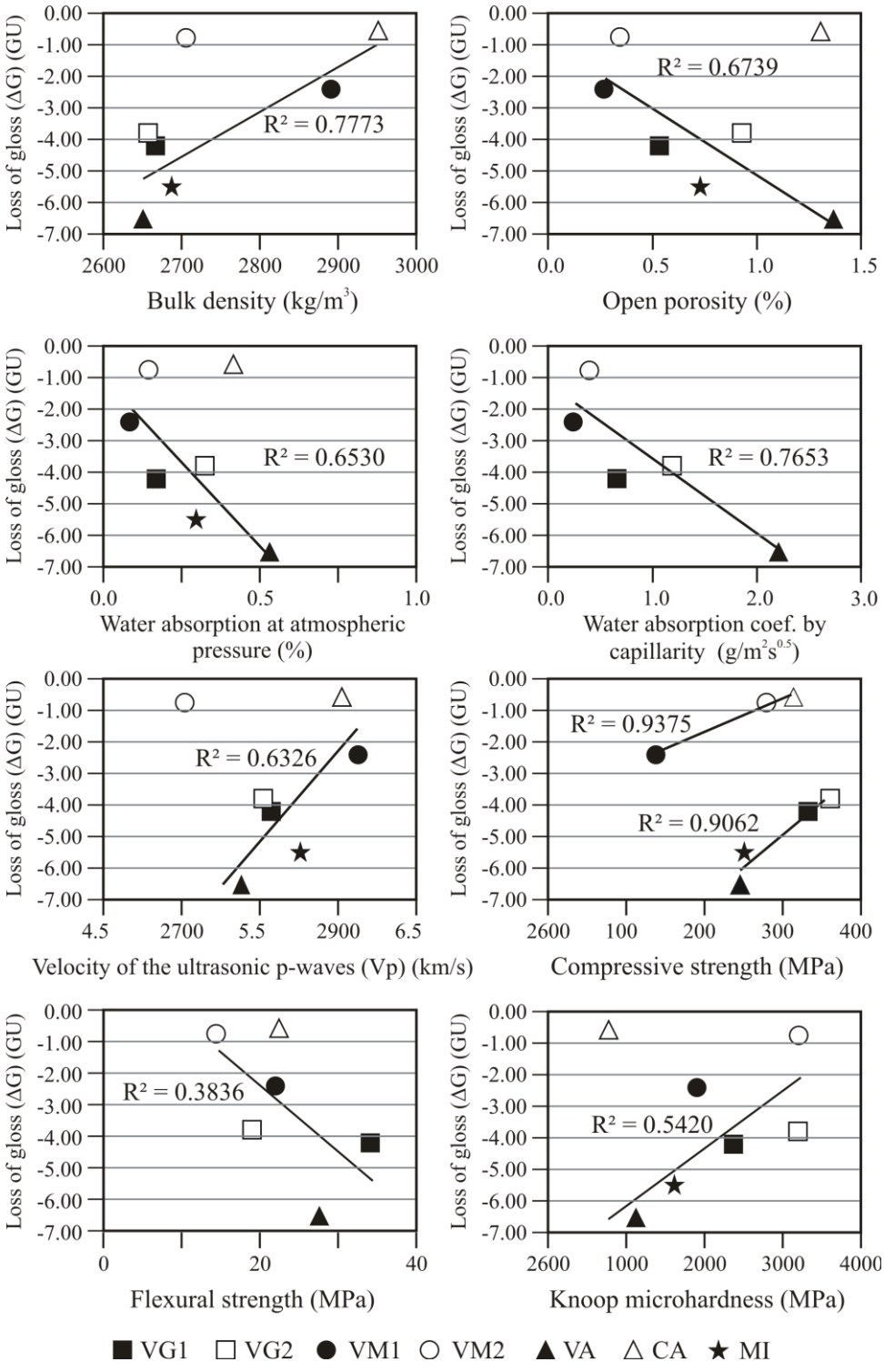

Fig. 1. Correlations between the different physical and mechanical parameters and loss of gloss.

No relationship was observed between the loss of gloss and flexural strength. Also, the differences that were observed for compressive strength, due to mineralogical composition, were not observed for flexural strength. With respect to the Knoop microhardness, a certain inverse relationship was also observed. The

samples exhibiting higher values for this parameter had less loss of gloss, which was the case for all of the samples analysed, except for the CA variety. In this sample the results were different, being similar to what was observed for the physical properties. However, it should be noted that the correlation coefficient was relatively low in this case.

Density, water absorption at atmospheric pressure, compression strength and flexural strength did not influence the gloss. Careddu and Marras (2013) indicate that only porosity and grain size are related to the loss of gloss in limestones and marbles. Our findings are in agreement with this, as it was observed that porosity, water absorption by capillarity and compression strength were the parameters with the best correlation. This is due to the fact that during the polishing treatment, the pores are sealed in the most superficial layer, increasing the gloss (Primavori 2006). This superficial layer will later be the one that will be most altered due to the action of ultraviolet rays, due to the change of its physical properties (Careddu and Marras 2013).

5. Conclusions

After carrying out the accelerated ageing experiment by applying UV radiation with an arc-xenon lamp onto polished samples of serpentinite it is observed that:

- The main parameters that define colour in the CIE L*a*b* space showed very little variation. In general, it was possible to observe a decrease in lightness, a tendency towards turning yellow and a decrease in colour saturation (Chroma).

