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Study on Serpentinites and the Consequence of the Misuse of Natural Stone in Buildings for Construction

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Abstract: Some rocks are very attractive as ornamental stone, but not all of them accomplish the needed requirements to be used in construction. Serpentinite can be an appealing rock, very commonly used as dimension stone. However, not all serpentinites can be used in such a way. The most widely used commercial serpentinite comes from Pakistan and India (i.e., Rajasthan Green), but other countries also produce serpentinites, such as Italy (Verde Alpi, Verde Polcevera, Verde Prato), United States (Vermont Verde Antique), and Spain (Verde Macael, Verde Pirineos). The geomechanical properties of a serpentinite are strongly related to its mineralogy (serpentine-group minerals and carbonates, mostly), and not all serpentinites are created equal. We have compared the mineralogy and mechanical properties of serpentinites that comply with the Standard Specification for Serpentinite as Dimension Stone (i.e., Rajasthan Green, Verde Macael) with those that do not (Verde Pirineos). Verde Pirineos consists of lizardite, and the carbonates, mostly dolomite, are restricted to veins. Rajasthan Green consists of antigorite, and most of the serpentine has been replaced by carbonates, dominantly magnesian calcite. Taking into account their composition and textures, rocks with such differences will evolve in different ways in a weathering environment. In replacing damaged serpentinite from a building, it is important that the new piece have the same characteristics, otherwise the replaced tiles will not look appropriate in aesthetic terms. This is particularly important in restoring monuments. Petrographic studies and x-ray diffraction are essential in analyzing the commercial potential of these rocks, and results are directly related to the geomechanical behavior of the samples. Knowledge of these characteristics would help to choose the right replacement in case of restoration of monuments and other buildings. **DOI:** **10.1061/(ASCE)MT.1943-5533.0000689.**

CE Database subject headings: Natural resources; Stones; Physical properties; Rock mechanics; Minerals; Case studies.

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Introduction

The serpentinization of rocks is a widespread process, present in most ultramafic massifs, in which the original rock changes its mineralogical composition owing to the hydration of the phases (Moody 1976; O'Hanley 1996). Serpentinization can take place through shearing and can affect the rock partially or totally. Depending on the alteration degree, the rock will have a different application in industry (Fig. 1). Serpentinites are commonly used in commercial buildings around the world in view of their attractive structures, colors, and patterns, mostly in various shades of green. There are many examples in civil construction around the world, like serpentine structures in buildings studied by Meierding (2005) in Philadelphia. However, many deterioration problems have been described on serpentinites used in exteriors (Melvin and McKenzie 1992), and a relationship has been established between this deterioration and acidic deposition due to contamination in urban areas (Meierding 2005).

Serpentinite has been used as well in monuments (Malesani et al. 2003; Marino et al. 2004). The high porosity that in some cases is present in serpentinites (up to 10% in the Appenine serpentinites: Malesani et al. 2003) can cause major problems of alteration and in durability, both indoors and outdoors, and the deterioration of the rock requires that a replacement be found. Knowledge of the composition of the rock and the provenance of the deteriorated tiles can help to preserve the exterior visual aspect of a building that has to be restored. This is also very important in the case of monuments (Šťastná et al. 2009). In any case, the mineralogical and geochemical characterization of serpentinites should be the first step in a planned restoration of a deteriorated part of a new or a historical building. It has been demonstrated that mineral transformations affecting the serpentinites, such as replacement by carbonate and/or talc, can derive in changes of physical and mechanical characteristics (Ismael and Hassan 2008). However, very little attention has been given to these rocks in the international literature regarding their proper behavior as construction rocks (Navarro 2011).

In this work, we present the results of our observations on various serpentinites. Traditionally, these rocks have been regarded as marbles, and we consider here the treatment that companies have done to these misinterpreted rocks. We analyze the evolution of serpentinites with weathering, and from the results of this analysis we suggest a proposal for a better understanding of their behavior as building material. From the conclusions, we advise the complete knowledge of a natural stone before presenting it for construction use, both for new building and for restoration.

