Influence of the commercial finishes of ornamental granites on roughness, colour and reflectance

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Abstract

The effects of the four common commercial finishes (polished, honed, disc cutting and bush hammering) on several properties of ornamental granites were evaluated. Two granites of different texture, widely used as ornamental stones in NW Iberian Peninsula, were selected. Roughness, colour and reflectance (properties used as cleaning indicators of cultural heritage objects) of the surfaces subjected to each finish were characterized and the correlations among these parameters were obtained.

The results show that the roughness generated by disk cutting and polishing is higher in Vilachán than in Rosa Porriño, showing that the finer the grain size, the higher the roughness generated. The lightness parameter (L^*) in CIELAB space is dependent on the roughness of the surface. The rest of the colour parameters (a^*, b^*, C^*_{ab} and h_{ab}) does not depend on roughness and they are related mostly with the colour of the surface by itself. Reflectance features are not affected by the different content of forming minerals and superficial colour which condition the reflectance intensity of each commercial finish. Reflectance is directly related with the roughness of the granite; the rougher the surface the higher the reflectance intensity. In addition, the lightness L^* and the reflectance are directly related while the other colour parameters do not show any relation with the reflectance.

These results can be used as an aesthetical criteria in order to standardize an objective methodology to evaluate the cleaning effectiveness in the conservation of cultural heritage objects.

Introduction

In cultural heritage constructed with stones, the interaction between rock materials, the environment and other building materials originates the formation, among other forms of alteration, of patinas and superficial crusts [1–3]. These patinas and crusts (together with graffiti paintings as vandalism) are removed by different cleaning procedures which must be carefully controlled to preserve the characteristics of the stone. In order to evaluate the efficiency of different cleaning procedures it is important to know the characteristics of the original stone free of deposit or coatings. Then, the general effectiveness can be obtained taking into account, not only the degree of extraction, but also the modi.cations that the cleaning procedures by themselves could induce on the original surface. Among the most important parameters to be considered in the evaluation of the cleaning there are the colour and the roughness of the stone surface [2,4–7]. Recently, it has been demonstrated that the differences in reflectance spectra of cleaned granite surfaces with respect to surfaces without any kind of deposit [8–11] can be used to obtain a quantitative index of the cleaning level. reflectance measurement were also used to evaluate the bio-cleaning with different lipases of acrylic marker pen inks from unglazed ceramic substrates [12].

Granites are the main construction materials of cultural heritage in NW Iberian Peninsula. Despite their low porosity, granites have a sufficient capillary absorption coefficient to be bio-receptive [13]. For this reason, it is usual, in a temperate humid climate as that of the NW Iberian Peninsula, to find the surfaces of buildings built with granite affected by biogenic patinas (green algae and cyanobacteria – [14 and references therein]). Likewise, although less receptive to the deposition of SO_2 than carbonate rocks, sulphated black crust is also reported in granite [15,16]. So, biogenic patinas, black crusts, together with graffiti (considered as vandalism) are therefore the usual causes of the cleaning interventions in the granitic heritage of NW Iberian Peninsula. On the other hand, the low porosity and the grained texture of granites make this rock to admit different superficial finishes (e.g. polished, honed, disc cutting and bush hammering) that increase its demand in the stone market. However, it has been reported that these different finishes affect to the colour, gloss and roughness of the stone [17]. Owing that some of these parameters are used to evaluate the cleaning effectiveness, it is of great interest to study the influence of different finishes on the superficial properties of the granite,

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particularly colour, roughness and reflectance. Besides, the knowledge of the relationship between the superficial properties for each finish could provide a standardized information for characterizing the granite finishes, which will be useful in the .eld of conservation and restoration of cultural heritage. In this sense, studies focused on this topic can be found in the literature, but mainly concerned the relationship between roughness and colour of the stone [17]. However, to the best of our knowledge, there is not any research on the relation between reflectance of the stone surface and its colour or roughness.

Some researchers working on more homogeneous substrates than granite, both in composition and colour (painting mock-ups or coloured metal), found that colour becomes lighter and less saturated when the surface becomes rougher [18,19]. This effect is more remarkable for darker or more saturated colour [18]. L^* coordinate was very sensitive to surface roughness in the specular component-excluded (SCE) measurement configuration, whereas it presented almost no change in the specular componentincluded (SCI) mode [20,21]. Relationships between a^* and b^* colour parameters and roughness differed regarding the substrate; also, a^* and b^* seemed to be more sensitive against roughness changes under the SCE measurement mode [20,21]. Chroma (C^*_{ab}) is much more sensitive to changes in roughness than hue (h_{ab}) following [18,20]. In the case of granites, differences in colour, especially in the lightness parameter (L^*), were reported for the same granite with different surface finishes [17]; however, no relationship was found between the roughness and the global colour change (ΔE^*_{ab}). In the same study and in agreement with [18], changes in colour related to roughness modi.cations are higher in stones with lowest L^* values [17].

Regarding the influence of roughness and colour on reflectance, only a work centred on commercial welding mask glass was found [21]. In this work, a direct relation between roughness and reflectance was reported measuring with SCE mode; the sensitivity of reflectance to increases in roughness when measuring in the SCI mode was, however, not so high. However, there are works that relate roughness with the capacity of a surface to reflect more light in directions close to the specular than in others, i.e., gloss [22]. Therefore, agreeing with works on materials different to granite [23,24], Sanmartín et al. [17] found an inverse relationship between gloss and roughness in granite; this relationship was only found for surfaces that are not too rough ($Ra < 5\,\mu$m) because, if the roughness is very high, gloss seemed more sensitive to colour of the surface. Therefore, the authors advise the need to consider the mineral composition (which determines the colour of granite) when evaluating the relationship between roughness and reflectance. In this context, characterized by the absence of works about the relationship between reflectance and roughness in ornamental stones, it is undoubted that the roughness would generate a deviation (scattering) around the mean angle of reflectance. A surface with zero roughness would only show specular reflectance, appearing glossy. Conversely, for a surface with certain roughness, the light would be reflected in all directions and the intensity of this reflection would depend on the angle between the surface normal and the illuminant (Lambert's law) [25]. Therefore, specular reflectance depends only on superficial roughness while diffuse reflectance also depends on the crystals orientation or particles disposition on the surface [21].

In the current study, the relationship between the roughness, the colour measured in the CIELAB and CIELCH spaces and the reflectance of four different commercial finishes of two commonly used granites from NW Iberian Peninsula were performed. Roughness was characterized by means of confocal microscopy and scanning electron microscopy. The colour of each surface was measured in CIELAB and CIELCH colour spaces and the total reflectance was measured by a hyperspectral camera.

Materials and methods

Rocks and surfaces finishes

Two commercial quality ornamental granitic rocks quarried on the NW Iberian Peninsula were selected, called Vilachán and Rosa Porriño. Vilachán is a .ne-grained panallotriomorphic heterogranular granite [26]. The modal analysis obtained under petrographic microscope, following [27] is: quartz (47%), K-feldspar (10%), plagioclase (15%), biotite (7%), muscovite (18%) and mineral accessories (3%). The grain sizes of the different minerals range between 2 mm and 0.3 mm. Water accessible porosity following [28] is 2.82%. Rosa Porriño is a two-mica coarse-grained granite with a panallotriomorphic heterogranular texture [26]. The mineralogical composition is quartz (40%), K-feldspar (27%), plagioclase (14%), biotite (8%), chlorite (4%), muscovite (2%) and mineral accessories (5%) following [27]. Grain sizes range through 10 mm (K-feldspar grains), 3.8–1.2 mm (quartz grains) and 2.0–0.3 mm (biotite grains). Water accessible porosity following [28] is 0.84%.

For both granite samples, four of the most common commercial finishes were used: polished, honed, disc cutting and bush hammering (Fig. 1). For each granite and finish, one slab of 10 cm×10 cm×2 cm was used. Polishing is the finishing process by which the surface is abraded by successively finer particles in order to obtain a .atty and glossy surface. By honing, a surface very similar to polished one is obtained, but without gloss. By means of this method, coarser particle size particles than in polishing are used, in order to obtain a matte surface. Disc cutting is a finish resulting from cutting the granite with steel sheets or diamond disks; it constitutes the finish on which the rest of the surface .nishes is usually applied. The appearance of disc cutting finish is matte and rough. Bush hammering is carried out by beating the stone with a bush hammer, producing a homogeneous roughness. The hammer used was designed by a peak-valley high of 4 mm and a peak width of 8 mm.

