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Development of a Solar-Reflective Ceramic Tile Ready for Industrialization

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Abstract

Solar-reflective surfaces represent an effective countermeasure to UHI. The market of "cool" materials is dominated by polymeric solutions which, under UV exposure, are damaged. On the other hand, an increasing attention was paid recently to ceramic-based solar-reflective surfaces, characterized by very long lifespan. A ceramic tile is typically made by a three layers structure: substrate-engobe-glaze. This structure has been exploited to develop a cool ceramic tile that can be produced in the same production facilities of common products to create a whole tile by merging technological results and industrial production needs, to achieve a compromise between performance and costs.

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Keywords: cool tile; solar reflective tile; solar reflectance; lightness; glaze; traditional ceramics

1. Introduction

In the last decades the temperature of urban areas increased markedly, worsening the difference in temperature with respect to the surrounding rural areas [1]. This well-known phenomenon, identified as the urban heat island (UHI) effect, is the result of several causes, some of them controllable, such as urban design, anthropogenic heat, air pollutants, and other ones uncontrollable, among these sun intensity, wind speed and anticyclone conditions. Moreover, the UHI effect is related to the increase of greenhouse gases ad CO_2 emissions, as well as to energy demand for air conditioning [2]. Finding effective solutions to UHI is one among the most urgent needs, not only for human comfort but also to preserve the environment from climate change [3]. One among the most effective solutions is increasing the passive cooling capabilities of the building by increasing its external albedo [4]. This can be achieved through green surfaces (green roof, green facades) which, however, show high installation and operation costs [5,6], or through solar reflective materials such as "cool roofs", which are characterized by the aptitude to reject solar

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radiation across both the visible and invisible portions of the solar spectrum [7–9]. Those materials are characterized by high solar reflectance (300-2500 nm) and high thermal emissivity (4000-40000 nm). The first generations of cool roofs were characterized by white color [10]. This made difficult to integrate cool materials in urban contests where roofs are visible from the streets, so new cool roof generations were implemented with the addition of pigments to create the so-called "cool colors" [11–14]. Those materials are generally characterized by a solar reflectance higher than other products with the same visual response, thanks to an increase of solar reflectance in the NIR spectral range. The first studies on both white and colored cool roof categories were carried out on polymeric materials (membranes and paints), which were easier to prepare since they do not require long industrial cycles and preparation. However, these materials are generally characterized by a relatively low durability against time: if a correct maintenance is not scheduled and performed properly, the lifetime of a cool roof is limited to few years [15]. The diffusion of cool roofs also to urban contexts where ceramic tiles and clay roof tiles are widely spread (i.e. European cities) is subordinated to the development of solar reflective materials which can be integrated on tiles. This ensures not only good solar performances but also good durability against time, since on a sloped roof, typically covered by tiles, cleaning operations are not recommended [16]. The development of new glazed and engobed solar reflective products is trying to help the diffusion of this technology even on ceramic tiles and clay roof tiles [17,18]. Such products, even if characterized by higher production costs, are also characterized by excellent durability against time and, in case of ceramic glaze, also by the capability to resist to soiling due to weathering and fouling [19]. Moreover a further implementation can be made on traditional tile systems to enhance not only solar reflectance but even insulation properties without changing in a substantial way the roofing setup [2]. The aim of this work is provide an overview of the most used pigments for external tile systems and analyze their behavior once embedded in ceramic glazes with different finishing (gloss, matt, with or without Zirconium Silicate), and subjected to traditional ceramics heating treatment. This will allow to provide a starting point for the production of a first generation of glazed cool colored tiles. This study was performed in collaboration with Sicer s.r.l. of Italy, which provided the industrial kiln for samples firing and all the raw materials used.

