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Characterization of ceramic tiles coated with recycled waste glass particles to be used for cool roof applications

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ABSTRACT

This study presents a novel proof of concept in the literature of using waste glass for solar reflectance purposes that can be useful for future research and applications with the aim of suggesting an appealing alternative for substituting raw materials in the ceramic industry. This research is framed in the field of the production of new sustainable and resilient construction materials, which is gaining a huge concern in the literature since using recycled materials is considered as a positive strategy for saving natural resources and mitigating potential environmental risks. In this context, waste glass (WG) being recycled globally is deemed insufficient compared to the discarded quantity intended for landfill disposal. The objective of this study is the characterization of ceramic roof tiles by using recycled WG particles as a coating in order to evaluate their potential use for cool roof applications. This characterization encompasses the assessment of their physical, mechanical and optical characteristics, including geometrical characteristics, flexural strength, permeability, frost resistance, fire behavior and leaching test in addition to colorimetric and normal solar radiation reflectance characterization. Furthermore, the study investigates the influence of WG type and shape of roof tiles on this characterisation. To conduct this study, flat and curved roof tiles models were produced on a laboratory scale following a single firing process. Two types of WG were used for the coatings: soda lime silica WG (WG1) and cathode ray tube (CRT) WG (WG2), which were specifically prepared for experimentation, varying in shape and type of WG. Then, a statistical analysis was performed to identify the influence of tiles shape and type of WG on the solar reflectance behavior, and to analyze the probability distributions for the different spectral normal reflectance measurements. The results demonstrated that the application of WG as a coating on roof tiles generally enhances their physical and mechanical characteristics. Specifically, the use of WG2 improves the normal solar reflectance performance of flat tiles, which makes it a sustainable solution to be introduced for cool roof applications and subsequently contribute to circularity and mitigation of urban heat island effect.

1. Introduction

Due to progressive urbanization worldwide, the construction industry is facing challenges in producing materials with sustainable life cycles and quality performance. For instance, cool materials had gained huge scientific attention recently, these materials are designed to reflect solar radiation and absorb less heat than conventional materials which influences the heat radiation into the atmosphere and reduce the temperature in buildings. Therefore, the development of such materials with solar radiation reflective properties proved promising results in the mitigation of urban heat island. The most discussed are cool roofs [1], as the heat-gain through the roof represents about 50–60% of the total heat gain in the building [2], while the cooling energy represents 2.9% to 6.7% of the world energy consumption [3]. This cooling strategy significantly reduces energy demand for cooling loads in air-conditioned houses in hot climate regions [4,5]. Recent study indicates that buildings using cool roofs achieve energy savings varying from 15.0% to 35.7% in different climatic zones [3]. In this context, designing clay roof tiles applicable for cool roof requirements will have the ability to enhance the building energy efficiency and indoor thermal comfort conditions [6].

On the other hand, the world economic growth has caused an increase in the waste production [7], and the scientific community is in

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constant research to develop new ways of recycling waste considering it as an alternative resource to alleviate the pressure from using natural raw materials. Among these wastes, as a non-biodegradable one, waste glass (WG) has attracted a considerable attention, only 21% is recycled globally, the rest of the waste is destined to landfill disposal [8]. Hence, the substitution of raw materials by secondary ones such as WG presents an interesting sustainable alternative in the fabrication of construction materials which proved promising results [9–21]. In this regard, and according to the literature, the construction industry is the main sector testing and using WG in the production of materials, which has been used and tested in different ways; incorporated as a raw material substitution in the fabrication of ceramic-based materials such as tiles and bricks, and in a complete or a partial substitution of clay [14,15]. In both forms, the main objective has been to improve the physical, mechanical and environmental performance of the products and decrease the melting time for the firing of ceramic-based materials [16]. In terms of pozzolanic properties, the use of WG in concrete as for partial substitution for cement or sand proved good results in terms of rigidity and durability [17–19], it improves the compressive strength of cement mortar cubes in an ultrafine material form with 10% of cement replacement [22]. More specifically regarding the type of WG used, the one derived from spent fluorescent lamps was successfully used to replace Portland cement in mortars [20], Dusan et al. [21] found that cathode ray tube (CRT) glass can be successfully used for making concrete blocks and paving flags. Moreover, the replacement of 50% of quartz sand with panel recycled glass with a fraction of 0.25/1.00 mm in the finishing layer, complies with the standards and does not affect the physical and mechanical characteristics. Other applications include the use of WG powder in asphalt mixtures which was discussed and proved significant benefits in constructing environmental friendly pavements [23–26].

The other way of incorporating WG in the production of ceramic-based materials is a partial substitution for glazes raw materials. In general, glaze suspensions are composed of non-plastic materials such as frits and feldspars, and plastic materials such as clay and kaolin and additives [27]. Due to the similar composition of frits and WG, which are mainly composed of mixtures of silicates and carbonates, it was proved that WG presents a good alternative for frits partial substitution [28]. According to Andreola et al. [29], up to 30% of recycled WG powder could be incorporated into glazes. More recently, Cardoso et al. [14] pointed out that it is feasible to use an 8 % mass of laminated WG for the manufacture of ceramic frits. In this context, the most common type of WG used was CRT glass that is mainly derived from TV sets and pc monitors and contains significant amounts of alkali and alkaline earth oxides. In their study, Andreola et al. [30] found that the use of CRT panel glass as a substitute for ceramic glaze frits reduces the environmental impact of the ceramic glaze production process, also it decreases the production expenses with a substitution of 25 wt% [31]. In this regard, Revelo et al. [32] found that the optimal amount to maintain the properties expected for ceramic glazes is 20 wt%. In all cases, this potential use of WG in ceramic-based materials alleviates the pressure from landfills and opens a new perspective for WG management.