Analytical techniques

Roughness characterization The roughness of each 10 cm×10 cm×2 cm-slab with different finishes was quantitatively characterized with a confocal microscope (PLu 2300 Sensofar®). For each sample, six extended profile of 8 mm with 10 and 2.5 objectives were performed in order to obtain the roughness parameters following [29]: R_a (average roughness), R_q (root mean square roughness), R_{pm} (average maximum peak heigh), R_{vm} (average maximum valley depth) and R_z (average maximum pro.le height).

Moreover, a qualitative characterization of the roughness was made by means of scanning electron microscopy (SEM; using a JEOL JSM-6700F and a Philips XL30) equipped with Energy Dispersive x-ray Spectroscopy (EDS), working in both modes, secondary electrons (SE) and backscattered electrons (BSE). For each sample, 2cm×2 cm-fragments were carbon coated and visualized. The optimum conditions of observation were obtained at an accelerating potential of 15–20 kV, a working distance of 9–11 mm and specimen current of 60 mA.

Colour Colorimetric analysis of each 10 cm×10 cm×2 cm-slab was accomplished by a Minolta CM-700d® spectrophotometer and a PC equipped with SpectraMagicTM NX software. Colour was expressed in the CIELAB and CIELCH spaces [20]. A total of 20 measurements were made for each surface to obtain statically representative results [30]. The measurements were made in Specular Component Included (SCI) mode with a spot diameter of 8 mm, D65 illuminant and an observer angle of 10°. The parameters measured were L^*, which is the lightness varying from black (0) to white (1 0 0); a^*, which varies from $+a^*$ (red) to $-a^*$ (green) and b^* * , which ranges from $+b^*$ (yellow) to $-b^*$ (blue). Also, C^*_{ab}, chroma or saturation or colour purity ($C^*_{ab} = \sqrt{a^{*2} + b^{*2}}$) and h_{ab}, hue, ($h_{ab} = \tan^{-1}(b^*/a^*)$) were computed. Colour of the disc cutting sur-faces was considered as a reference in order to calculate the colour differences ΔL^*, Δa^*, Δb^*, ΔC^*_{ab} and ΔH^*_{ab} and the global colour change, $\Delta E^*_{ab} = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}$). In order to detect statistical differences between the colour parameters of the superficial finishes a one-way analysis of variance and a post-hoc Tukey HSD test were performed.

Reflectance The hyperspectral imaging technique was applied in each 10 cm×10 cm×2 cm-slab using a system that integrates an imaging spectrograph with a monochrome matrix array sensor. The spectral sensor used is a Pulnix TM-1327 GE with readout mode (1392×1040) and with an objective lens (10 mm focal length). The spectrograph used is an ImSpector V10 model 2/3″ with nominal spectral range 400–1000 nm and nominal spectral resolution 4.55 nm. The system incorporates a CINEGON 1.9/10 mm COMPACT optics and a Schott DCR® III light source with cylindrical lens, producing an illuminated area of 51 mm length and 0.89 mm width. The object under analysis can be moved vertically. So that the camera scans the surface, line by line, to obtain an image at each wavelength. The ratio pixel/mm is estimated as a function of the object-camera distance and, combined with the vertical displacement of the object, allows to accurately assign a hyperspectral image to each point of the surface. The spectra data obtained was referenced to an ideal white (maximum reflectance) and corrected by the camera signal background. The 450–680 nm ranged VIS reflectance spectra was obtained for all the different surfaces.

Results and discussion

Roughness characterization

Table 1 shows the values for the roughness parameters (R_a, R_q, R_{pm}, R_{vm} and R_z) of Vilachán and Rosa Porriño granites with the four commercial finishes studied (polished, honed, disc cutting and brush hammering). Firstly, it was observed that, in both granites, the finish that showed the highest roughness values is the bush hammering. Regarding the other three finishes (polished, honed and disc cutting), slightly different results on the two granites were obtained: 1) polished-and honed-Vilachán showed similar values of roughness parameters, being lower than those of the disc cutting surface; 2) in Rosa Porriño, conversely, honed finish gave higher roughness values than polished-and disc cutting-finishes. The values of the roughness parameters of polished-and disc cuttingsurfaces were generally lower in Rosa Porriño than in Vilachán granite; in this sense, grain size could be the determining property of this difference, since it is the property that most differs between both rocks. On the contrary, the values of roughness parameters of bush hammering were higher in Rosa Porriño than in Vilachán.

R_q and R_a showed similar trends in both granites regarding the finishes but it is noteworthy that R_q parameter, which is more sensitive than R_a to the presence on the surface of localized irregularities, showed higher values than R_a in all the cases. This fact would confirm that the irregularities are not uniformly distributed along the profile. This would fit with the intrinsic textural heterogeneity of these rocks: 1) on the one hand, the resistance offered by each granite forming-mineral to the finishing process is different; 2) on the other hand, the distribution of minerals in the rock is random, both in terms of the spatial relationship of each other and the grain size.

Generally, R_{pm}, R_{vm} and R_z showed similar trends to R_a; however, on polished-, honed- and disc cutting-surfaces, R_{vm} showed higher values than R_{pm}, while for the bush hammering surface, an inverse trend was detected: R_{pm} was slightly higher than R_{vm}. The difference between R_{pm} and R_{vm} was higher in Rosa Porriño than in Vilachán. Finally, it was detected that R_z was higher in Vilachán.

Abbot-Firestone curves are useful to discuss these parameters as they allow to represent the cumulative probability density function of the surface profile's height [31]. Fig. 2 shows the Abbot-Firestone curves of the surfaces with the different commercial finishes. These curves confirmed 1) a different behaviour of the bush hammering surfaces regarding roughness comparatively to the other finishes; and 2) on Vilachán (Fig. 2A) roughness increased from polished -and honed-(similar roughness) surfaces to disc cutting surface while whereas on Rosa Porriño (Fig. 2B), roughness increased from polished-, disc cutting- and honed-surfaces.

In Fig. 2A and B, the probability density was higher for the negative z values, which indicates that there was a higher concentration of valleys (negative values of z) than of peaks (positive values of z). In Fig. 2C, the Abbot-Firestone curves of the bush hammering surfaces of both granites are shown. In general terms, their behaviour was similar; the main feature on both curves was the existence of a higher probability density for negative z values than for positive z values (i.e., the pro.le is characterized by a higher concentration of valleys than peaks). However, two subtle differences between the two granites were found: 1) the curves of Rosa Porriño have less slope at their ends, which means that the height of the peaks and the depth of the valleys are lower than in Vilachán; and 2) in Vilachán granite, the curves of the four finishes, but specially of the disc cutting and the bush hammering, extended towards more negative z values than the curves of Rosa Porriño surfaces, indicating that valleys on Vilachán surfaces are deeper than those of Rosa Porriño surfaces. This different behavior could be related to the different water open porosity of these rocks: Vilachán shows a higher porosity than Rosa Porriño, so, its mechanical strength is lower. In this sense, Vilachán granite would show a greater susceptibility to the most aggressive surface finishes, which would cause, in this granite, a greater degree of granular extraction and mineral fracture. The greater height of the peaks and depth in the valleys could reflect this higher susceptibility.

SEM observations (Fig. 3) allowed to qualitatively illustrate the quantitative characterization performed with confocal microscopy. Note that the roughness measurements were performed in wider areas (six extended profile of 8 mm) than SEM-SE observations (50x magnification). Regarding the polished finish, the surface of Rosa Porriño is smoother (Fig. 3B) than that of Vilachán (Fig. 3A) which showed more voids and .ssures. These features would be in accordance with data of Table 1 and Abbot-Firestone curves (Fig. 2A and B): polished Rosa Porriño R_a is approximately half of Vilachán polished R_a, and the curves showed deeper valleys on Vilachán than on Rosa Porriño. As was reported before, the formation of deeper valleys in Vilachán granite could be related to its lower mechanical strength derived from its higher water accessible porosity comparatively to Rosa Porriño.