- The greatest variation was observed in the gloss, which decreased within the range of 0.46 GU (0.73%) to -6.54 GU (10.44%) and was lower in the samples with a more diverse mineralogical composition.

- This loss of gloss was generally well correlated with physical parameters such as bulk density, porosity, absorption at atmospheric pressure and absorption by capillarity.

This work highlights the importance of the correct characterization of this type of material. The determination of the variation of colour caused by UV radiation is an aspect that is not usually taken into consideration when placing materials outdoors. The orientation in which the materials are placed determines how long they are exposed to sunlight and UV radiation. Thus, the colour of the stone can vary from one area to another, as well as aesthetic damage. In some cases the application of conservation treatments and even the replacement of certain pieces are sometimes necessary, which can result in economic losses. Additionally, determining colour in dimension stones and its variation due to UV radiation is essential in the study of the application of chemical and biological consolidating treatments, coatings or the application of cleaning processes that may damage the stone.

However, this topic has not been studied in detail and additional research is necessary for establishing how these treatments may interact with the properties of the stone. The possible alteration of colour due to different climatic conditions (intensity of UV radiation, time of exhibition or humidity), different

finishes (e.g. polished, hammered or brushed) and materials is an area that could lead to new research opportunities.

Acknowledgements

This work was made possible thanks to the FCT (Fundação para a Ciência e a Tecnologia, I.P.) and by the research project UID/Multi/00073/2013 of the Geosciences Centre of the University of Coimbra (Portugal), Spanish Science and Technology Ministry (MCT) through the projects: BTE2003–04812, CGL2004–03048, CGL2005-03048/BTE, CGL2006–05128/BTE and CGL2010-18579/BTE, Spanish Geological Survey (I.G.M.E.) through the project CONSTRUROCK (Natural stone and their relation with historic and monumental heritage and new building network) and the General Foundation of the University of Salamanca through the call Doctor TCUE 2015.

References

ASTM C-1526-02 (2002) Standard specification for serpentine dimension stone. American Society for Testing and Materials International, Pennsylvania (US)

Bams V, Dewaele S (2007) Staining of white marble Mater Charact 58:1052-1062 https://doi.org/10.1016/j.matchar.2007.05.004

Becherini F, Pastorelli Gç, Valotto G, Gambirasi A, Bianchin S, Favaro M (2017) Effects of protective treatments on particle deposition and colour variation in stone surfaces exposed to an urban environment Prog Org Coat 112:75-85 https://doi.org/10.1016/j.porgcoat.2017.06.029

Benavente D et al. (2003) Influence of surface roughness on color changes in building stones Color Res Appl 28:343-351 https://doi.org/10.1002/col.10178

Blanchard I (2012) The use of natural stone for internal flooring Proc Inst Civ Eng Constr Mater 165:177-187 https://doi.org/10.1680/coma.10.00015

Careddu N, Marras G (2013) The effects of solar UV radiation on the gloss values of polished stone surfaces Constr Build Mater 49:828-834 https://doi.org/10.1016/j.conbuildmat.2013.09.010

Carmona-Quiroga PM, Jacobs RMJ, Martinez-Ramirez S, Viles HA (2017) Durability of anti-graffiti coatings on stone: natural vs accelerated weathering PLoS ONE 12:e0172347 https://doi.org/10.1371/journal.pone.0172347

Carmona-Quiroga PM, Martinez-Ramirez S, Viles HA (2018) Efficiency and durability of a self-cleaning coating on concrete and stones under both natural and artificial Appl Surf Sci 433:312-320 https://doi.org/10.1016/j.apsusc.2017.10.052

Cayless A, Coaton JR, Marsden AM (eds) (2011) Lamps and Lighting (4 ed.). Taylor & Francis, New York

CIE (2004) CIE 15 Technical Report: Colorimetry, 3rd. edition vol 15.4. International Commission on Illumination, Vienna

Cooper BJ (2015) The "Global Heritage Stone Resource" designation: past, present and future. In: Pereira D, Marker B, Kramar S, Cooper B, Schouenborg B (eds) Global Heritage Stone: Towards International Recognition of Building and Ornamental Stones, vol 407 Geol Soc London Spec Publ, London, United Kingdom, pp 11-20. http://dx.doi.org/10.1144/SP407.7

Erdogan M (2000) Measurement of polished rock surface brightness by image analysis method Eng Geol 57:65-72 https://doi.org/10.1016/S0013-7952(99)00148-9

Esbert RM, Ordaz J, Alonso FJ, Montoto M, González-Limón T, Álvarez de Buergo-Ballester M (1997) Manual de diagnosis y tratamiento de materiales pétreos y cerámicos. Collegi d'Apparelladors i Arquitectes Tècnics de Barcelona, Barcelona

Gómez-Pugnaire MT, Braga JC, Martin JM, Sassi FP, Del Moro A (2000) Regional implications of a

Palaeozoic age for the Nevado-Filabride cover of the Betic Cordillera, Spain Schweiz Mineral Petrogr Mitt 80:45-52

Grossi MC, Esbert RM, Alonso FJ, Valdeón L, Ordaz J, Díaz-Pache F Colour changes and reactivity to SO2 of some cladding stones at the "Gran Teatre del Liceu" (Barcelona; Spain). In: Fassina V (ed) Ninth International Congress on Deterioration and Conservation of Stone, Venice, June 19-24 2000. Elsevier B.V., pp 323-328