According to the state of the art, the use of WG was discussed as a partial substitution of raw materials for the manufacturing of ceramic products, and the assessment of the influence of using WG particles for the exterior surface of roof tiles as a coating, including solar radiation reflectance assessment has not been discussed in literature and no other studies has been found about it yet to the best knowledge of the authors. Hence, the objective of this study is the characterization of ceramic roof tiles using recycled WG particles as a coating in order to evaluate their potential use for cool roof applications. For this purpose, this research was focused on evaluating three main assumptions: (i) recycled WG could be used as coatings without affecting the performance of the roof tiles; (ii) the surface color of roof tiles coated with WG measured by lightness, influences the solar reflectance of the tiles; and (iii) the type of WG and shape of tiles influence the solar reflectance.

To this end, a performance evaluation in terms of roof tiles shape and WG type is conducted by testing the geometrical characteristics, the flexural strength of the tiles, permeability, frost resistance, potential risk for human health and the environment (lead leaching), solar reflectance and colorimetric characteristics. The results of the tests conducted were interpreted and discussed, and a statistical analysis was performed to identify the influence of tiles shape and type of WG on the solar reflectance behavior. Finally, a set of the main conclusions obtained were stated to summarize the contributions of this study and future investigations were proposed.

This research builds on the previous findings of a prior work, where a preliminary characterization of sample clay specimens covered with WG (small square samples covered with WG) was performed in terms of the influence of the following parameters: the type of WG, the temperature and holding time of firing treatment, and thickness of WG coating. This present work represents the main stage of the research using real shape of roof tiles, where the starting point is based on the best experimental results obtained from the characterization of different samples clay tiles reported in [33], i.e. by using the optimal combinations of parameters, and using only those two types of WG that showed good results and a firing temperature of 1000 °C.

2. Materials and methods

The research was carried out using a structured approach divided into three stages: (i) The preparation of raw materials and the fabrication of two types of clay roof tiles (flat and curved) with WG coatings at laboratory scale; (ii) the experimental process performance for the tiles characterization; (iii) and finally the interpretation and the analysis of the results obtained based on the comparison of the types of WG and the shape of roof tiles on the basis of a statistical analysis conducted for the solar reflectance data. In the sections below, the materials used, the fabrication procedure and the tiles characterization setup of the laboratory testing are described.

2.1. Raw materials

For this research, clay roof tiles (curved and flat) covered with WG were manufactured through a single firing process, and two types of WG were used for the coatings: soda lime silica waste glass (WG1) derived from recycled hollow glass and lead silica waste glass (WG2) derived mainly from CRT of television sets. The chemical composition of WG is shown in Table 1. The used particle size of WG was reduced and passed through a 0.63 mm sieve for better sintering process and adhesion to the clay body. The glass-collection plant “Camacho Recycling” located in Albacete, Spain, provided the WG; this company has developed glass-collection systems for all the different types of glass, regardless of the origin, composition, and quantities that can be generated on factories or homes. The WG undergoes a processing line that consists of cleaning, milling, sieving and drying. For the ceramic body (substrate), the preparation was carried out using a local prefabricated terracotta clay paste with 22% of humidity that provides ceramic bodies characterized with a high environmental degradation resistance and a high frost resistance. In addition, the commercial engobe Cerograf, an opalescent solution, was used for the intermediate layer between WG coat and the substrate to enhance the adhesion between the two materials. For each tile, two coats of the engobe were applied.

2.2. Fabrication procedure of experimental samples

A number of 36 flat roof tiles measuring 19x13.5x1cm and 36 curved roof tiles measuring 19x10x4cm were fabricated at the laboratory scale. The size of the tiles adopted in this study conform to the Spanish technical building code (CTE) and it was reduced to 50% from the common size in the market due to the capacity of the laboratory kiln, but keeping the shape and scale. This is not really a limitation since measurements

Table 1
Chemical composition in wt.% of WG1 and WG2 obtained from FRX analysis.

	SiO_2	Na_2O	CaO	MgO	Al_2O_3	Fe_2O_3	K_2O	TiO_2	P_2O_5	BaO	PbO
WG1	70–74	12–14	7–11	1–3	0 0.5–3	<0,1	0.2–0.9	–	–	–	
WG2	50–55	5–7	2–4	1–2.5	1.5–3	0.05–0.1	6–9	0.05–1	–	1–3	17–24

are not affected by the actual size of the specimens since they are made to scale. Table 2 presents the identification code of the six groups of tiles used in the tests. For the fabrication process of the samples, the clay was shaped manually with the help of laboratory molds to form the required shape for the production of flat and curved roof tiles (Fig. 1). The curved roof tiles were pressed on later using molds to obtain the bent shape. At the first stage, the samples were dried using laboratory furnace for a temperature degree of 100 °C for 48 h; the second stage was for the WG particles application with a thickness of coating less than 1 mm and a quantity of 450 g/m^2 on the exterior surface of the tiles after applying an intermediate engobe coat to ensure the adhesion of WG particles to the tiles, the one coat was determined as the optimal choice resulted from experiencing the influence of different number of coats on the coherence between the WG coat and the substrate. The last stage was for the firing treatment of the samples at the laboratory kiln using a temperature degree of 1000 °C during 60 min with a heating rate of 1.22 °C/min that takes up to 1.5 h to get to 1000 °C. In general, the manufacturing of tiles could be through a single firing process that consists of one single firing treatment phase, or following a double firing process that takes on two phases of firing treatment, the first phase is for the biscuit preparation and the second is for the glaze melting. In this study, the single firing process was followed to decrease the energy consumption at the greatest extent, and to ensure a better adhesion of materials.