The honed surface showed a similar appearance in both granites (Vilachán: Fig. 3C and Rosa Porriño: Fig. 3D) which is in agreement with the roughness parameters R_a and R_q, which were quite close to each other. In both granites, irregularities not uniformly distributed were clearly observed, in agreement with the fact that R_q values was higher than R_a.

The disc cutting finish showed a clear different appearance in both granites. Rosa Porriño surface appeared less rough (Fig. 3F) than Vilachán surface, which is in agreement with roughness parameters: R_a of disc cutting Rosa Porriño surface was approximately half of R_a of disc cutting Vilachán surface. In addition, the Abbott-Firestone curves depicted higher probability density of irregularities in the case of Vilachán (i.e., higher negative and positive z-values) than Rosa Porriño.

On the bush hammering finish surfaces (Vilachán: Fig. 3G and Rosa Porriño: Fig. 3H), SEM confirmed the clear difference of these surfaces regarding the other finishes, seen as a notable increase of peaks and valleys.

SEM-EDS also allowed to confirm different sensitivities of the minerals to the procedure of the commercial finishes (Fig. 4). On the disc cutting finish surfaces, the deep valleys were mainly associated to quartz grains which broke according to a cleavage fracture (Fig. 4A). To a lesser extent, holes were also observed affecting K-feldspar and biotite grains; the planes of rupture are clearly conditioned by the exfoliation planes of these minerals, laminar in the case of biotite (whose 001 planes appeared separated and open) and almost orthogonal in the case of K-feldspar and plagioclase.

On polished surfaces, holes also affected mainly to quartz grains; the appearance of valleys was similar to that of disc cutting finish. The rest of the minerals was apparently intact, however, that does not mean that they have not suffered stress from the mechanical process and even that they have not been broken or extracted (Vilachán: Fig. 4B and Rosa Porriño: Fig. 4C).

On honed surfaces, although the largest part of the holes is generated in the quartz (Fig. 4D, E), there are also holes in K-feldspar and plagioclase grains, but not as large or deep as in quartz. These holes are in a greater quantity than in the polished and disc cutting samples. Deformations are observed in the phyllosilicates (opening of exfoliation planes in the biotite-Fig. 4E).

On bush hammering samples (which was the finish that produced the highest roughness increase) holes in quartz, K-feldspar and plagioclase grains were observed; the shape of these holes is clearly conditioned to the exfoliation planes of each mineral (Fig. 4F and G).

Colour

Table 2 shows the colorimetric differences for each commercial finish taking as reference the colour of the disc cutting surface. For all the cases except for the colour of the polished Rosa Porriño, the colour change was mainly affected by L^*. This parameter exhibited increases for bush hammering surfaces of both granites and for honed Rosa Porriño. However, for polished surfaces of both granites and for honed Vilachán, L^* decreased. Analysing the relationship between roughness and colour, it can be seen that the rougher the surface the lighter (higher ΔL^*) the colour, as was reported by other authors in other substrates [18,19] and also on granites [17]. Regarding a^* and b^*, these coordinates increased with all the finishes on both granites except for honed Rosa Porriño. Therefore, polished-, honed-and bush hammering-Vilachán and polished-and bush hammering-Rosa Porriño surfaces showed more reddish and yellowish colourations than the disc cutting .nish. However, for honed Rosa Porriño a slight decrease on coordinate a^* was registered resulting in a decrease of the reddish colour and in an increase of the yellowish colour. No relations between the roughness and these parameters were observed. Choma (C^*_{ab}) increased in both granites after being subjected to honing, polishing and bush hammering, resulting in a more vivid colours, except in honed Vilachán; in this last, chroma showed a reduction and, in consequence, colour became greyish. C^*_{ab} suffered a higher increase in Rosa Porriño than in Vilachán, reaching values of ΔC^*_{ab} higher than 4 CIELAB units in the former. Hue also increased on the two granites after being subjected to honing, polishing and bush hammering; the highest increase occurred on polished Rosa Porriño and the lowest increase occurred on honed Rosa Porriño. Therefore, neither a^* and b^* nor chroma and hue showed a relation with the roughness; similar results were obtained in [17,21].

As indicated before, ΔE^*_{ab} were computed considering the colour of the disc cutting surfaces as the reference. The trends of the ΔE^*_{ab} were similar for all the surfaces in both granites; ΔE^*_{ab} values were lower for honed surface, followed by polished samples and the highest values of ΔE^*_{ab} were detected for the bush hammering samples. The ΔE^*_{ab} of polished surfaces were similar on both granites; conversely, the magnitude of the colour change after being subjecting to honing and bush hammering differed between granites, especially for bush hammering (DE* 5.4 CIELAB units for Vilachán and ab: ΔE^*_{ab} did not show any ab:11. 61 CIELAB units for Rosa Porriño). DE* clear relation with the roughness, but it was easy to detect that the surfaces which suffered the highest ΔE^*_{ab} were those that showed the highest increase on roughness. The visible perception of the ΔE^*_{ab} depends on the criteria applied; following [32] and [33], all the colour changes obtained for both granites after being subjected to all the finishes, except honed Rosa Porriño, would be detectable for a human eye. However, only colour changes suffering by bush hammering surfaces would be outside of the acceptable threshold in conservation of cultural heritage ($\Delta E^*_{ab} < 5$ [34]).

In Fig. 5, scatter L^*˘C^*_{ab} graphs are shown, representing the ab colour data for each granite with different commercial finishes (A: Vilachán, B: Rosa Porriño). This figure clearly depicts the direct relationship between roughness and lightness for both granites. Moreover, in the case of Vilachán a direct relationship between roughness and chroma was also found.

Reflectance

The VIS reflectance spectra (450–680 nm range) of the surfaces evaluated are shown in Fig. 6. There were no difference in the reflectance features for the studied finishes; also, the spectra features are in agreement with previous data about one of the studied granite (Rosa Porriño) with disc cutting surface [9,11]. In these spectra, six spectral signatures were identified (Fig. 6):

The first two signatures correspond to bands at the blue region (460 nm and 470 nm) in which reflectance suffered slight increases. These bands would be related with the Fe absorption: iron reflects energy in the red portion of the electromagnetic spectrum and absorbs it in the blue portion [35,36]. Harris et al. assigned these spectral features to the presence of Fe-rich minerals present in the psammites (sandstone) [36]. In Rosa Porriño and Vilachán granites, Fe is present in biotite grains (Vilachán 7% biotite and Rosa Porriño 8% biotite). It also very important to highlight that this higher content in biotite for Rosa Porriño is also reflected in the reflectance spectra, because the bands at 460 nm and 470 nm are slightly more intense for Rosa Porriño than for Vilachán.

A third signature corresponds to a steep slope between 480 nm and 620 nm (green and yellow region respectively), which is assigned by other authors to the presence of quartz in other stones, such as monzogranite and quartzite [36]. As a consequence of the steep slope, an intense reflectance peak centred at 620 nm was detected.

A fourth signature is reflected by an intense trough at 670 nm, due to the high absorption of both granites at this wavelength.

The fifth relevant feature corresponds to a very low intense trough at 675 nm that can be attributed to the presence of quartz [36].

Finally, a slight higher reflectance in the VNIR wavelengths was detected.

Harris et al. working with silicate stones with higher content in quartz assigned higher intensities, particularly in the VNIR, to the higher content in quartz; quartzite (100% quartz) and monzogranite (>50% quartz) are characterized by higher intensities than others with less quartz content, e.g. psammite (sandstone) and metatonalite (granodiorite) with quartz content <30% [36]. The same authors stated that the reflectance of high quartz-content stones throughout the whole spectrum (but especially in VNIR region) presented around 10–15% higher intensities than other stones with less content of quartz. They suggested that these higher intensities are due to the higher albedo of quartz. In the current study, Vilachán has 47% quartz and Rosa Porriño 40% quartz. Therefore, based on the results of the authors previously cited, the higher intensity of reflectance on these bands in Vilachán (a 15% higher than Rosa Porriño) could be due to its higher quartz content. This occur for all the surfaces except for the bush hammering surfaces in the blue and green regions (450–550 nm): in this region, the reflectance intensity of Vilachán is lower than in Rosa Porriño. This can be related to the greater amount of quartz in Vilachán and also to the differential behaviour of this mineral compared to the other minerals under bush hammering procedure. Quartz is the most sensitive mineral to this surface finish (as previously commented): in the SEM micrographs, many holes appear on quartz, suggesting that the quartz breaks into pieces and is extracted from the surface. Because Vilachán has more amount of quartz, the loss of this mineral will be greater in this granite, being reflected in a lower reflectance in the 450–550 nm.