Cases in Our Study

In this study, we have compared serpentinites from various ultramafic massifs that are or have been quarried for use as dimension

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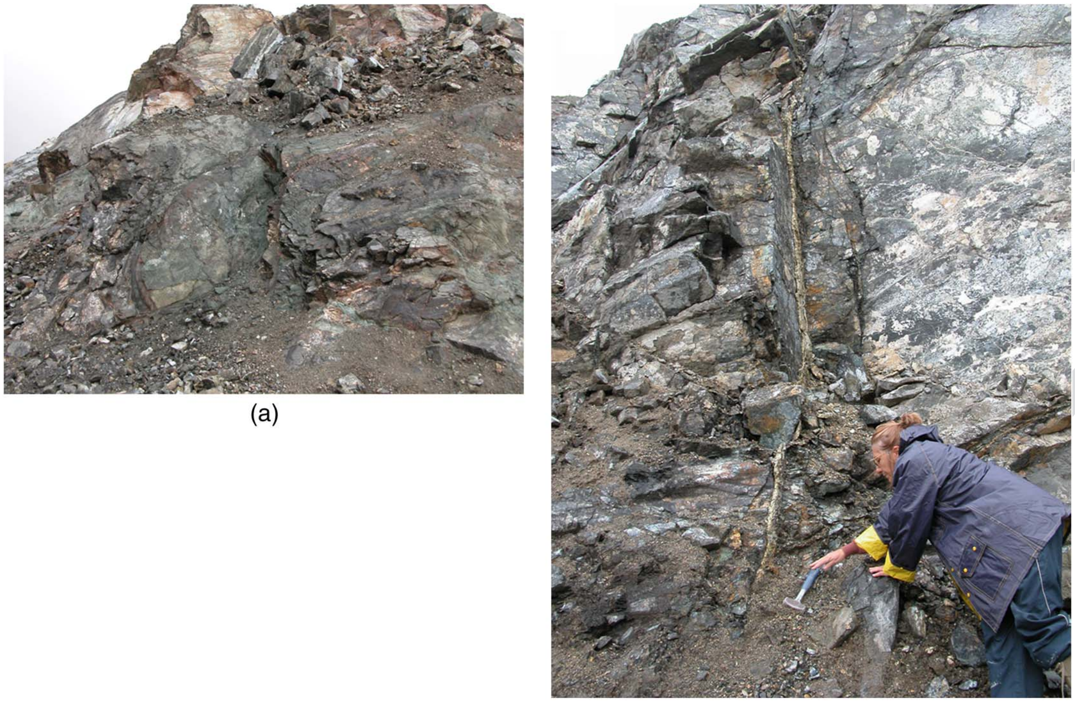
(a)

(b)

Fig. 1. Evolution of an ultramafic rock (a) from the fresh outcrop; (b) to the serpentinized outcrop cut by a shearing system (pictures are from a dunite quarry for refractory concrete in Cabo Ortegal)

stone. Serpentinites from Cabo Ortegal massif, northwest Spain (Pereira et al. 2004, 2008) were quarried in the municipality of Moeche and commercialized under the marketable name of Verde Pirineos. This quarry was closed down some years ago, probably because of undesirable characteristics of this rock where used in construction (see later). Most serpentinites used as dimension stone in Spain come from Pakistan, India (i.e., the Rajasthan massif), and Guatemala (Verde Guatemala). They are sold under various commercial names, depending on the color or structures (for example, Forest, Ocean, Grace, Jaim, Emerald, Seagreen), but most of them are found in catalogues under the "marble" section, called in general green marble. We understand that the reason for this denomination comes from the high degree of transformation of the minerals of these rocks to carbonates; most of the serpentine is now carbonate, and, from a petrographic point of view, they could well be classified as carbonated rocks. However, some remnants of the original assemblage of minerals can be found (i.e., olivine, pyroxene, spinel-group minerals), and some mineral transformations can differ from the described ones. The evolution of the original minerals with weathering can be very different, either to serpentine, to talc, and/or to carbonates. It is a striking feature that even where serpentinites are pervasively transformed into carbonates (or to talc), they maintain their appearance in color and structures, adding more confusion to their classification.

Methods of Study

The main tools used in our study of the serpentinites are the optical microscope and X-ray diffractometry. Geochemical analysis is an excellent method to characterize the rocks and could be used to identify their origin (Pereira et al. 2010). We have studied 39 thin sections of rocks from Cabo Ortegal (including fresh, serpentinized, and carbonate-serpentinized samples) and six thin sections of the Indian Green marble. The latter are samples from the International Natural Stone Fair that takes place in Madrid (Spain) every two years. Several of those samples from Cabo Ortegal and from the Indian Green marble have been analyzed by x-ray diffraction. In Tables 1 and 2, we present results for that selection, representing the complete set.