As it was mentioned before in the introduction section, the holding time of the firing treatment is an influential parameter depending on the WG type. Consequently, a preceding testing was conducted for real shape roof tiles, and it showed that using a thickness of less than 1 mm of WG, and adopting a firing treatment during one hour for 1000 °C resulted in homogenous products that can ensure the reproducibility of data for this work. Hence, this pattern was adopted in the preceding research.

2.3. Tiles characterizations

For the experimental part, the technical characteristics described in the standard UNE EN 1304 [34] that includes geometrical characteristics, flexural strength, permeability, frost resistance and fire behavior were measured. In addition, a leaching test [35], and the colorimetric and normal solar radiation reflectance characterization through the VIS and NIR solar radiation spectrum [36] were performed. Table 3 shows the tests conducted according to the above standards and the samples used for each one.

In order to evaluate the difference between the samples in terms of color and the influence of the application of WG coatings on the lightness of tiles, a colorimetric characterization was conducted. Moreover, the solar reflectance test was performed to evaluate the reflectance behavior of the samples according to the type of WG and shape of tiles.

Table 2
Identification code of the configurations of tiles based on the shape of tiles and type of waste glass.

Shape of tiles	Configurations	Identification code
Flat tiles	Reference	**FRef**
	With WG1	**FWG1**
	With WG2	**FWG2**
Curved tiles	Reference	**CRef**
	With WG1	**CWG1**
	With WG2	**CWG2**

The colorimetric characterization and the normal solar reflectance tests were performed using the fiber optic Stellarnet spectrometer. The spectrometer works in the radiation range of 350–1100 nm, taking measurements for the VIS and NIR radiation according to ASTM E903 standard for laboratory measurements [36]. All the experimental set up is presented in the previous study [33]. The colorimetric coordinates of the samples representing WG1, WG2 and the reference were determined in compliance with the CIE-1976 l*a*b* colorimetric method defined by the international commission on Illumination (CEI EN 60335–2-27).

Before starting the measurements, the calibration procedure was applied and checked through the dark and response corrections using a piece of white Teflon from which the data collected is scaled in reflectance as to the reference material. For a more accuracy of the results, the process of measuring the normal solar reflectance was repeated for nine points for each sample of each of tiles used: three FRef, three FWG1, three FWG2, three CRef, three CWG1 and three CWG2, which makes a total number of measurements of 162 spectral readings. The tile samples were subjected to normal radiation emitted by the source that has a detector integrated (Fig. 2). In the experiment, the light strikes the surface from a perpendicular source integrating a sensor that receives the reflected radiation. The test was performed at same conditions for all samples (curved and flat); the tilt sides of the curved tiles were measured by keeping the distance between the detector and the surface the same in all points where the beam hit the surface. Subsequently, the data were processed using the Spectrawizz OS v5.33 software.

The spectral solar reflectance $\rho\lambda$, was integrated over the solar irradiance spectrum $Isol,\lambda$, based on ASTM E-490 to obtain the solar reflectance of samples ρsol, at wavelength λ:

$$\rho sol = \frac{\int_{350}^{1100} \rho\lambda Isol,\lambda d\lambda}{\int_{350}^{1100} Isol,\lambda d\lambda} \tag{1}$$

In this study, the normal solar reflectance behavior of the samples was evaluated taking into consideration the perpendicular striking radiation coming from the illuminating source, and the measurements conducted using the procedure described above characterize the behavior of the tile in normal radiation incidence.

Finally, the results obtained of the experimental process were analysed and interpreted based on a comparison for the WG type and roof tiles shape. For this matter, a statistical analysis was conducted to identify the influence of tiles shape and type of WG on the solar reflectance behavior, and to analyze the probability distributions for the different spectral reflectance measurements. The data acquired were processed starting with the normality test, the distributions were not normal; hence the non-parametric Mann–Whitney *U* test was performed in all cases for the overall distribution of reflectance.

3. Results and discussions

The results of the physical, mechanical, and optical characteristics of the tiles studied are shown in Table 4. According to these results as discussed below, the samples showed clear different behaviors with regard to the type of WG coatings applied and the shape of roof tiles. Nevertheless, all samples comply to the standards of the tests performed and the results demonstrated that using WG as coatings for roof tiles does not affect the performance of roof tiles regardless of the shape and WG type.