For both granites, the highest value of reflectance was exhibited by bush hammering surface. The surfaces with disc cutting-, honed-and polished-finishes showed reflectance spectra very close: 1) in Vilachán (Fig. 6A) it is possible to distinguish among the four spectra (higher reflectance for disc cutting and lower reflectance for polished); 2) in Rosa Porriño, the spectra of the honed and the disc cutting are overlapped (Fig. 6B), whereas the polished surface spectrum exhibited a lower reflectance values. For both granites, polished samples exhibited the lowest reflectance values. Therefore, considering R_a values of each finish, the rougher surfaces the higher reflectance intensities. This fact is in agreement with [21], who reported that the rougher the surfaces of welding mask glasses the higher the reflectance. In this paper, the reflectance measured is the combination of specular and diffuse reflectances. The reflected light (which depends on the angle between the surface normal and the illuminant) has higher intensity in rougher surfaces than in smoother surfaces [25].As the evaluated surfaces were not specular, following Lambert´s law [25] when the surfaces became rougher, the reflectance will be dependent on the diffuse reflectance due to the roughness and also on the refractive index of the forming-minerals [17,23,24].

By summary, comparing the reflectance spectra for different finishes of the same granite, no different spectral features were observed. Nevertheless, different intensities were found due the roughness; the rougher the surface the more intense the reflectance intensity.

Comparing granites, both rocks showed the same spectral signatures, but they differed in their intensity. Vilachán granite spectra showed higher intensities than Rosa Porriño spectra, regardless the finish. This difference seems to depend more on the mineralogical composition (specifically on the percentage of quartz and ma.c minerals) than on other properties such as water accessible porosity. As previously stated, the higher water accessible porosity of

Vilachán could explain that valleys and peaks became more accused after the disc cutting and bush hammering, due to a lower mechanical strength. However, no clear relationship between porosity and R_a was found.

Correlation between roughness, colour and reflectance of granite surfaces with different finishes.

The influences of the topography of the stone surfaces in the colour and reflectance have been analysed, for each granite individually, by linear correlation techniques. The average roughness R_a was used to characterize the topography or finish of each surface. First of all, the dependence of colour parameters L^*, C^*_{ab} and h_{ab} with the surface roughness was analysed. In Fig. 7, CIELAB and CIELCH parameters L^*, C^*_{ab} and h_{ab} are represented versus R_a including a linear fit. As it can be observed the lightness L^* is correlated with R_a (correlation coefficients, $R^2 = 0.89$ for Vilachán and $R^2 = 1.0$ for Rosa Porriño). Nevertheless, neither C^*_{ab} nor h_{ab} are correlated with R_a.

Secondly, correlation between reflectance and R_a was analysed. Fig. 8 depicts the linear fit in Vilachán (Fig. 8A) and Rosa Porriño (Fig. 8B). In both cases a correlation coefficient R^2 of 0.97 was obtained indicating that the higher the roughness of the granite, the higher the reflectance.

Thirdly, the analysis of the dependence between colour parameters L^*, C^*_{ab} and h_{ab} and reflectance was studied. As it is depicted in Fig. 9, the lightness, L^*, measured in CIELAB space is correlated with the reflectance (correlation coefficient $R^2 > 0.97$ for both granites). However there is no correlation between the colour parameters C^*_{ab}, h_{ab} and the reflectance.

Conclusions

The study of the influence of the roughness due to different commercial superficial finishes of granites on their colour and reflectance revealed that:

1. The roughness generated by disk cutting and polishing is higher in Vilachán than in Rosa Porriño; taking into account the textural differences of both rocks, grain size could be the main determining property of this different behaviour. So that, for these finishes, the finer the grain size, the higher the roughness generated. However, this is not fulfiled for bush hammering; in this case, the highest roughness is generated in the coarser grained granite. The influence of the water accessible porosity on the roughness generated for the superficial finishes studied is unclear. Only after treatment of disc cutting and bush hammering, the height of peaks and the depth of the valleys are slightly higher in the more porous granite, which might reflect a lower mechanical resistance against these specific finishes.
2. Among all the minerals of the granite, quartz seems to be the mineral most sensitive to surface finishes. reflectance measurements suggest that this mineral is extracted from the surface, modifying the reflectance of the rock. Other minerals, such as phyllosilicates, seem to better adapt to the mechanical procedure, deforming through their exfoliation planes and remaining on the rock.
3. In each studied granite, a direct significant correlation between roughness (Ra) and lightness (L^*) was found; the rougher the surface the higher the L^*. However, no significant relation was stated neither between R_a and C^*_{ab} nor R_a and h_{ab}. These parameters do not change with roughness variations.
4. In each granite and finish studied, a direct significant correlation between roughness and reflectance was found: the rougher the surface the higher the reflectance.
5. Comparing granites, the differences on the roughness values and on the intensity of the reflectance spectra obtained for each finish seem to be better explained by means of the grain size and the mineralogical composition (proportion between quartz and other minerals). The influence of the water accessible porosity on these parameters seems not to have, in this case, a relevant influence.

This information should be considered as an aesthetical criterion in order to evaluate cleaning effectiveness achieved by different methods as chemical, mechanical and laser procedures in the conservation of granitic cultural heritage. In studies evaluating the effectiveness of cleaning methods, consistent conclusions regarding the harmful effects on colour and reflectance exerted by each cleaning method can be drawn only if samples with similar roughness to the rock under study are used.

Conflict of interest

None.

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References

[1] ICOMOS, Ilustrated Glossary on Stone Deterioration Patterns. Monuments and Sites XVICOMOS-ICS, 2008.

[2] C.A. Doehne, E. Price, Stone Conservation: An Overview of Current Research, Second Ed., Getty Conservation Institute (GCI), Los Angeles, USA, 2010.

[3] J.S. Pozo-Antonio, T. Rivas, A.J. López, M.P. Fiorucci, A. Ramil, Effectiveness of granite cleaning procedures in cultural heritage: a review, Sci. Total Environ. 571 (2016) 1017–1028.

[4] C.M. Grossi, F.J. Alonso, R.M. Esbert, A. Rojo, Effect of laser cleaning on granite color, Color Res. Appl. 32 (2) (2007) 152–159.

[5] J. Delgado Rodrigues, A. Grossi, Indicators and rating for the compatibility assessment of conservation actions, J. Cult. Heritage 8 (2007) 32–43, https:// doi.org/10.1016/j.culher.2006.04.007.

[6] M. Carvalhão, A. Dionísio, Evaluation of mechanical soft-abrasive blasting and chemical cleaning methods on alkyd-paint graffiti made on calcareous stones, J. Cult. Heritage 16 (4) (2015) 579–590.

[7] J.S. Pozo-Antonio, T. Rivas, M.P. Fiorucci, A.J. López, A. Ramil, Effectiveness and harmfulness evaluation of graffiti cleaning by mechanical, chemical and laser procedures on granite, Microchem. J. 125 (2016) 1–9.

[8] J.S. Pozo-Antonio, M.P. Fiorucci, A. Ramil, A.J. López, T. Rivas, Evaluation of the effectiveness of laser crust removal on granites by means of hyperspectral imaging techniques, Appl. Surface Sci. 347 (2015) 832–838.

[9] J.S. Pozo-Antonio, A. Ramil, M.P. Fiorucci, A.J. López, T. Rivas, The use of hyperspectral imaging technique to detect the most suitable graffiti-cleaning procedure, COLOR Res. Appl. 41 (3) (2016) 308–312.