Harrell JA, Broekmans MATM, Godfrey-Smith DI (2007) The origin, destruction and restoration of colour in Egyptian travertine Archaeom 49:421-436 https://doi.org/10.1111/j.1475-4754.2007.00312.x

INM (2004) Guía resumida del clima en España 1971-2000: Plan Estadístico Nacional, 2001-2004 vol 2013. Series Estadísticas, vol 2/12/2013. Dirección General del Instituto Nacional de Meteorología-Ministerio de Medio Ambiente Madrid

ISO 11341 (2004) Paints and varnishes. Artificial weathering and exposure to artificial radiation. Exposure to filtered xenon-arc radiation. A.E.N.O.R., Madrid

Marker B (2015) Procedures and criteria for the definition of Global Heritage Stone Resources. In: Pereira D, Marker B, Kramar S, Cooper B, Schouenborg B (eds) Global Heritage Stone: Towards International Recognition of Building and Ornamental Stones, vol 407 Geol Soc London Spec Publ, London, United Kingdom, pp 1-4. http://dx.doi.org/10.1144/SP407.7

Martín-Algarra Ac et al. (2004) Zonas Internas Béticas. In: Vera JA (ed) Geología de España. Sociedad Geológica de España (S.G.E.)-Instituto Geológico y Minero de España (I.G.M.E.), Madrid, Spain, pp 395-444

Meierding TC (2005) Weathering of serpentine stone buildings in the Philadelphia region: a geographic approach related to acidic deposition. In: Turkington AV (ed) Stone decay in the architectural environment, vol 399. Geol Soc Spec, pp 17-25

Navarro R (2016) Estudio de las serpentinitas del Complejo Nevado-Filábride (Cordillera Bética). Usos y aplicaciones en construcción y patrimonio arquitectónico. Dissertation, University of Salamanca

Navarro R, Pereira D, Gimeno A, Del Barrio S (2018) Influence of natural carbonation process in serpentinites used as construction and building materials Constr Build Mater 170:537-546 https://doi.org/10.1016/j.conbuildmat.2018.03.100

Navarro R, Pereira D, Rodríguez-Navarro C, Sebastian-Pardo E (2015) The Sierra Nevada serpentinites: the serpentinites most used in Spanish heritage buildings In: Pereira D, Marker B, Kramar S, Cooper B, Schouenborg B (eds) Global Heritage Stone: Towards International Recognition of Building and Ornamental Stones, vol 407 Geol Soc London Spec Publ, London, United Kingdom, pp 101-108. http://dx.doi.org/10.1144/SP407.7

Navarro R, Pereira MD, Gimeno A, Del Barrio S (2013) Verde Macael: a serpentinite wrongly referred to as a marble Geosci 3:102-113 https://doi.org/10.3390/geosciences3010102

Navarro R, Sánchez-Valverde J, Baltuille JM (2014) The natural stone in the historic buildings of the city of Granada (southern Spain). Features as a possible candidate for the designation of "Global Heritage Stone Province". In: Lollino G, Manconi A, Guzzetti F, Culshaw M, Bobrowsky P, Luino F (eds) Engineering geology for society and territory. Volume 5: urban geology, sustainable planning and landscape exploitation, vol 5. Springer International Publishing AG, Switzerland, pp 229-232. 10.1007/978-3-319-09048-1_40

Ozcelik Y, Careddu N, Yilmazkaya E (2012) The effects of freeze–thaw cycles on the gloss values of polished stone surfaces Cold Regions Sci Tech 82:49-55 https://doi.org/10.1016/j.coldregions.2012.05.007

Pereira MD, Peinado M, Blanco JA (2013) Misuse of natural stone for construction and the consequences in buildings. Case of study of serpentinites J Mater Civ Eng 25:1563–1567 https://doi.org/10.1061/(ASCE)MT.1943-5533.0000689

Pereira MD, Yenes M, Blanco JA, Peinado M (2007) Characterization of serpentinites to define their appropriate use as dimension stone. In: Prikryl R, Smith BJ (eds) Building Stone Decay: From Diagnosis to Conservation, vol 271 Geol Soc London Spec Publ, London, United Kingdom, pp 55-62

Poli T, Toniolo L, Sansonetti A (2006) Durability of Protective Polymers: The Effect of UV and Thermal Ageing Macromol Symp 238:178-183 https://doi.org/10.1002/masy.200650611

Pozo-Antonio JS, Montojo C, Lopez de Silanes ME, de Rosario I, Rivas T (2017) In situ evaluation by colour spectrophotometry of cleaning and protective treatments in granitic Cultural Heritage Int Biodeterior Biodegradation 123:251-261 https://doi.org/10.1016/j.ibiod.2017.07.004

Pozo-Antonio JS, Rivas T, Jacobs RMJ, Viles HA, Carmona-Quiroga PM (2018) Effectiveness of commercial anti-graffiti treatments in two granites of different texture and mineralogy Prog Org Coat 116:70-82 https://doi.org/10.1016/j.porgcoat.2017.12.014

Prieto B, Sanmartín P, Silva B, Martínez-Verdú F (2010) Measuring the color of granite rocks: A proposed procedure Color Res Appl 35:368-375 https://doi.org/10.1002/col.20579

Primavori P (2006) Technologies for stone extraction and processing. In: Promorana SRL (ed) Directory Marmomacchine. Associazione Italiana Marmomacchine, Milano (Italy),