For the analysis of the specimens, it is interesting to mention the

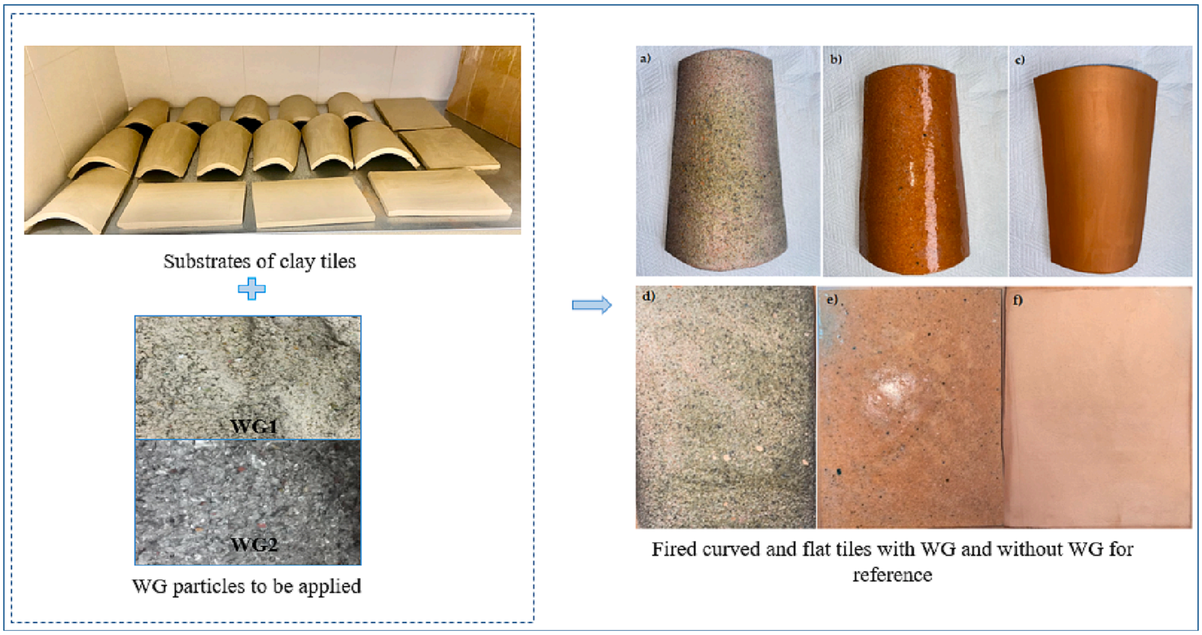

Fig. 1. Main stages of roof tiles fabrication and appearance of the curved and flat tiles used in the experimental setup **a)** CWG1, **b)** CWG2, **c)** CRef, **d)** FWG1, **e)** FWG2 and **f)** FRef.

Table 3
Tests performed for the characterization of tiles with the standards followed and the samples used for each one.

Tests	Standard	Samples used
Geometrical characteristics	UNE-EN 1304 [34]	All Tiles
Flexural strength	UNE-EN 538 [37]	3 samples of each tile
Permeability	UNE-EN 539–1[38]	3 samples of each tile
Frost resistance	UNE-EN 539–2 [39]	3 samples of each tile
Fire behavior	UNE-EN 1304 [34]	All Tiles
Leaching test	UNE-EN 12457–1 [35]	Ratio of 2 l/kg
Colorimetric characteristics	CEI EN 60335–2-27	3 samples of each flat tile
Solar reflectance	ASTM E903 [36]	3 samples of each tile

difference in the visual appearance of the tiles with WG1 and WG2 coatings and the reference. As it can be seen in Fig. 1, samples with WG2 coating show a notably transparent and a glossy surface, unlike WG1 coated samples that have a rough and porous green surface; while both reference and WG2 samples have the same color, a terracotta-type color, which is the real color of the substrate. In the following subsections, it is described the main findings of the measurement tests.

3.1. Geometrical characteristics

The geometrical characteristics for all the tiles used in this study conform to the standard. More specifically, tiles covered with waste glass showed no difference from reference tiles in terms of form, regularity, rectitude and dimensional tolerance. This result demonstrates that using WG can be considered as normal coatings for roof tiles and its

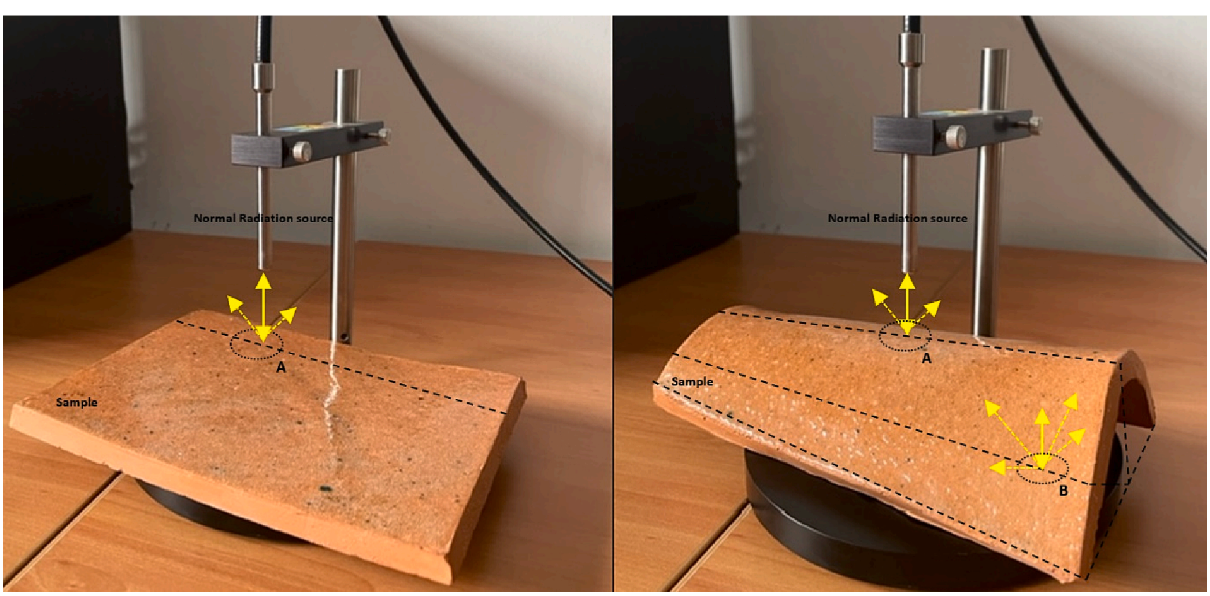

Fig. 2. Normal radiation reflectance experimental set-up using the Stellarnet spectrometer at different points of a curved tile and a flat tile.

Table 4
Tests results of flat tiles, curved tiles and reference tiles according to UNE-EN 1304.