2. Materials and Methods

2.1. Sample Preparation

As suggested by Levinson et al. [12], in case of a NIR-absorbing substrate, it is necessary to create a two-coat system to obtain a cool roof product: the lower coat is represented by a NIR-reflective basecoat, the upper one is a cool topcoat. This layout matches perfectly with the typical structure of a glazed ceramic tile where the lower layer is represented by the engobe, whereas the upper layer is represented by a ceramic glaze. In this case a zirconium silicate based engobe was used (B_2O_3 3 wt%, Na_2O 3 wt%, Al_2O_3 19 wt%, SiO_2 64 wt%, K_2O 1 wt%, CaO 1 wt%, ZnO 1 wt%, ZrO_2 7 wt%), which ensures opacity and a good degree of solar reflectance. Previous studies formulated a more performing engobe [17] but the high percentage of zirconium silicate present in that formulation doesn't match with the need of the ceramic industry to keep the final production costs affordable. The glazes were prepared, starting from a gloss frit (Al_2O_3 9.27 wt%, SiO_2 61.97 wt%, K_2O 3.93 wt%, CaO 14.81 wt%, ZnO 10.02 wt%), a matt frit (Na_2O 5.45 wt%, MgO 2.90 wt%, Al_2O_3 21.29 wt%, SiO_2 56.66 wt%, K_2O 4.71 wt%, CaO 8.99 wt%,), zirconium silicate ($ZrSiO_4$ 100 wt%) and pigments, according to formulations reported in Table 1.

Table 1. Glazes composition (wt%).

	G3	G6	M3	M6	G3Z	G6Z	M3Z	M6Z
Gloss frit	100	100			85	85		
Matt frit			100	100			85	85
ZrSiO4					15	15	15	15
Pigment	3	6	3	6	3	6	3	6
Carboxymethyl cellulose	0.2	0.2	0.2	0.2	0.2	0.2		

14 different pigments were selected, divided into four family colors according to commercial classification. Pigments are identified with a letter (B for black, R for red, G for green and Y for yellow) and a progressive number. Chemical compositions of the four families are reported in Table 2. Resulting glazes were labelled merging the glaze and the pigment codes (i.e. G3_B1: gloss glaze with a 3 wt% of black pigment B1). All the components were poured in a porcelain jar with 50 wt% of water and sintered alumina grinding media with different diameters. After the milling process, a 300 μm glaze layer was applied onto engobed tiles, then dried for 24 hours at 110°C and eventually fired in an industrial roller kiln at the maximum temperature of 1200°C for 15 min in a cold to cold cycle which totally lasts 40 min. All the characterizations were performed on the surface of the fired samples.

Table 2. Pigments chemical composition.

	B1	B2	B3	B4	R1	R2	R3	R4	G1	G2	G3	Y1	Y2	Y3
Na_2O		1.54												
MgO					1.39									
Al_2O_3	14.78			1.86		5.19	8.02	2.49	14.66	7.19				11.39
SiO_2	21.32	4.27	36.91	23.69	24.24	22.17	16.06	10.69		26.05		38.42	18.21	26.99
K_2O	0.55													
CaO	1.36													
TiO_2					1.66							1.64	74.01	
Cr_2O_3	24.43	38.65	21.39	23.51		14.07	26.08	33.15	85.35	45.51	100.00		1.92	8.01
MnO	2.83		6.58	12.77				19.50						
FeO	15.77	27.16	17.06	18.40	24.95	14.49	17.08	28.46						10.13
CoO	6.05	28.26	11.49	20.60						21.24				
NiO	11.83		6.59	17.29										
ZnO	1.09	3.54				39.22	32.77	7.63						43.48
ZrO_2					47.76	4.85						55.39		
Sb_2O_3													5.86	
Pr_2O_3												4.56		

2.2. Samples characterization

Spectral reflectance of each sample was measured using a UV-Vis-NIR spectrophotometer (Jasco V-670) with a 150 mm integrating sphere, in accomplishment to ASTM E90 [20]. The solar reflectance value ρ_{sol} of every surface was calculated by integrating over the range from 300 to 2500 nm the measured spectral reflectivity $\rho\lambda$ (defined as the ratio of reflected part and total amount of incident radiation at the considered wavelength λ), weighted by the standard spectral irradiance of the sun at the earth surface, $I_{sol,\lambda}$ [Wm^{-2} nm^{-1}] described by the AM1GH irradiance spectrum [21,22] (1):

$$\rho_{sol} = \frac{\int_{300}^{2500} \rho_\lambda I_{sol,\lambda} d\lambda}{\int_{300}^{2500} I_{sol,\lambda} d\lambda} \quad (1)$$

L*a*b* measurements were also performed using an X-Rite SP-60 colorimeter on the final samples to quantify sample colors in an objective way. A particular attention is paid to the L* parameter through correlation between solar reflectance and L* itself.