We have tested the samples from Cabo Ortegal to get the physical and mechanical characteristics of the rocks as a function of various degrees of serpentinization. The values obtained have been compared to those published elsewhere (Pereira et al. 2005). Samples were tested for the Absorption and Bulk Specific Gravity (ASTM C97) to correlate this value with the degree of serpentinization of the rock and for compressive strength (ASTM C170) to determine if these rocks achieve the minimum requirements in terms of resistance. We used the ASTM norms because these refer specifically to serpentinites instead of being these rocks included within the marble requirements section. Values for the

Table 1. Mineralogical Composition (%), Density (δ, g/cm^3), and Compressive Strength (σ_1, MPa) Values for 13 Serpentinite Samples from Galicia (Spain)

Sample	δ	σ_1	Lizardite	Talc	Magnesite	Dolomite
Mo-1	2.62	119.6	88.67	0	0	11.33
Mo-2	2.76	50.5	47.62	9.78	39.55	3.04
Mo-3	2.66	24.2	49.33	7.60	40.97	2.10
Mo-4	2.70	59.6	78.28	5.56	16.16	0
Mo-5	2.70	59.6	79.38	3.76	16.85	0
Mo-6	2.79	68.2	52.97	9.32	37.71	0
Mo-7	2.76	59.6	50.51	25.28	24.20	0
Mo-8	2.83	52.2	61.79	8.96	29.26	0
Mo-9	2.84	87.4	70.94	6.05	23.01	0
Mo-10	2.82	70.0	78.86	6.73	14.42	0
Mo-11	2.73	58.5	83.69	3.97	12.34	0
Mo-12	2.78	35.8	68.34	6.48	25.19	0
Mo-13	2.60	35.1	100	0	0	0

Table 2. Physical and Mechanical Properties of Indian Green Marbles and Samples from Cabo Ortegal, Compared to the ASTM Requirements (Data from Pereira et al. 2007)

		Absorption (%) Ext./Int.	Density (kg/m^3)	Compressive strength (MPa)
ASTM requirements		0.20 (max)/ 0.60 (max)	2.56 (min)	69 (min)
Indian Green marbles[a]		<0.50	2.55–2.70	70–140
Selection of samples from Cabo Ortegal (Spain)	Sample 1	0.09	2.94	78.22
	Sample 2	13.06	1.69	1.91
	Sample 3a	0.25	2.76	50.50
	Sample 3b	0.13	2.66	24.20
	Sample 4	0.09	2.50	84.10

[a] Indian Green marble data from http://www.naturalstoneimpressions.com/marble.html.

Indian Green marble come from the web pages of the commercial companies.

Discussion

When ultramafic rocks reach a high percentage of water, we say that these rocks are weathered. Weathering can be due to surface exposition of the rock to meteoric fluids or fluids coming from other origins, and the process can take place in the outcrop or once the rock has been emplaced for building. Our observations of the evolution of serpentinites used in buildings as a result of the weathering have taken us to the study of the behavior of these rocks as dimension stone. It is surprising that, being these rocks widely used and with such bad demonstrated behavior when used in exteriors (Meierding 2005), there is little information about the physical and mechanical characteristics in the international literature. We conclude from our study that the differences in mechanical response are strongly dependent on the mineralogy and structures of the rock. Verde Pirineos is a rock made up mainly of serpentine, cut by few veins of the fibrous serpentine variety (chrysotile) and carbonates, and many times severely replaced by talc (Table 1). Most of the original mineralogy (forsterite, enstatite, augite, metamorphic tremolite) has been largely transformed to lizardite, and the latter is transformed in part to magnesian carbonates (Figs. 2 and 3, Table 1).

The Green Marble product from India is made up mostly of a carbonate (magnesian calcite), with some remnants of amphibole (tremolite) and serpentine. However, in the case of Indian serpentinites, the serpentine-group mineral is antigorite rather than lizardite (Fig. 4).