Test	FREF	FWG1	FWG2	CREF	CWG1	CWG2
Geometrical characteristics	Conform to the standard					
Flexural strength (KN)	0.62	0.65	0.66	0.97	1.07	1.16
Permeability	No water drops					
Frost resistance	No damage					
Fire behavior	Conform to the standard					
Colorimetric characteristics (L*)	70.95	58.32	98.43	70.95	58.32	98.43
Solar reflectance	0.42	0.39	0.8	0.3	0.26	0.41

use does not impact the geometrical characteristics requirements.

3.2. Flexural strength test

According to the results of the flexural strength test (Table 4), the specimens of flat tiles hit the breakage conforming to the standard that establishes a limit for a bearing capacity of 0.6kN for flat tiles. For specimens of curved tiles, they conformed as well to the standard that establishes a limit of 1kN, with a slight deficiency of the reference tiles which is not relevant due to the difference of the specimens manufacturing process between industrial and laboratory scale. In terms of the flexural strength, it is significantly influenced by the shape of tiles since it has increased for curved tiles by approximately 50% with regard to flat tiles, which is not surprising since it is known that curved tiles are more resistant to flexion due to the bent structure.

With regard to the WG type, the application of WG showed an improvement of the flexural strength regardless of the shape of tile, as it increased by 4.61% and 6.06% for FWG1 and FWG2 respectively, and by 9.34% and 16.37% for CWG1 and CWG2 respectively. This improvement can be due to several factors: the chemical composition of WG that creates a good adhesion between the substrate and the coatings, and the presence of oxides in their composition that decreases the melting time and favors the sintering behavior during the firing treatment by generating a strong transition zone. As a result, the application of WG2 showed better flexural strength performance with regard to WG1 for both shapes of tiles. In general, according to previous investigations, the use of waste glass in the fabrication of ceramic construction materials generally increase the flexural strength, while decreasing the particle size of glass [15,40–42]. In a study conducted by Dusan et al. [21], it was proved that when using CRT glass in the finishing layer formulation of concrete paving blocks and flags, the strength quality prescribed by the standard was not degraded, which is consistent with the results obtained in this work.

3.3. Permeability

For the permeability test, all the specimens showed high impermeability (Table 4), no water drops were observed during the period of the test (20 ± 4)h for all the samples, therefore the tiles are considered waterproof. More specifically, tiles covered with WG2 showed less moisture on the lower surface, unlike the rest of the tiles that developed more moisture on the lower surface. This is explained by the porous texture of WG1 and the reference specimens, as the permeability of ceramics is normally reduced when decreasing water absorption and implicitly decreasing porosity [43], which allows to reduce the thickness of the product.

3.4. Frost resistance

As for the frost resistance test, according to the results, no damage occurred to the tiles specimens during the 30 freeze and thaw cycles. The freezing and thawing cycles did not cause any structure cracks or show any quality reduction, both in WG coating and substrate, which means good levels of porosity and absorption (Table 4). These results are consistent with the results of permeability test, accordingly, and considering that the frost resistance as one of the important parameters influencing the durability of clay building materials [44,45], it can be affirmed that the tiles studied are able to withstand adverse climatic conditions, and subsequently presents a positive durability potential.

3.5. Fire behavior

Regarding fire behavior of the tiles, the test was not required because all types of tiles fulfill the Commission Decision 2000/553/CE conditions taking in account that the material of covering is inorganic. Moreover, the tiles can be classified for fire reaction in A1 class without testing according to the Commission Decision 96/603/CE.

3.6. Leaching test

The use of recycled waste glass in this study, fundamentally for WG2 derived from CRT of computers and TV monitors, requires the performance of a leaching test, especially due to its lead concentration. Hence, a leaching test was performed to evaluate and determine any potential risk for human health and the environment in the eluate obtained, in which a ratio of 2 l/kg (water/solid) was adopted.

According to the ASTM D3987 (2012) [46], lead ratio in the sludge sample obtained from tiles must be lower than 5 mg/l [34], As it can be seen in Table 5, the concentrations of lead (Pb) and cadmium (Cd) comply with the permissible limit of the regulatory standard, unlike the rest of the metals, and the possibility of lead leaching contained in the CRT WG is null. Moreover, and contrary to what was expected a priori, concentrations of metals measured in tiles made with CRT waste glass (WG2) are lower than the soda lime silica waste glass (WG1) ones. The rest of the elements reported in the test are used as a raw material for glass manufacturing, and apparently, the clay used in this study has traces of these elements as well. As it is seen in the Table 5, the concentrations of Arsenic (As) and Barium (Ba) exceed the permissible limits; however, their similar quantity in all the samples shows that their origin is not from the WG but from the clay used for the substrate. In addition, the high chromium (Cr) content could be quite compromising for the elaborated ceramic pieces; however, it is observed in both raw materials. It is used as a coloring element in green glasses which is coherent with its high concentration found in the glazed tiles, especially in those ones made with WG1. The results demonstrate that the use of WG does not present a potential risk for human health and the environment in this study, especially that lead leaching contained in the CRT WG was null. In general, the low toxicity levels obtained when using CRT WG in ceramic materials fabrication ensures the viability of this use and encourages the recycling of such WG type for the substitution of other materials [12,15,47].

3.7. Colorimetric characterization

Regarding the colorimetric characterization, the results of the lightness coordinates of the three configurations (WG1, WG2 and the reference regardless of the shape of tiles) are shown in Fig. 3 and Table 6. These results indicate that tiles covered with WG2 have the

Table 5
The metals most reported as toxic concentrations in Reference, WG1 and WG2 samples in mg/l.