3. Results and Discussion

Starting from 8 different ceramic glazes and 14 pigments, 112 different samples were produced. Due to the high number of samples involved, all the four color families will be treated separately to facilitate the discussion. Fig. 1 shows the spectral reflectivity of all the produced samples. Samples without the addition of zirconium silicate are reported on the first and third column while samples characterized by the addition of zirconium silicate are on the second and fourth column. The first line summarizes the black samples, then there are the red, green and yellow samples. One general observation can be drawn from Fig. 1 and since all the samples analyzed present spectral reflectivity in the infrared range significantly higher than that in the visible portion of the irradiance spectrum, also thanks to the solar reflective engobe under the glaze. The only exception is represented by all the four samples with pigment Y1. Moreover, it is important to notice that the solar reflectance of the engobed substrate without any glaze is ρ= 0.54 while color coordinates are L*= 78.93, a*= 0.03 and b*=1.82. . Regarding Black samples, spectral reflectivity of black samples is reported on the first line of Fig. 1. To facilitate the comprehension of the figures above, the solar reflectance calculated in combination with the air mass 1 global horizontal (AM1GH) irradiance spectrum are reported in Table 3.

Table 3. AM1GH solar reflectance of black samples.

	G3_	G6_	M3_	M6_	G3Z_	G6Z_	M3Z_	M6Z_
B1	0.19	0.09	0.16	0.11	0.21	0.14	0.26	0.18
B2	0.13	0.10	0.13	0.10	0.14	0.13	0.23	0.16
B3	0.18	0.10	0.14	0.11	0.19	0.16	0.27	0.19
B4	0.27	0.07	0.11	0.10	0.29	0.10	0.18	0.12

On all the series the reflectance of samples containing 3 wt% of pigment is, as expected, higher than that of samples containing 6 wt% of pigment. In general, samples containing CoO are characterized by an absorption peak in the wavelength range between 1000 and 1800 nm that causes a decrease in solar reflectance. This effect is caused as well by the presence of Fe_2O_3. Analyzing the color, all samples containing zirconium silicate present L* values significantly higher than the corresponding samples without $ZrSiO_4$. Considering the overall performance, the best performing samples are the ones containing pigment D4 for gloss samples (G3_D4 and G3Z_D4), while among matt samples the best performing ones are M3_D1 and M3Z_D3. The less performing samples for all the analyzed groups are the ones containing pigment D4. Analyzing together solar reflectance and L* parameter, as reported in Fig. 2 (left), it can be noticed how both solar reflectance and L* decrease with the increase of pigment wt%. This occurs even if L* and solar reflectance trends do not always match. All the red samples present good values of solar reflectance even if it remains under 0.50. Each sample also presents values of infrared reflectance higher than that in the visible range of the irradiance spectrum. The variation of solar reflectance among red samples (Table 4) presents the same trend as reported above if the variation in concentration of the pigments is considered. In fact, with a higher amount of pigment, solar reflectance decreases sharply.

Table 4. AM1GH solar reflectance of red samples.

	G3_	G6_	M3_	M6_	G3Z_	G6Z_	M3Z_	M6Z_
R1	0.39	0.33	0.43	0.42	0.47	0.40	0.55	0.47
R2	0.30	0.27	0.31	0.29	0.42	0.35	0.46	0.38
R3	0.21	0.19	0.27	0.24	0.31	0.27	0.39	0.32