From our study, it seems that those serpentinites consisting of antigorite are more likely to be replaced by carbonates; those made by lizardite as the main serpentine phase are likely to remain with that same mineralogy or replaced by talc. In the latter case, carbonates are found only in shears and veins cutting the rock. Serpentinites that are dominantly made of lizardite (i.e., Verde Pirineos) are more fragile, are susceptible to low values for compressive strength, and do not achieve the requirements for use as dimension stone in most of their uses. Serpentinites transformed to carbonates (i.e., Green Marble in all different commercial names) seem to be more resistant to further alteration and do satisfy the specifications. It could be concluded that carbonate minerals act as a perfect cement to maintain the other components together, unlike the samples that have not suffered a carbonation process.

We have used the statistical parameter R (Pearson's correlation coefficient) to measure the intensity of the relationship between mineralogy and strength of the rock. Correlation between carbonates and compressive strength in the case of Verde Pirineos is negative ($R = -0.66$ in the case of magnesite and $R = -79$ in the case of dolomite; Table 1). Correlation between serpentine (lizardite) and carbonates (magnesite) is negative as well ($R = -0.94$).

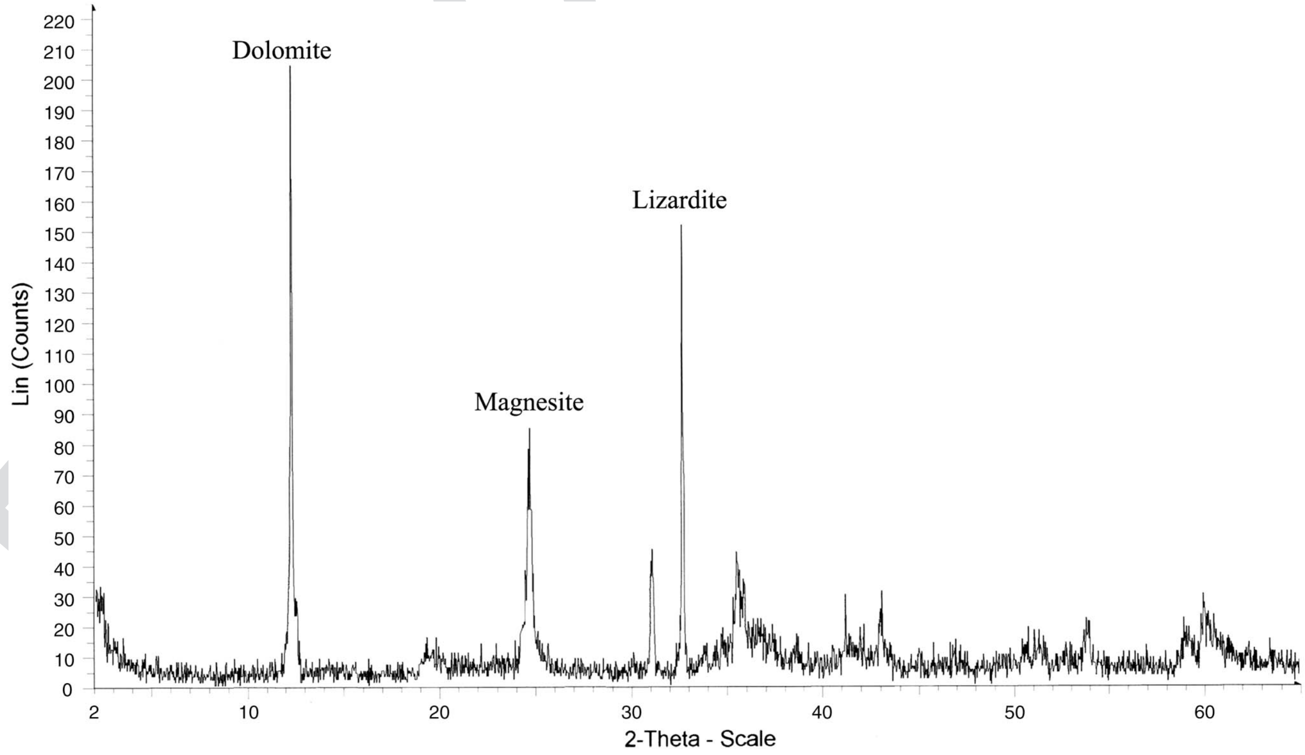

Fig. 2. X-ray difractogram of a carbonated serpentinite from Cabo Ortegal where Verde Pirineos used to be mined (Pereira et al. 2008)