Samples	As	Ba	Cd	Cr	Pb
Reference	12,427	166,426	0,023	134,961	0
WG1	12,179	166,444	0,211	284,130	0
WG2	12,103	147,099	0,125	190,195	0
Regulatory	5	100	1	5	

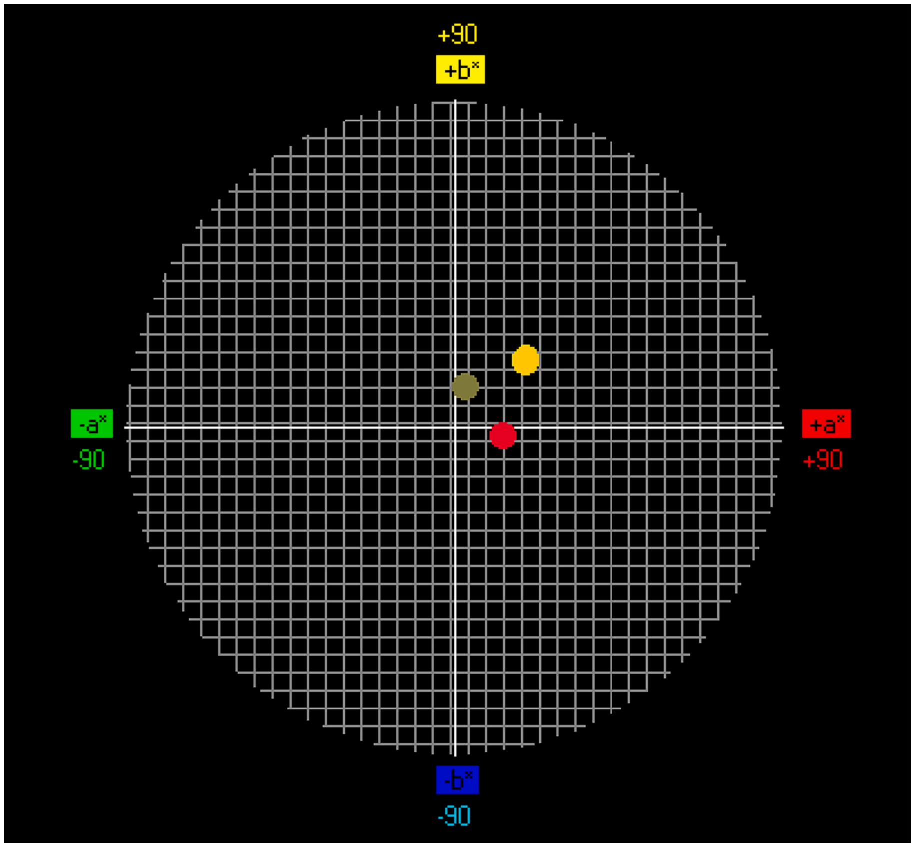

Fig. 3. Lightness coordinates of the three configurations: tiles with WG1 (green), tiles with WG2 (Red) and tiles without WG for reference (Yellow). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 6
Lightness coordinates of WG1, WG2 and the reference. The percentage of saving regarding the reference is reported between brackets.

Samples	L*	a*	b*	Chroma	Hue
Reference	70.95	+18.685	+19.579	27.06	46.33
WG1	58.32 (-18%)	+2.610	+9.710	10.06	74.94
WG2	98.43 (28%)	+8.723	−1.598	8.87	−10.38

highest lightness which increased by 28% with regard to the reference, and the application of WG1 decreases the lightness by 18% with regard to the reference. From the visual appearance of the tiles shown in Fig. 1, it can be seen that roof tiles with WG2 have a transparent gloss surface due to the composition of WG, especially the existence of K_2O [14], conversely to roof tiles with WG1, that exhibit a textured green and matte surface compared to WG2 and reference that both show a light orange terracotta-type color. Therefore, it can be set that the application of WG2 coating improves the lightness which is consistent with the results obtained in [33], that show a decrease in lightness with the use of WG2 on the small clay pieces. The color surface of the materials implemented in cool roof structures was a key to improve their performance as a first strategy before using white pigments. This point is further analysed in the next section (3.8. Solar reflectance). According to previous studies, the use of light colored materials enhance the solar reflectance response [48–51], which makes the application of WG2 coatings on clay roof tiles the better potential candidate with regard to WG1 for cool roofs applications.

3.8. Solar reflectance

The results of the spectral solar reflectance tests conducted to evaluate the effect of the type of WG and the shape of tiles on the solar reflectance, are plotted in Figs. 4 and 5 which show the behavior of the spectral reflectance of the flat and curved tiles respectively. The procedure of gathering the data was described in the methodology section, and then these data were processed with the Spectrawizz software associated to the spectrometer, taking into account the reference samples and the calibration adjustments. These figures show the spectral reflectance in % compared to the perfectly emitter (white sample) in terms of the main wavelength range coming from the solar spectrum. As observed from these figures, WG2 coated tiles tend to have the highest reflectance in the wavelength range, both in flat and curved cases with regard to Reference and WG1. The profiles also show that FWG2 reflects more radiations than the rest of the configurations, moreover, all the samples depict the peak of reflectance around the wavelength 1000 nm in the NIR range.

In the range between 400 and 550 nm of the VIS spectrum, tiles with WG1 have more reflectivity than the reference tile samples. This behavior shows that the application of WG1 is not totally affected by the color of the substrate in this spectrum range. Moreover, tiles with WG2 reflect more in the spectral range between 700 and 1100 nm of the NIR. To have a closer evaluation of the normal solar reflectance between the different types of tiles, a comparison was set between the samples at the specific wavelength of 1000 nm, which represents the relevant solar spectral range between 700 nm and 1100 nm in the NIR range. The results are reported in Table 7, which show that the samples prove the same behavior as for the spectral range studied, being noteworthy the increase of the solar reflectance with the application of WG2.