[10] J.S. Pozo-Antonio, M.P. Fiorucci, T. Rivas, A.J. López, A. Ramil, D. Barral, Suitability of hyperspectral imaging technique to evaluate the effectiveness of the cleaning of a crustose lichen developed on granite, Appl. Phys. A 122 (2) (2016) 100.

[11] J.S. Pozo-Antonio, M.P. Fiorucci, T. Rivas, A. Ramil, A. López, Hyperspectral imaging as a nondestructive technique to control the laser cleaning of graffiti on granite, J. Nondestruc. Eval. 35 (3) (2016) 44.

[12] G. Germinario, I.D. van der Werf, G. Palazzo, J.L. Regidor Ros, R.M. Montes-Estelles, L. Sabbatini, Bioremoval of marker pen inks by exploiting lipase hydrolysis, Prog. Org. Coatings. 110 (2017) 162–171, https://doi.org/10.1016/j.porgcoat.2017.02.019.

[13] B. Prieto, B. Silva, Estimation of the potential bioreceptivity of granitic rocks from their intrinsic properties, Int. Biodeter. Biodegr. 56 (2005) 206–215.

[14] A.Z. Miller, P. Sanmartín, L. Pereira-Pardo, A. Dionísio, C. Saiz-Jiménez, M.F. Macedo, B. Prieto, Bioreceptivity of building stones: a review, Sci. Total Environ. 426 (2012) 1–12.

[15] T. Rivas, S. Pozo, M. Paz, Sulphur and oxygen isotope analysis to identify sources of sulphur in gypsum-rich black crusts developed on granites, Sci. Total Environ. 01 (s482–483) (2014) 137–147.

[16] J.S. Pozo-Antonio, M.F.C. Pereira, C.S.A. Rocha, Microscopic characterization of black crusts on different substrates, Sci. Total Environ. 584–585 (2016) 291– 306.

[17] P. Sanmartín, B. Silva, B. Prieto, Effect of surface finish on roughness, color and gloss of ornamental granites, J. Mater. Civ. Eng. 23 (8) (2011) 1239–1248.

[18] L. Simonot, M. Elias, Color change due to surface state modification, Color Res. Appl. 28 (2003) 45–49.

[19] M. Yonehara, T. Matsui, K. Kihara, H. Isono, A. Kijima, T. Sugibayashi, Experimental relationship between surface roughness, glossiness and color of chromatic colored metals, Mater. Trans. 45 (2004) 1027–1032.

[20] CIE S014-4/E, Colorimetry Part 4: CIE 1976 $L^*a^*b^*$ Colour Space, Commission Internationale de l'eclairage, CIE Central Bureau, Vienna, 2007.

[21] F.L.S. Cuppo, A. García-Valenzuela, J.A. Olivares, Influence of surface roughness on the diffuse to near-normal viewing reflectance factor of coatings and its consequences on color measurements, COLOR Res. Appl. 38 (3) (2013) 177–187.

[22] ASTM, Standard test method for specular gloss, D523-1995, Philadelphia. 1995.

[23] I. Ariño, U. Kleist, L. Mattsson, M. Rigdahl, On the relation between surface texture and gloss of injection-molded pigmented plastics, Polym. Eng. Sci. 45 (10) (2005) 1343–1356.

[24] V. Briones, V.J. Aguilera, C. Brown, Effect of surface topography on color and gloss of chocolate samples, J. Food Eng. 77 (4) (2006) 776–783.

[25] R.J. Lee, H.E. Smithson, Low levels of specularity support operational color constancy, particularly when surface and illumination geometry can be inferred, J. Opt. Soc. Am. A Opt. Image Sci. Vis. 33 (3) (2016) A306–A318.

[26] IGME-Mapa Geológico de España E 1:50000, Sheet 261 Tui, second edition. Servicio de Publicaciones del Ministerio de Industria y Energía, Madrid, Spain, 1981.

[27] UNE-EN 12407:2007. Natural Stone Test Methods – Petrographic Examination. AENOR.

[28] RILEM Recommandations provisoires, Essais recommandés pour mesurer l'altération des pièrres, Test n. II.1 Open Porosity, Commission 25 PEM, Protection et Erosion des Monuments, 1980.

[29] UNE-EN ISO 4288, Geometrical Product Specifications (GPS). Surface texture: Profile method, terms, definitions and surface texture parameters, 1999.

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

[31] G.W. Stachowiak, Engineering Tribology, Butterworth Heinemann, Boston, 2001.

[32] W.S. Mokrzycki, M. Tatol, Colour difference DE – A survey, Machine Graph. Vision 20 (4) (2011) 383–411.

[33] R.S. Berns, Billmeyer, Saltzman's Principles of Color Technology, third edition., Wiley, New York, 2000.

[34] O. García, K. Malaga, Definition of the procedure to determine the suitability and durability of an anti-graffiti product for application on cultural heritage porous materials, J. Cult. Heritage 13 (2012) 77–82. doi.org/10.1016/j. culher.2011.07.004.

[35] F. Sabins, Remote Sensing Principles and Interpretation, third edition., W.F. Freeman and Co., New York, N.Y., 1996.

[36] J.R. Harris, D. Rogge, R. Hitchcock, O. Ijewliw, D. Wright, Mapping lithology in Canada's Arctic: application of hyperspectral data using the minimum noise fraction transformation and matched filtering, Can. J. Earth Sci. 42 (2005) 2173–2193.

Table 1: *Roughness parameters: R_a (average roughness), R_q (root mean square roughness), R_{pm} (average maximum peak heigh), R_{vm} (average maximum valley depth), R_z (average maximum pro.le height) measured in the surfaces of Vilachán and Rosa Porriño granites with different commercial finishes.*

Surface finish	R_a (mm)	R_q (mm)	R_{pm} (mm)	R_{vm} (mm)	R_z (mm)
VILACHÁN					
Polished	5.8 ± 2.5	8.1 ± 3.5	14.8 ± 5.6	34.0 ± 14.2	48.9 ± 17.8
Honed	5.8 ± 0.5	8.3 ± 1.3	27.0 ± 18.5	33.6 ± 12.7	60.6 ± 27.5
Disc cutting	10.0 ± 1.2	13.0 ± 2.10	31.0 ± 13.6	40.5 ± 11.6	71.5 ± 13.3
Bush hammering	50.9 ± 19.8	61.1 ± 20.3	135.8 ± 28.5	129.9 ± 61.2	265.7 ± 67.5
ROSA PORRIÑO					
Polished	3.0 ± 1.1	4.2 ± 1.5	8.7 ± 2.2	23.4 ± 12.2	32.1 ± 13.7
Honed	6.2 ± 1.7	9.1 ± 3.5	14.6 ± 1.7	44.8 ± 27.9	59.4 ± 29.4
Disc cutting	4.9 ± 1.8	6.3 ± 2.1	12.9 ± 3.1	23.0 ± 6.9	35.9 ± 9.5
Bush hammering	70.5 ± 11.6	86.3 ± 12.5	269.0 ± 48.4	238.0 ± 33.8	507.0 ± 57.2

Table 2: *Colorimetric differences ΔL^*, Δa^*, Δb^*, ΔC^*_{ab} , ΔH^*_{ab} and global colour change ΔE^*_{ab} of the surfaces of Rosa Porriño and Vilachán with different finishes. The disc cutting surface data was considered as reference, so the greater the difference, the higher the colour modification. $^{\varphi}$: Significant differences (n.s. < 0.05).*

Surface finish	ΔL^*	Δa^*	Δb^*	ΔC^*_{ab}	ΔH^*_{ab}	ΔE^*_{ab}
VILACHÁN						
Polished	3.67^{φ}	0.31	1.33	0.34	1.81	3.91
Honed	2.52^{φ}	0.63^{φ}	1.92^{φ}	0.39	2.66^{φ}	3.23
Bush hammering	4.76^{φ}	0.57	2.69^{φ}	0.53	3.64^{φ}	5.49
ROSA PORRIÑO						
Polished	0.69	0.96^{φ}	3.45^{φ}	4.20^{φ}	4.09^{φ}	3.64
Honed	0.74	0.17	0.65	4.49^{φ}	0.63	1.00
Bush hammering	11.49^{φ}	0.02	1.63	6.57^{φ}	1.79	11.61