Puga E, Díaz de Federico A, Nieto JM (2002) Tectonostratigraphic subdivision and petrological characterisation of the deepest complexes of the Betic zone: a review Geodinam Acta 15:23-43 https://doi.org/10.1080/09853111.2002.10510737

Sáez-Pérez MP, Rodríguez-Gordillo J (2008) The influence of solar radiation on the deterioration of the marble columns in the courtyard of the lions in the Alhambra Stud Conserv 53:145-157 https://doi.org/10.1179/sic.2008.53.3.145

Sancho Ávila JM, Riesco-Martín J, Jiménez-Alonso C, Sánchez de Cos-Escuin MC, Montero-Cadalso J, López-Bartolomé M (2012) Atlas de radiación solar en España utilizando datos del SAF de Clima de EUMETSAT. AEMET-MAAMA, Madrid

Sanmartín P (2012) Cuantificación del color en el estudio de la formación de biofilms en rocas graníticas del Patrimonio histórico artístico. Dissertation, University de Santiago de Compostela

Sanmartín P, Silva B, Prieto B (2011) Effect of surface finish on roughness, color and gloss of ornamental granites J Mater Civ Eng 23:1239-1248 https://doi.org/10.1061/(ASCE)MT.1943-5533.0000285

Siivola J, Schmid R (2007) List of mineral abbreviations. In: Fettes D, Desmons J (eds) Metamorphic rocks: a classification and glossary of terms. Cambridge University Press, Cambridge, U.K., pp 93-110

Simonot L, Elias M (2003) Color change due to surface state modification Color Res Appl 28:45-49 https://doi.org/10.1002/col.10113

Smith KJ (1997) Colour-order systems, colour spaces, colour difference and colour scales. In: McDonald R (ed) Colour Physics for Industry. Society of Dyers and Colourists, West Yorkshire, England, pp 121-208

Sousa LMO, Gonçalves BMM (2012) Color Assessment of Granitic Rocks and Implications For Their Ornamental Utilization Color Res Appl 37:375-383 https://doi.org/10.1002/col.20681

Sterflinger K (2011) Biodeterioration of Stone. In: Siegesmund S, Snethlage R (eds) Stone in Architecture: Properties, Durability (4th edition). Springer-Verlag Berlin, Berlin, pp 291-302

Urosevic M, Yebra-Rodríguez A, Sebastián-Pardo E, Cardell C (2012) Black soiling of an architectural limestone during two-year term exposure to urban air in the city of Granada (S Spain) Sci Total Environ 414:564–575

Vera JAe (2004) Cordillera Bética y Baleares. In: Vera JA (ed) Geología de España. Sociedad Geológica de España (S.G.E.)-Instituto Geológico y Minero de España (I.G.M.E.), Madrid, pp 345-464

Völz HG (2001) Industrial color testing: fundamentals and techniques. 2nd Edition. Wiley-VCH, Weinheim (Germany)

Winkler EM (1979) The Lightness (Reflectance) of Stone in the Stone Industry APT Bull 11:7-16 http://www.jstor.org/stable/1493694

Winkler EM (1997) Stone in architecture: properties, durability (3rd. edition). Springer-Verlag, Berlin Heidelberg 10.1007/978-3-662-10070-7

Wood KA, Cypcar C, Hedhl L (2000) Predicting the exterior durability of new fluoropolymer coatings J Fluor Chem 104:63-71 https://doi.org/10.1016/S0022-1139(00)00228-1

Figure 1

Click here to download Figure Fig 1.JPG

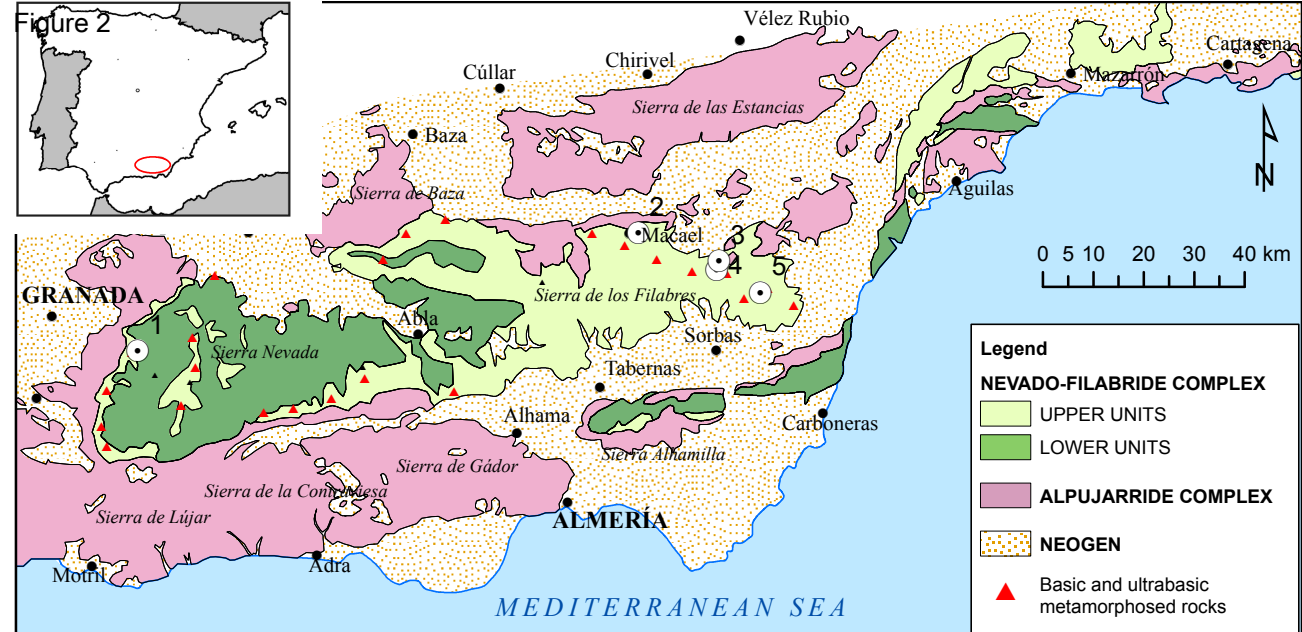
Figure 2
Vélez Rubio
Chirivel
Cúllar
Cartagena
Mazarrón
Sierra de las Estancias
Baza
Sierra de Baza
Aguilas
2
Macael
3
4
5
0 5 10 20 30 40 km
GRANADA
Sierra de los Filabres
Abla
1
Sierra Nevada
Sorbas
Tabernas
Alhama
Carboneras
Sierra Alhamilla
Sierra de Gádor
Sierra de la Contraviesa
Sierra de Lújar
ALMERÍA
Motril
Adra
MEDITERRANEAN SEA
Legend
NEVADO-FILABRIDE COMPLEX
UPPER UNITS
LOWER UNITS
ALPUJARRIDE COMPLEX
NEOGEN
Basic and ultrabasic metamorphosed rocks