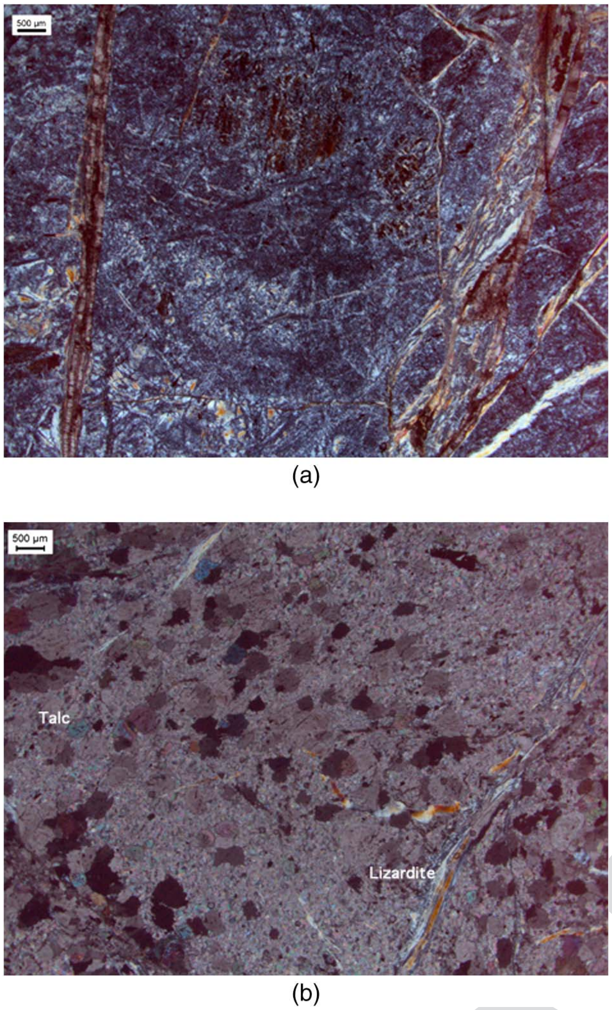

Fig. 3. Microphotographs of (a) serpentinite from Galicia (Mo-1, Table 1); (b) carbonatized serpentinite from Galicia (Mo-2, Table 1)

The more magnesite we find in the rock, the less serpentine remains untransformed, which means that the magnesite is replacing the serpentine, while in the case of the Indian serpentinites, the carbonate (magnesian calcite) is found filling porosities and acting as cement. In this last case, carbonate has a different correlation with serpentine, as there is no replacement of one for the other and carbonates are added to the whole serpentine composition. Carbonates in this case are derived from direct precipitation.

But even where these rocks achieve the normative specified values, they can give some problems once they are placed on a building. The degree of serpentinization of the rocks directly influences the physical (absorption) and mechanical (compressive strength) properties. Samples with the highest degree of alteration (e.g., sample 2 in Table 2) give absorption values below the normative requirements and a very low compressive strength (Pereira et al. 2007). Sample 3a in Table 2 does not present evidence of a high degree of alteration and gives absorption values of 0.25%, which allows its use in interiors, but not exteriors. The compressive strength value is clearly below the requirements. Samples 1b and 4 in Table 2 present very low absorption percentages and very high values of compressive strength, consistently within the requirements; however, these last two samples cannot be classified as serpentinites, as they do not have a sufficient degree of hydration. Moreover, they preserve most of the original minerals (olivine, enstatite, spinel-group minerals, and metamorphic tremolite), together with some serpentine.

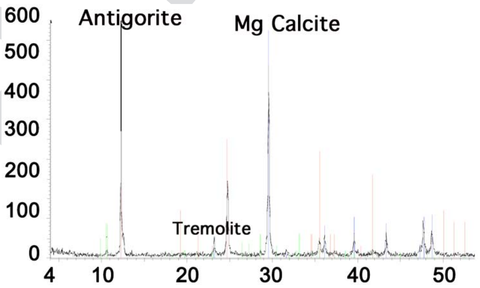

Fig. 4. X-ray difractogram of Indian Green Marble, the sample of which comes from the Stone International Fair in Madrid