According to the results shown in Table 7, a clear influence on the solar reflectance parameter can be observed regarding the shape and type of waste glass of the pieces tested. In flat tiles, the solar reflectance of FWG2 increases by 47,5% with regard to FRef, whereas the solar reflectance of FWG1 decreases by 7% with regard to FRef. For the curved

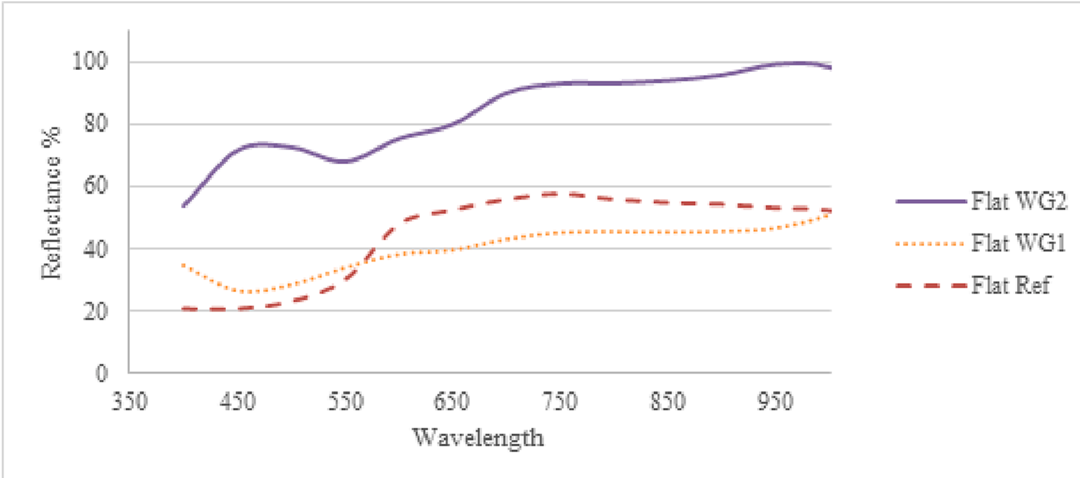

Fig. 4. The spectral solar reflectance profiles of FWG1, FWG2 and FRef.

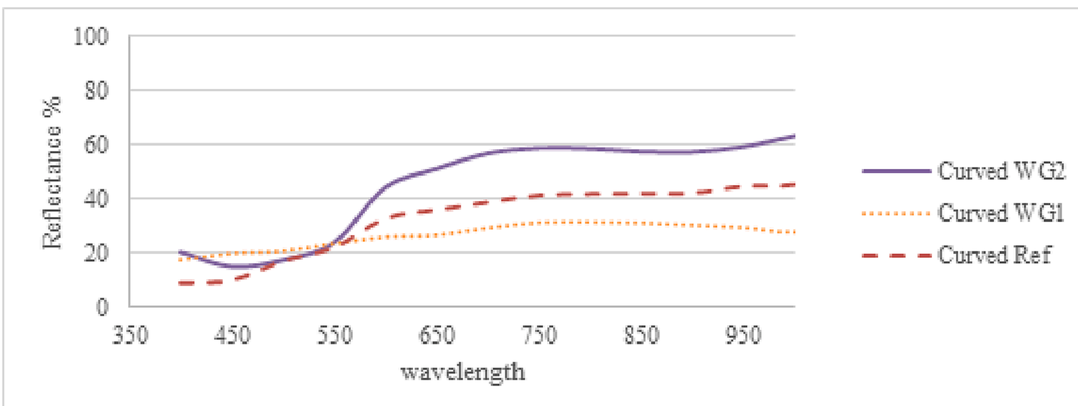

Fig. 5. The spectral solar reflectance profiles of CWG1, CWG2 and CRef.

Table 7
Solar reflectance at 1000 nm of the different types of tiles in comparison with total solar reflectance. The percentage of difference regarding the reference is reported between brackets.

Tiles	FREF	FWG1	FWG2	CREF	CWG1	CWG2
Solar reflectance	0.42	0.39 (-7%)	0.8 (47.5%)	0.3	0.26 (-13%)	0.41 (27%)
Reflectance at 1000 nm	52	50 (-4%)	98 (47%)	45	28 (-38%)	63 (28%)

Table 8
Mann-Whitney U to identify the difference in the overall distribution of reflectance across groups of tiles through the spectrum range 400–1000 nm in terms of the average rank between groups.

Groups	FWG1/ FWG2	FWG1/ FRef	FWG2/ FRef	CWG1/ CWG2	CWG1/ CRef	CWG2/ CRef
Asymp. Sig. (2-tailed)	0.000	0.168	0.000	0.024	0.035	0.022

tiles, the application of WG2 registered the highest solar reflectance as well with an increase of 27% compared to CRef, while the application of WG1 decreases the solar reflectance with 13%. Furthermore, the solar reflectance of flat tiles was the highest with regard to curved tiles.

Conversely to flat tiles, the spectral profiles of curved tiles show almost the same behavior for much of the spectral range, according to Fig. 4, all samples overlap around 550 nm and increase toward the NIR radiation. To identify if there are differences in the overall distribution of reflectance across the different groups of tiles in the spectrum range 400–1000 nm, the U Mann-Whitney statistical test was performed for the averages of the spectrum range (Table 8). According to Table 8, the overall distribution of reflectance along the wavelength range adopted for the study shows statistically significant differences across the following groups: FWG1-FWG2 and FWG2-FRef. The distribution of the spectral reflectance in terms of the average rank of FWG2 is not the same as FWG1 and FRef. In other terms, the results show that in contrary to FWG1 and FRef that display little difference from each other in terms of reflectance behavior across the spectrum, FWG2 has a specific response and mode of distribution. For curved tiles, all the groups show no difference in the overall distribution of the spectral reflectance.