Figure 3

Click here to download Figure Fig 3.tif

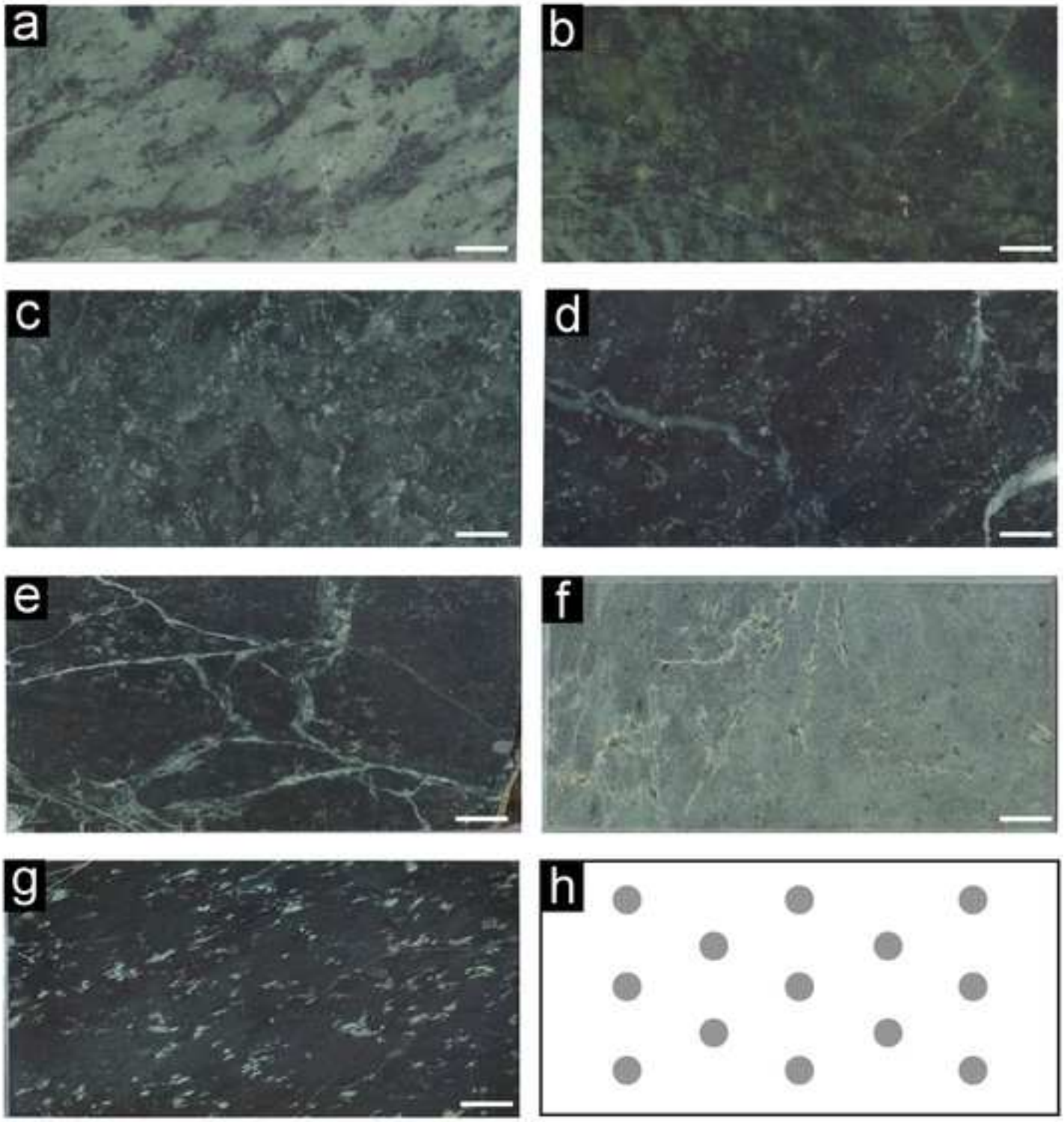

Figure 4

Click here to download Figure Fig 4.tif

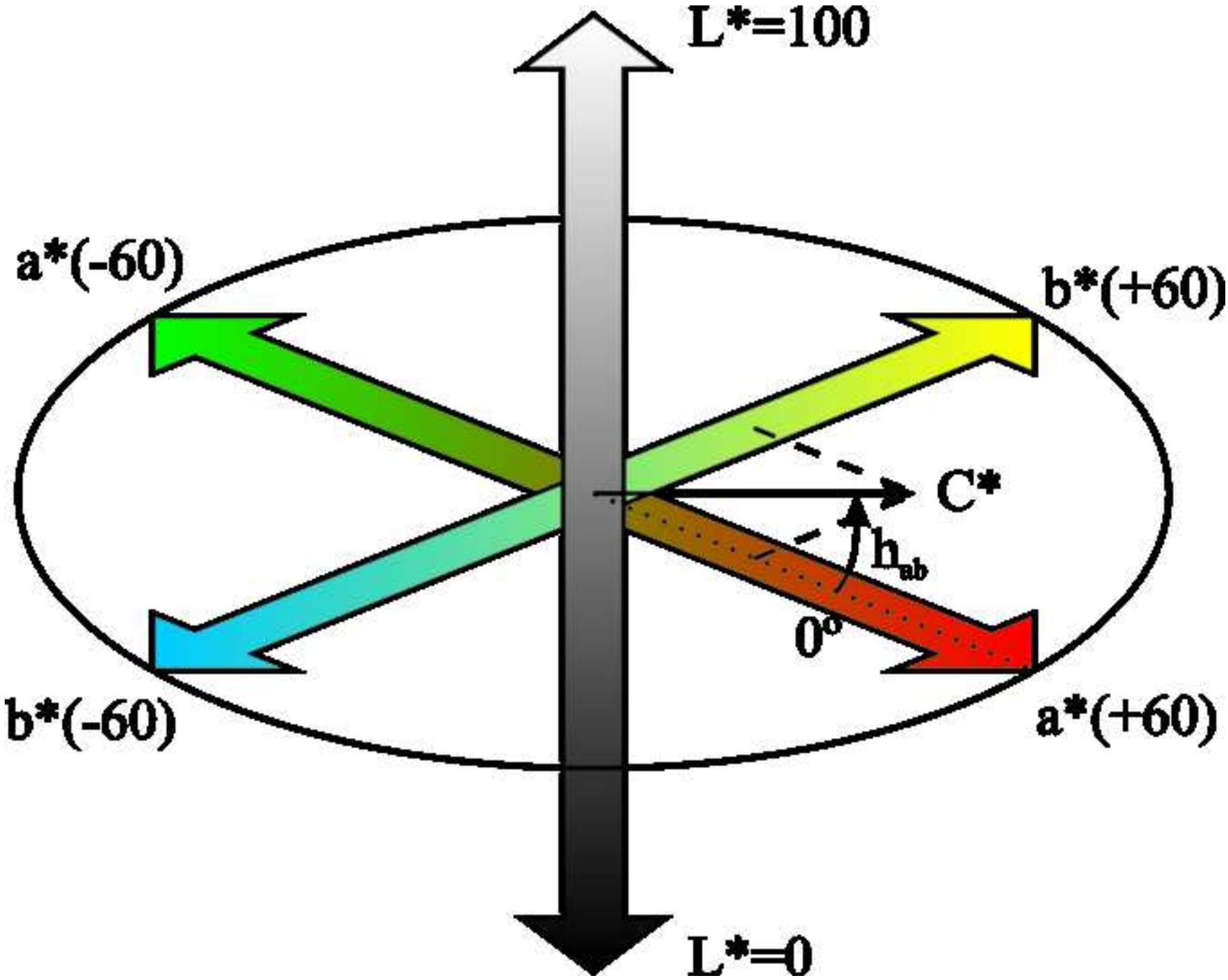

Figure 5

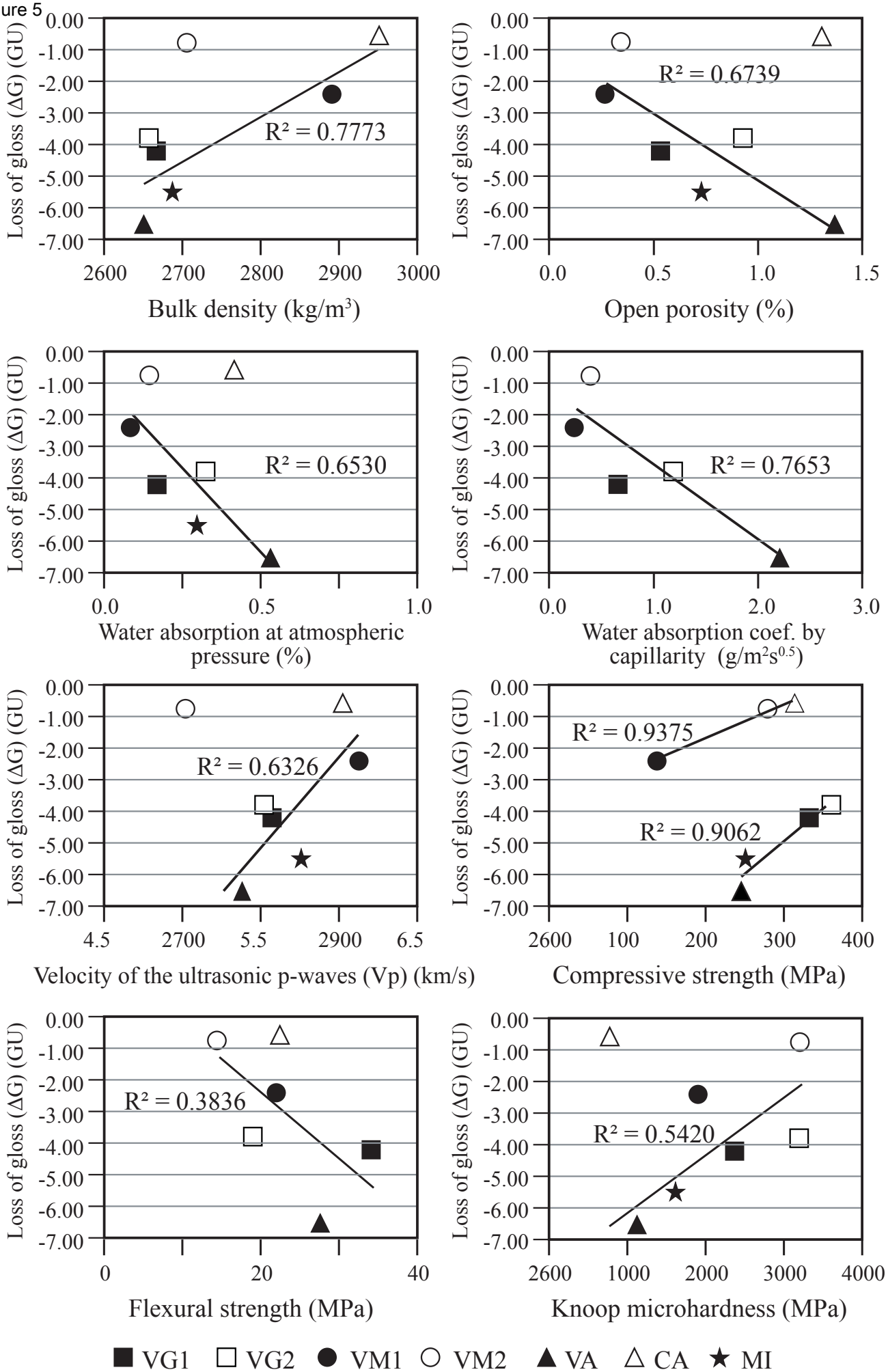
Loss of gloss (ΔG) (GU)
0.00
-1.00
-2.00
-3.00
-4.00
-5.00
-6.00
-7.00
R² = 0.7773
2600
2700
2800
2900
3000
Bulk density (kg/m³)
R² = 0.6739
0.0
0.5
1.0
1.5
Open porosity (%)
R² = 0.6530
Water absorption at atmospheric pressure (%)
R² = 0.7653
Water absorption coef. by capillarity (g/m²s^0.5)
R² = 0.6326
4.5
5.5
6.5