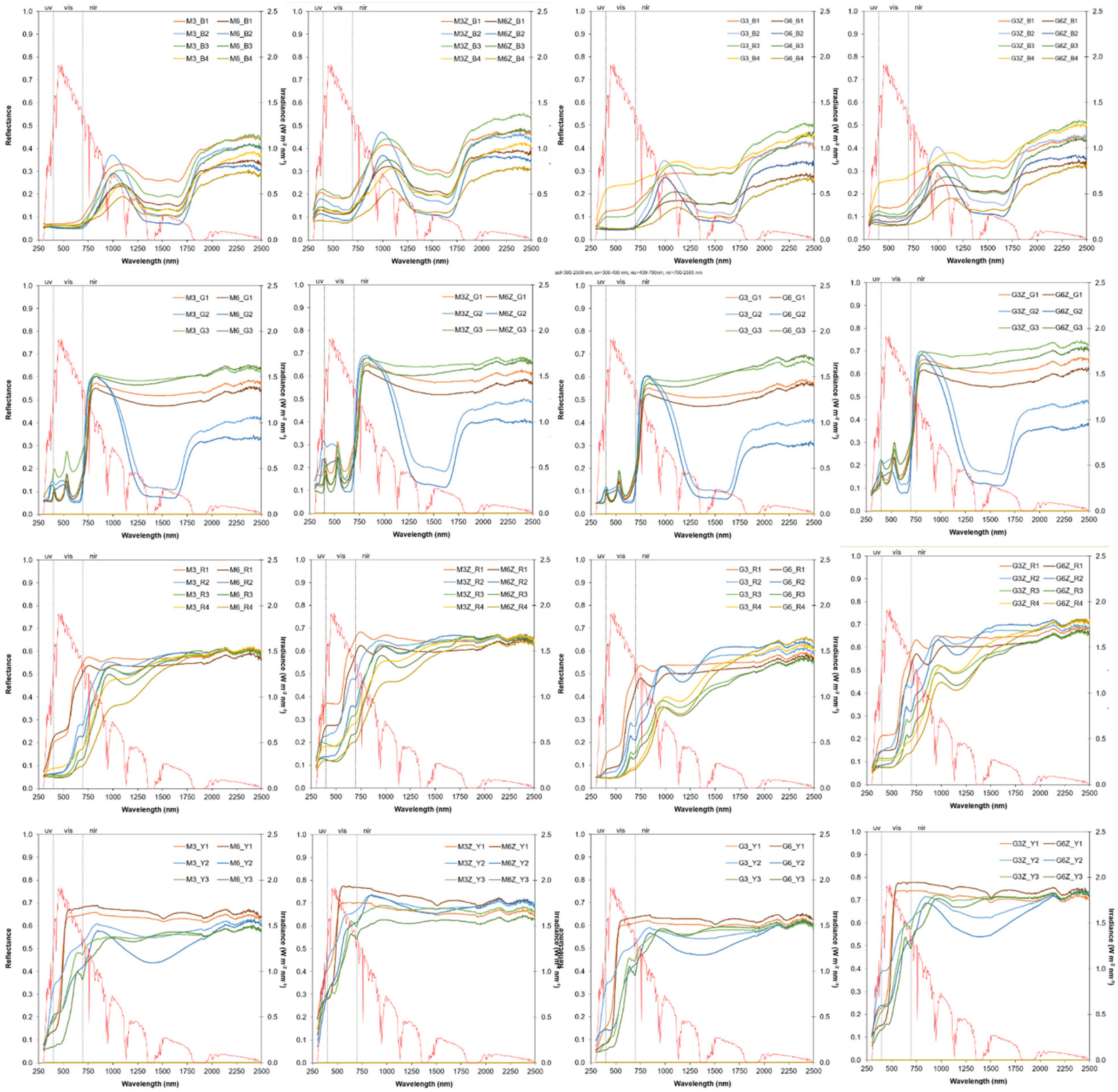

Fig. 1. Gloss samples spectral reflectivity (left) Matt samples spectral reflectivity (right). The red irradiance spectrum represents the AM1GH spectrum, used to integrate the spectral reflectivity in order to calculate the solar reflectance.

Samples characterized by the presence of pigment R1 (with a relevant amount of $ZrSiO_4$) are characterized by higher values of both solar reflectance and L* (Fig. 2 right) if compared with other samples containing the same amount of pigment. The presence of iron oxide tends to counteract the effects of zirconium oxide due to its absorptive behavior, while the presence of chromium oxide tends to promote both solar reflectance and L*. Despite the visual appearance of the samples, which look deeply different each from the other, the difference between solar reflectance and L* appears quite regular. Moreover, matt samples tend to reach values of solar reflectance and L* higher than the corresponding glossy samples. This can be motivated by the formation, during the heating treatment, of mineralogical phases which reflect solar radiation slightly more.