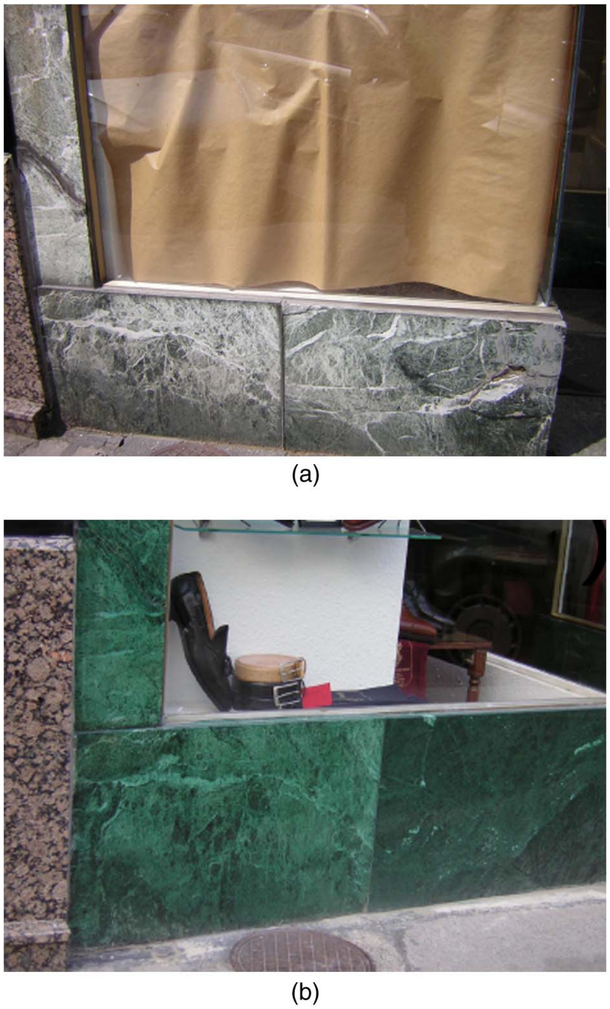

Fig. 5. Exterior walls of a commercial building in Vienna showing major differences in aspect: (a) the deterioration of one of the tiles; (b) the new tile used in the restoration

We have investigated the consequences of serpentinite weathering in commercial and historical buildings. Fig. 5 shows aspects of the exterior wall of a shop in Vienna, before and after the replacement of a tile. One of the original lower tiles was broken and, when restoring the assembly of the façade, builders replaced this tile by a new one. However, the appearance of this new tile is very different from the previous one in color and in the rock structures. Although a covering product was used to disguise the differences, these can be noticed easily; the new tile has a darker color and a total different aspect. This outcome is not a severe problem in a commercial building (Pereira et al. 2010), but it is an unacceptable result if we are dealing with a historical building (Malesani et al. 2003; Marino et al. 2004). We will address this issue in a separate paper.

Conclusions

To prevent unexpected results in serpentinites used as dimension stone, we recommend knowing their complete characterization regarding mineralogy (X-ray diffraction, petrography) and physical and mechanical specifications. Unfortunately, the technical information that the suppliers provide in most instances is quite inadequate. For some rocks, they claim that the stone has the hardness of most granites (just over 6 on the Mohs scale of hardness) when their constituent minerals are related to talc (hardness 1) and chlorite (hardness around 2 1/2). Clearly, the suppliers of this stone need to acquire a decent study of petrography, to be included with the rest of the information.

Evolution and weathering of rocks in the outcrop can serve as a natural analogue for the evolution of a natural rock once emplaced in a construction site. Knowledge of the provenance of the rocks from which the tiles were made will assure a predicted pattern of their evolution through weathering, avoiding unwanted contrasts in appearance. This is important in commercial buildings but mandatory in restoration of monuments. For this last reason, protecting historical quarries from overexploitation has become an important issue (Fort et al. 2006).

From our study, we can conclude that there is very little international literature on this issue, and we propose to promote research on methods of building with serpentinites. The existence of veins and brittle fractures in serpentinites, a very common feature, can lead to the disintegration of a tile. Moreover, there is the high porosity of some of these rocks, which can trigger their breakdown. Applying reinforcement or a method of consolidation could solve both problems. There is already research along this line (Proudfoot et al. 1988) as well as regarding consolidation experiments using resins leading to a diminution of porosity and providing an enhanced resistance (Selwitz 1992). More investigations along these research lines are needed because the conclusions of misbehavior of serpentinites can be applied to many other natural stones, requesting their study in detail before using them in civil construction or restoration work.

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