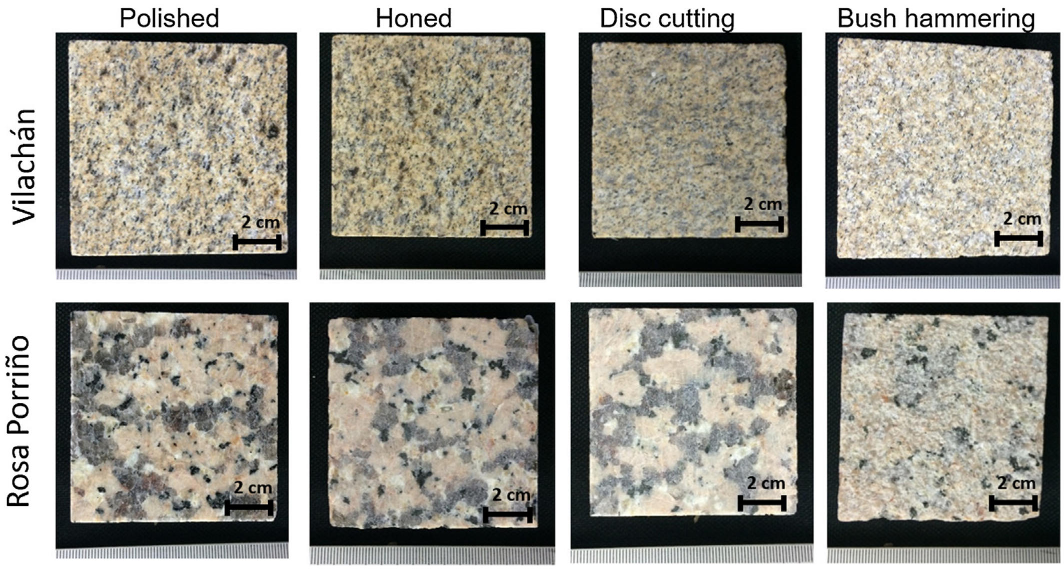

Figure 1: *Digital photographs of the granitic samples with the different commercial finishes.*

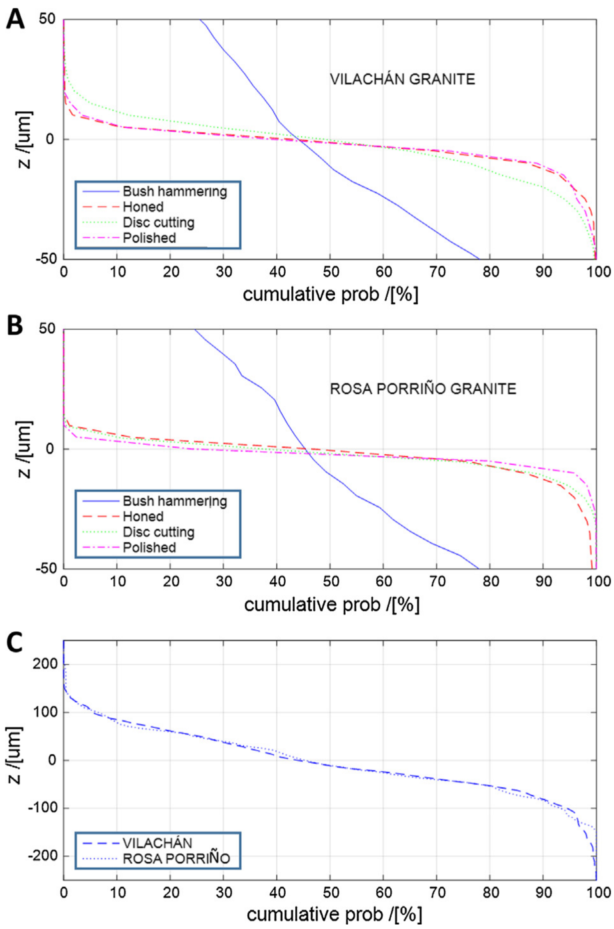

Figure 2: *Abbot-Firestone curves of data for the commercial finishes (bush hammering, honed, disc cutting and polished) of Vilachán (A) and RosaPorriño (B) granites. C shows the Abbot-Firestone curves for the bush hammering surfaces of both granites.*

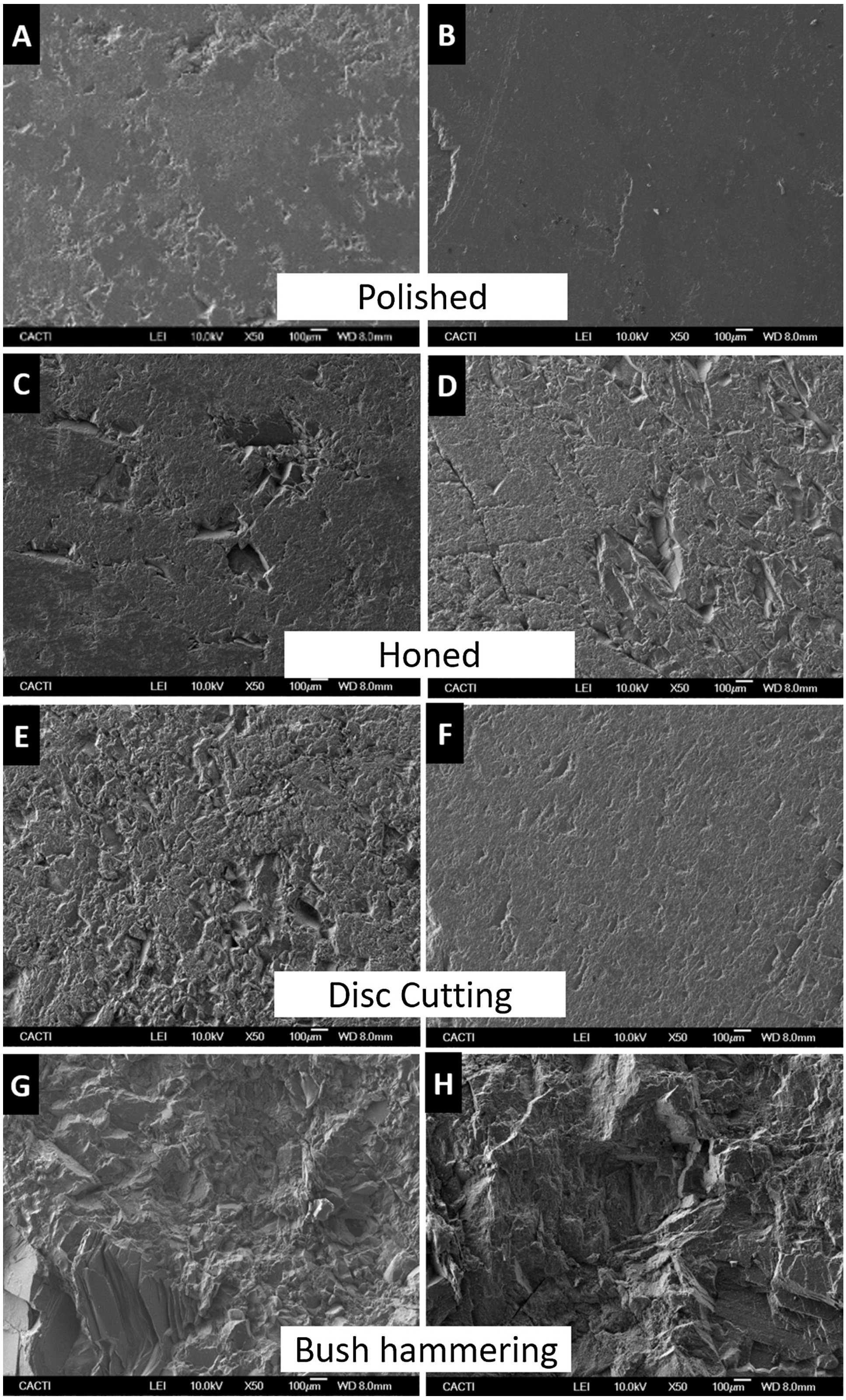

Figure 3: *SEM-SE micrographs of the surfaces of the two granites (Left: Vilachán and right: Rosa Porriño) subjected to the four commercial finishes.*