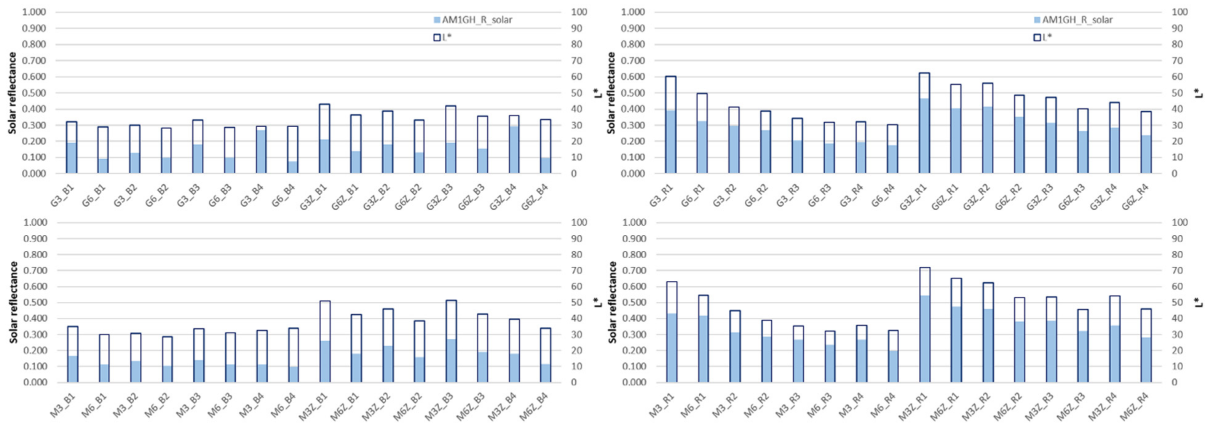

Fig. 2. Comparison between Solar Reflectance and L* for black (left) and red (right) samples.

In contrast with other samples analyzed up to now, the difference in solar reflectance on green samples (Table 5) does not vary significantly with the pigment amount. Among green samples, a big variation in visual response can be noticed, in particular due to the presence of cobalt oxide in G2 pigment. The presence of cobalt oxide is shown in reflectivity spectra (Fig. 1) by the absorption peak in the wavelength range between 1000 and 1800 nm.

Table 5. AM1GH solar reflectance of green samples.

	G3_	G6_	M3_	M6_	G3Z_	G6Z_	M3Z_	M6Z_
G1	0.29	0.27	0.30	0.28	0.39	0.35	0.40	0.35
G2	0.25	0.22	0.26	0.24	0.33	0.28	0.37	0.32
G3	0.32	0.31	0.38	0.31	0.41	0.40	0.40	0.35

The best performing samples in this set are the glazes containing pigment G3, which is constituted by 100 wt% of chromium oxide, known for its reflecting properties. As reported above, the presence of zirconium silicate increases significantly both solar reflectance and L* of all samples of this set (Fig. 3 left). If compared to trends observed on red samples, on this set both solar reflectance and L* tend to decrease for gloss and matt samples with the same amount of pigment and zirconium silicate. Yellow samples are the ones that present higher solar reflectance even if samples with pigments Y2 and Y3 are more similar to light terracotta. Among yellow samples, those characterized by the presence of pigment Y1 seem to not be influenced by the variation in pigment's percentage in the formulation, differently from pigments Y2 and Y3. A significant contribution to the good performance of pigment Y1 is given by the high amount of zirconium oxide in the pigment (Table 6). Samples characterized by the presence of pigment Y3 are characterized by slightly lower solar reflectance probably due to the amount of iron oxide in the pigment.

Table 6. AM1GH solar reflectance of yellow samples.

	G3_	G6_	M3_	M6_	G3Z_	G6Z_	M3Z_	M6Z_
Y1	0.51	0.52	0.55	0.56	0.63	0.63	0.63	0.66
Y2	0.49	0.38	0.49	0.40	0.57	0.48	0.62	0.56
Y3	0.39	0.36	0.42	0.35	0.53	0.46	0.54	0.51

Fig. 3 (right) highlights how samples containing pigment Y1 are characterized not only by higher solar reflectance but also by higher L*. Analyzing the L*a*b* measurement it can be noticed what was already stated previously with regard to the difficult of distinguishing between red and yellow samples since some red point trespass the yellow cloud as well as some yellow points. Green and black samples are represented by two limited clouds of points. The black one not only remains in the lower part of the L* axis but also remains close to the intersection between a* and b*, thus representing a neutral black color without other chromatic influences. The green cloud is slightly affected by the pigment G2 which is shifted on the positive part of the b* axis.