Velocity of the ultrasonic p-waves (Vp) (km/s)
R² = 0.9375
R² = 0.9062
100
200
300
400
Compressive strength (MPa)
R² = 0.3836
0
20
40
Flexural strength (MPa)
R² = 0.5420
1000
2000
3000
4000
Knoop microhardness (MPa)
VG1
VG2
VM1
VM2
VA
CA
MI

Table 1

Sample	Main	Accessory	Secondary
VG1[+]	Atg	Ol, Px, Am, Chl, Cb, Crt, Mag, Py, Sp, Ilm, Hem	Cb, Tlc, Trm
VG2[+]	Atg	Ol, Px, Am, Chl, Cb, Crt, Mag, Py, Sp, Ilm, Hem	Cb, Tlc, Trm
VM1[+]	Cb	Atg, Px, Amp, Mag, Py, Ttn, Sp	Cb, Hmt
VM2[+]	Atg, Cb	Ol, Px, Chl, Crt, Mag, Ilm, Py, Sp	Cb, Ilm, Hem
VA[+]	Atg	Cb, Crt, Chl, Mag, Py, Ilm, Hem, Grt	Cb, Tlc, Hem
CA[+]	Px; Amp; Ep	Atg, Crt, Chl, Mag, Ilm, Chr, Hem	Cb, Tlc, Hem
MI[x]	Atg	Am, Crt, Chl, Mag, Ilm	Cb, Tlc, Hem

Table 1: Main mineral contents of the serpentinites studied ([+]Navarro et al. 2018; [x] Navarro et al. 2013). Abbreviations: Atg: antigorite; Am: amphibole; Cb: carbonates; Chl: chlorite; Chr: chromite; Crt: chrysotile; Ep: epidote; Grt: garnet; Hem: hematites; Ilm: ilmenite; Mag: magnetite; Ol: olivine; Px: pyroxene; Py: pyrite; Sp: sphalerite; Tlc: talc; Trm: tremolite; Ttn: titanite; (Abbreviations: Siivola and Schmid 2007)