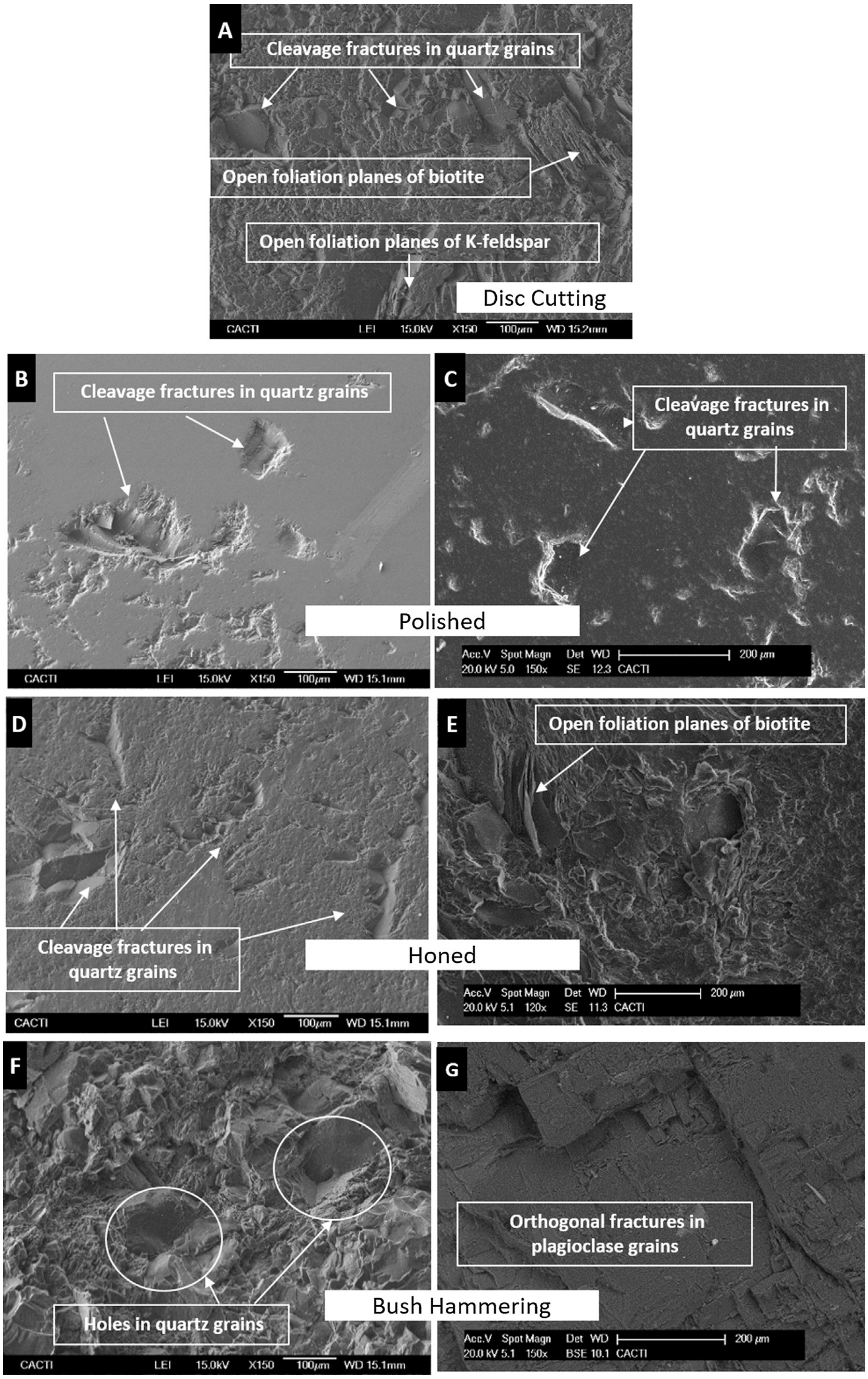

Figure 4: *A: SEM micrographs of the disc cutting surface of Vilachán. B-G: SEM micrographs of surfaces with different commercial finishes of Vilachán (left) and Rosa Porriño (right).*

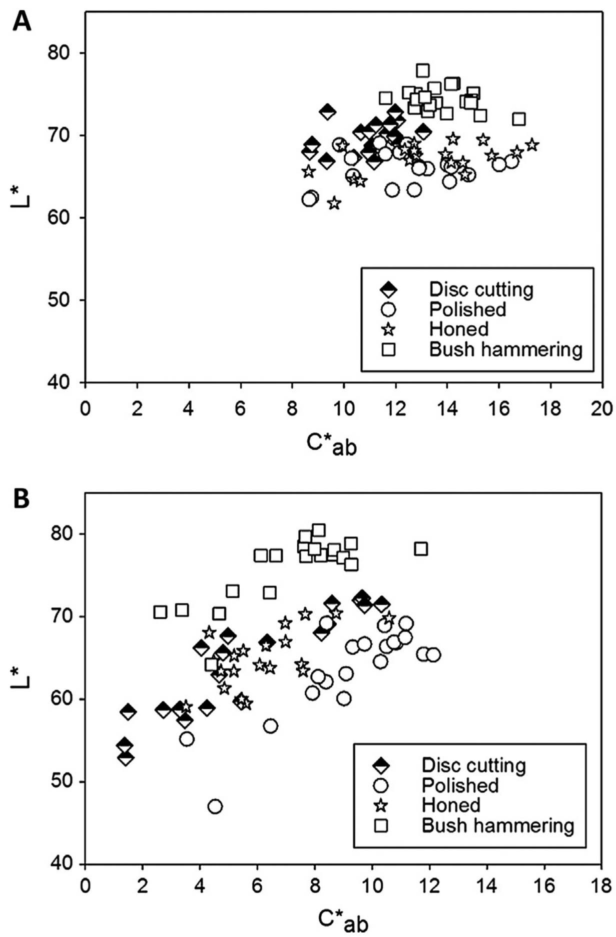

Figure 5: *Scatter $L^* - C^*_{ab}$ graphs representing colour data for the surface finishes of Vilachán (A) and Rosa Porriño (B).*

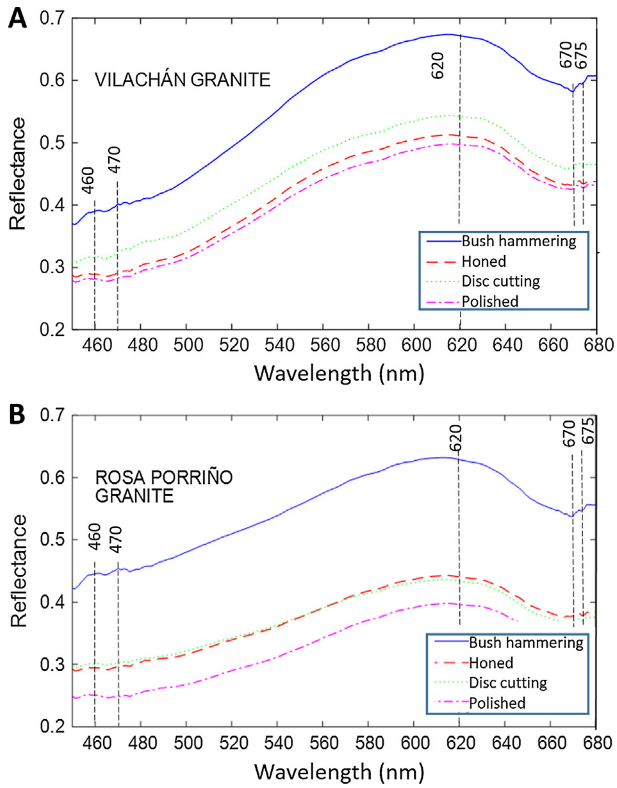

Figure 6: *Reflectance spectra obtained by hyperspectral imaging of granite surfaces with the commercial finishes (bush hammering, disc cutting, honed and polished). A: Vilachán. B: RosaPorriño.*