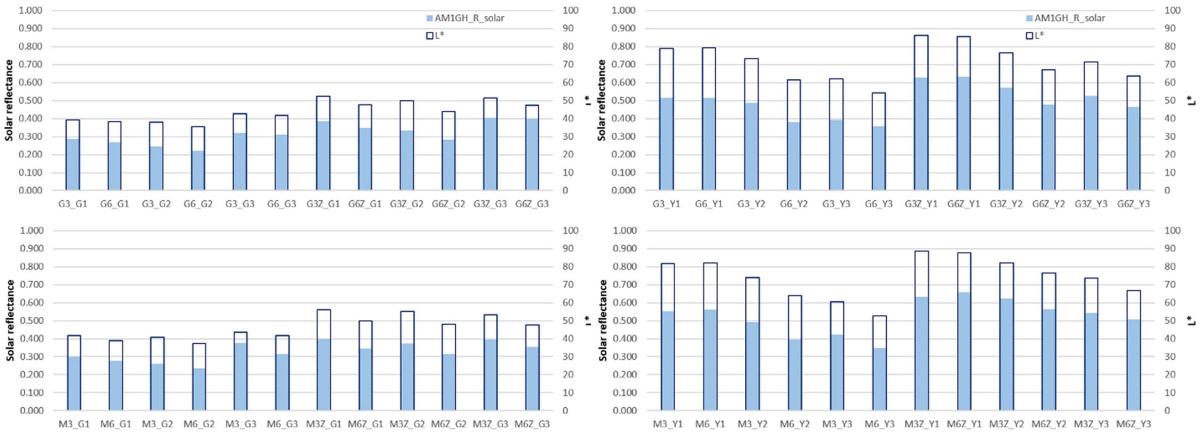

Fig. 3. Comparison between Solar Reflectance and L* for Green samples (left) and Yellow samples (right).

4. Conclusions

Solar reflective materials are among the most effective ways to counteract the urban heat island effect. Recently, inorganic materials such as those for roof tiles and ceramic tile mantles are being developed to provide solutions more durable under weathering and characterized by easier cleaning processes. This study is aimed to propose a first overview on several ceramic glazes by analyzing their solar properties, in the perspective of creating a range of solar reflective ceramic glazed tiles. Fourteen different pigments with different color, as well as different chemical and mineralogical composition, were provided, chosen among the most used ones for the external building envelope. Combining the pigments with different glazes, 112 samples were obtained. This work is focused on the analysis of their solar reflectance and color, as well as on how these parameters are affected by the pigments features. The first parameter influencing solar reflectance and color is, obviously, the chemistry of the pigment: in particular, the presence of zirconium and chromium oxides tends to enhance the solar reflectance, whereas cobalt and iron tend to reduce it. The addition of zirconium silicate to the glazes further improves the solar reflectance. On the other hand, it alters the visual appearance of the samples. The presence of a matt frit, moreover, improves the solar reflectance if compared to a gloss frit. This can be due to the formation of crystalline phases, which reflect in a better way solar radiation than an amorphous glossy coating. In general, the solar reflectance of the samples is also influenced, as expected, by the pigments concentration within the glaze. Analyzing the visual response of the sample, and considering each color family, samples which appear lighter are generally characterized by a higher solar reflectance than others. Concerning roofing solution, it is important to remember that solar reflectance values are sensibly lower than commercial cool roof coatings but, if compared to colored products with the same visual response, the samples produced in this study present pretty interesting values. In sight of this, only products with solar reflectance higher than 0.50 may be defined as true “cool colors”. This requirement is pretty stringent in this case as just few samples can be defined “cool”, and all of them belonging to the yellow family of colors apart from one from the red family. In conclusion, this study provides an overview on the behavior of ceramic glazes with different finishing in terms of solar reflectance. Guideline are drawn to help in the creation of a new series of ceramic cool-colored tiles that couple the advantages brought by cool materials with the durability vs. time and the long term affordability of ceramics if compared with organic ones.

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