Table 2

Sample	BD (kg/m^3)	OP (%)	WAA (%)	WCC ($g/m^2s^{0.5}$)	V_p (km/s)	CS (MPa)	FS (MPa)	KM (MPa)
VG1[+]	2666 ± 27	0.53 ± 0.36	0.19 ± 0.12	0.66 ± 0.59	5.65 ± 0.48	331 ± 41	34 ± 7	2398 ± 1123
VG2[+]	2658 ± 9	0.92 ± 0.28	0.33 ± 0.07	1.17 ± 0.49	5.53 ± 0.44	361 ± 60	19 ± 9	3198 ± 1115
VM1[+]	2888 ± 59	0.27 ± 0.11	0.10 ± 0.03	0.26 ± 0.13	6.13 ± 0.43	139 ± 42	23 ± 10	1928 ± 721
VM2[+]	2704 ± 5	0.34 ± 0.07	0.16 ± 0.02	0.40 ± 0.09	6.02 ± 0.19	315 ± 43	14 ± 15	3235 ± 1432
VA[+]	2650 ± 11	1.37 ± 0.39	0.53 ± 0.11	2.19 ± 1.05	5.38 ± 0.45	246 ± 37	27 ± 12	1150 ± 319
CA[+]	2949 ± 256	1.31 ± 0.14	0.42 ± 0.08	*	5.03 ± 0.89	279 ± 84	22 ± 3	790 ± 106
MI[x]	2687 ± 17	0.73 ± 0.24	0.30 ± 0.08	*	5.76 ± 0.51	252 ± 34	*	1631 ± 583

Table 1: Main physical and mechanical parameters (mean and standard deviation) ([+]Navarro et al. 2018; [x] Navarro et al. 2013). Abbreviation: BS: Bulk density; OP: Open porosity; WAP: Water absorption at atmospheric pressure; WCC: Water absorption coefficient by capillarity; Vp: velocity of the ultrasonic p-waves; CS: Compressive strength; FS: Flexural strength; KM: Knoop microhardness. * Not tested.

Table 3

Sample	L*	a*	b*	C*	h_{ab} (º)	G^8 (GU)
VG1	39.57 ± 4.61	-6.70 ± 1.55	2.67 ± 1.58	7.21 ± 1.82	-21.75 ± 13.67	75.08 ± 7.69
VG2	23.33 ± 2.53	-3.09 ± 1.58	1.73 ± 1.60	3.54 ± 1.97	-29.33 ± 24.27	63.96 ± 6.43
VM1	27.82 ± 2.59	-5.66 ± 0.78	0.59 ± 0.50	5.69 ± 0.81	-5.94 ± 4.91	65.69 ± 5.72
VM2	22.19 ± 5.16	-3.69 ± 0.98	-0.21 ± 0.45	3.70 ± 0.97	3.27 ± 6.70	63.04 ± 8.47
VA	19.36 ± 3.45	-2.92 ± 1.01	-0.86 ± 0.55	3.04 ± 0.88	16.37 ± 13.98	62.65 ± 10.64
CA	47.55 ± 2.88	-6.11 ± 0.62	1.67 ± 1.18	6.33 ± 0.82	-15.26 ± 9.48	71.58 ± 6.84
MI	24.15 ± 3.37	-2.46 ± 0.52	-1.19 ± 0.78	2.73 ± 0.49	25.79 ± 16.56	80.08 ± 7.96

Table 3. Initial mean values and standard deviation of the main colour parameters.

Table 4

Sample	L*	a*	b*	C*	h_{ab} (º)	G^8 (GU)
VG1	38.56 ± 4.46	-6.53 ± 1.38	2.57 ± 1.52	7.02 ± 1.68	-21.47 ± 12.13	70.85 ± 9.37
VG2	23.37 ± 2.45	-2.97 ± 1.57	1.79 ± 1.55	3.47 ± 2.05	-31.12 ± 16.81	60.15 ± 5.53
VM1	26.98 ± 2.45	-5.69 ± 0.71	0.81 ± 0.42	5.75 ± 0.74	-8.12 ± 3.63	63.35 ± 4.82
VM2	22.14 ± 5.30	-3.92 ± 0.78	0.06 ± 0.35	3.92 ± 0.80	-0.83 ± 4.59	62.58 ± 5.40
VA	19.48 ± 3.16	-2.67 ± 0.93	-0.61 ± 0.50	2.74 ± 0.83	12.97 ± 13.82	56.12 ± 9.86
CA	46.55 ± 3.06	-6.01 ± 0.62	1.69 ± 1.16	6.24 ± 0.82	-15.71 ± 9.58	70.96 ± 5.83
MI	23.83 ± 3.25	-1.88 ± 0.54	-1.04 ± 0.87	2.15 ± 0.64	28.85 ± 22.42	74.58 ± 7.46

Table 4. Final mean values and standard deviation of the main colour parameters after 126 cycles.

Table 5

Sample	ΔL*	Δa*	Δb*	ΔC*	Δh	ΔG^8 (GU)	ΔE_{ab}	ΔH
VG1	-1.02	0.17	-0.10	-0.19	0.28	-4.23	1.04	0.03
VG2	0.04	0.12	0.06	-0.07	-1.79	-3.81	0.14	0.11
VM1	-0.84	-0.03	0.22	0.05	-2.18	-2.35	0.87	-0.22
VM2	-0.04	-0.22	0.27	0.22	-4.10	-0.46	0.35	-0.27
VA	0.11	0.25	0.24	-0.30	-3.40	-6.54	0.37	-0.17
CA	-1.00	0.10	0.02	-0.09	-0.44	-0.62	1.01	-0.05
MI	-0.32	0.58	0.15	-0.59	3.06	-5.50	0.68	0.13

Table 5. Variation of the mean values after 126 cycles.