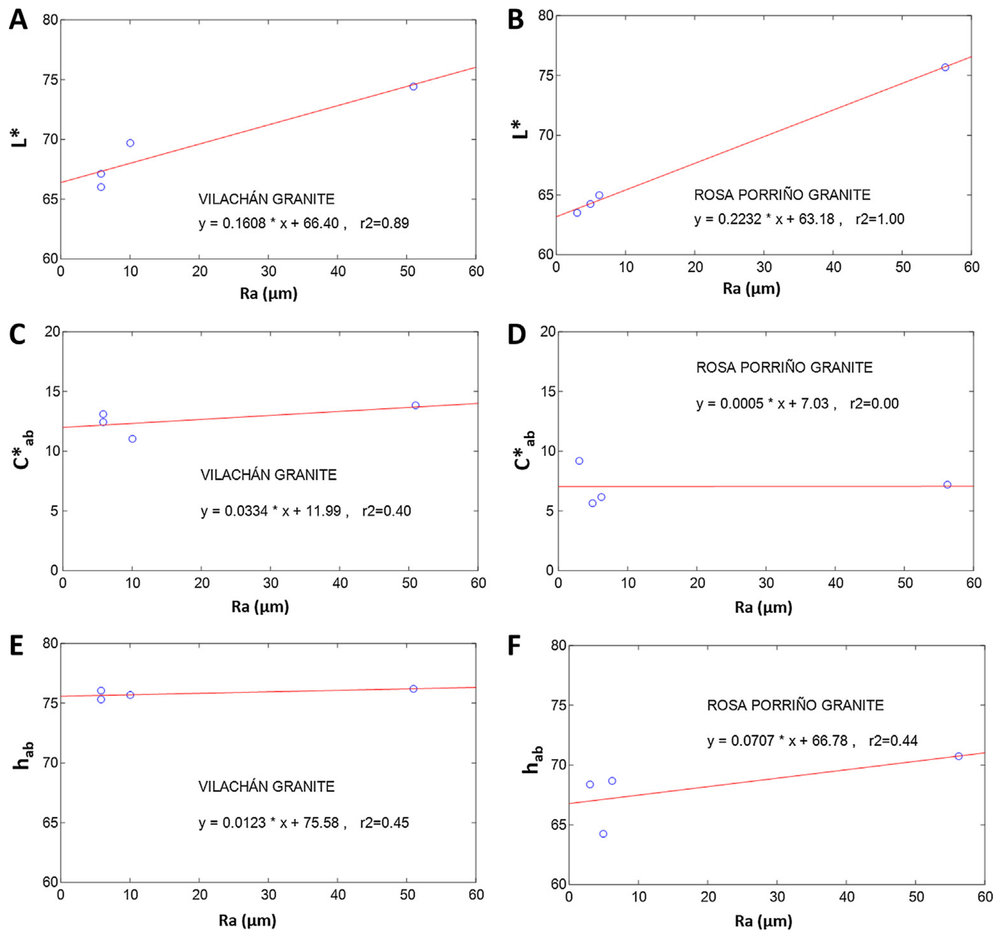

Figure 7: *Linear correlations between R_a (average roughness, mm) with the parameters of CIELAB and CIELCH spaces L^*, C^*_{ab} and h_{ab}. A, B: R_a and L^*. C, D: R_a and C^*_{ab}. E, F: R_a and h_{ab}. Left: Vilachán. Right: Rosa Porriño.*

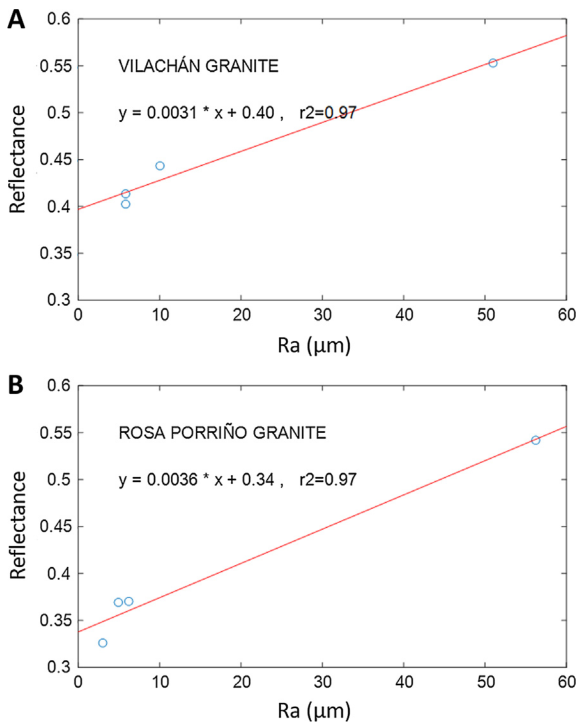

Figure 8: *Linear correlationof R_a (average roughness, µm) with the reflectance of Vilachán (A) and Rosa Porriño (B) granites with different commercial finishes. Left: Vilachánan, right: Rosa Porriño.*

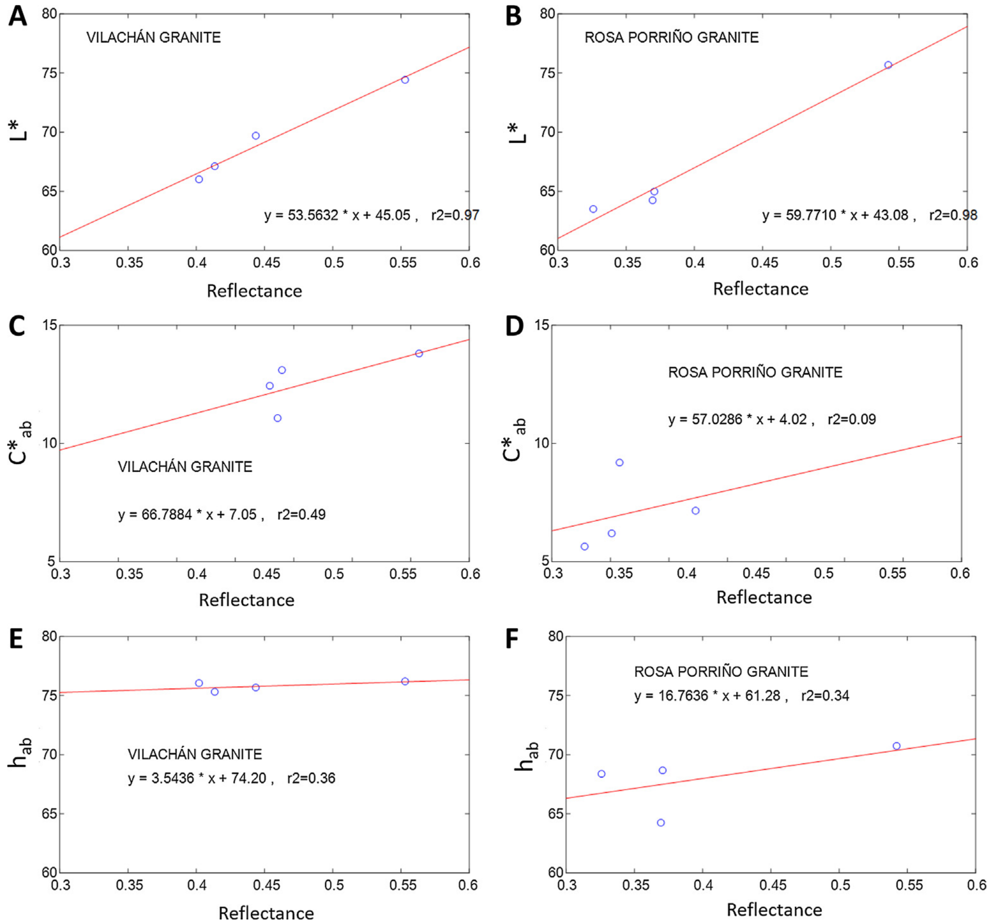

Figure 9: *Linear correlations between reflectance with the parameters of CIELAB and CIELCH spaces L^*, C^*_{ab} and h_{ab}. A, B: reflectance and L^*. C, D: reflectance and C^*_{ab}. E, F: Reflectance and h_{ab}. Left: Vilachán and right: Rosa Porriño.*