The application of WG2 significantly increases the solar reflectance of tiles in accordance with the results found in [33], and flat tiles showed better reflectance in comparison with curved tiles in normal incidence experiments. These results are consistent with the colorimetric characterization results showed in Table 4, where the difference in surface colors of the samples influences the degree of lightness, and samples with WG2 has the highest lightness which is close to literature findings [14]. According to Xie et al. [48], the color lightness influences the spectral reflectance in the visible radiation region, the higher the lightness the higher the visible light reflectance. These results highlight the influence of the color of the material exposed to solar radiation on the solar reflectance, which conform to the results obtained by Kiletico et al. [49] investigating the use of recycled glass as a substitute for asphalt shingles aggregates to enhance the solar reflectance. Replacing the top surface granules with green glass and clear glass enhanced the reflectance by 44% and 55% respectively, however the material did not achieve cool roof attributes unless a white pigment was added to reach a reflectance of 0.275. This principle of using light color materials was highly used for cool roofs to improve their solar reflectance [50,51]. In this study, using light color clay roof tiles was a positive point for the WG coating substrate choice in coherence with previous findings.

To study the different response to WG2 of curved tiles with regard to flat tiles, a detailed analysis was conducted for the spectral reflectance readings of one curved tile in nine different points. Taking into consideration the fact that they have the same type of WG still flat tiles reflects more, by evaluating the response of each point of a curved tile (Fig. 6); it was found that at the highest points (P1, P2 and P3 in Fig. 6) of the curved tile where the radiation hit normally as it is shown in Fig. 2 at the point A, a high amount of energy is received. However, when the radiation hits the tilt sides of the curved tiles represented by point B in Fig. 2 (P4, P5, P6, P7, P8 and P9 in Fig. 6), the spectral reflectance decreases because of the radiations angle of incidence. The tilt sides may redistribute the percentages of reflectivity behavior of the sample according to the inclination angle of the reflected radiation. From this result it can be stated that if the sample is curved, it might maintain the same spectral reflectance behavior when exposed to different angles of the incident radiation. In this case, the solar radiation received by curved tiles will be normal to the surface for a long diurnal period than flat tiles. To study this assumption, it would be interesting to use a system of detectors distributed all along the hemisphere to receive the spectral reflectance response according to the angular distribution and the angle of incidence. Rossi et al. [52] presented an example of the reflectivity behavior of a white diffusive material according to different angles of incident radiation.

Finally, according to the results it can be stated that the samples drew different spectrums of normal solar reflectance being influenced by the type of WG applied and shape of tiles, which is explained by the difference in their compositions of WG and their role in shaping the sample surfaces' structure and color, moreover to the substrate color. The existence of the fluxing oxides K_2O and PbO in WG2 with a higher amount compared to WG1 influenced the sintering process during the firing treatment, and favored the development of a gloss transparent surface, conversely to WG1 that developed a rough, porous surface. This difference in surface structure and color influenced the spectral response to the incident normal radiation of the illuminating source for each sample. According to Spragg [53], the surface roughness affects significantly the scattering behavior of a surface.

The difference in solar reflectance behavior of the samples could be owed as well to technical conditions, considering the spectroscopy as delicate measurements. Many factors can influence the behavior of the solar radiation reflectance such as the orientation and angle of incidence, size and numbers of particles in a light beam, all this may influence the intensity of the light scattered which makes quantifying the intensity of the spectra difficult [54]. Xie et al. [48] found that each light source has a specific spectral power distribution that might affect the spectral response of the sample measured; it depends on where the spectral emission is focused.

4. Conclusion

This study provides a first proof of concept for using WG on roof tiles for solar reflectance purposes and serves as a reference for future researchers works. The goal of this paper was based on the characterization of a novel utilization of WG in clay roof tiles and to evaluate their applicability for cool roofs solutions. The tiles were manufactured with two types of recycled WG coatings with a particle size under 0,63 mm and a thickness less than 1 mm with regard to a reference tile without WG. Soda lime silica waste glass (WG1) derived from recycled hollow glass and lead silica waste glass (WG2) derived mainly from CRT of television sets were used and two shapes of tiles (curved and flat) were taken into consideration. A series of experimental tests were performed from which the following relevant conclusions can be drawn:

- WG coatings enhance the flexural strength of the tiles, showing a significant difference from both the type of the WG used and the shape of the tiles. The flexural strength was also increased up to 6.06% and 16.37%, for FWG2 and CWG2 respectively.
- Roof tiles with WG coatings are considered to be waterproof with a good level of frost resistance which could be used in cold and wet climates.
- The WG used in this study does not present a potential risk for human health and the environment, moreover, the possibility of lead leaching contained in the CRT WG was null.
- According to the colorimetric characteristics, the color of the substrate and the surface structure influence the lightness of the tiles, the application of WG2 increased the lightness by 28%.

The shape of tiles strongly influences the solar reflectance behavior; flat tiles reflect more radiation than curved tiles for the normal solar reflectance measurements of a single beam. Besides, the application of WG2 enhances the normal solar reflectance response of the tiles along the spectrum range of 350–1100 nm, with an increase of 47.5% and 27%, for flat and curved tiles respectively.

According to this study and the findings of this research, it is concluded that the application of recycled WG for roof tiles coatings complies with the standards and does not affect the performance of roof tiles. Moreover, the application of WG2 for flat clay roof tiles presents the better option for the tile's normal solar reflectance compared to

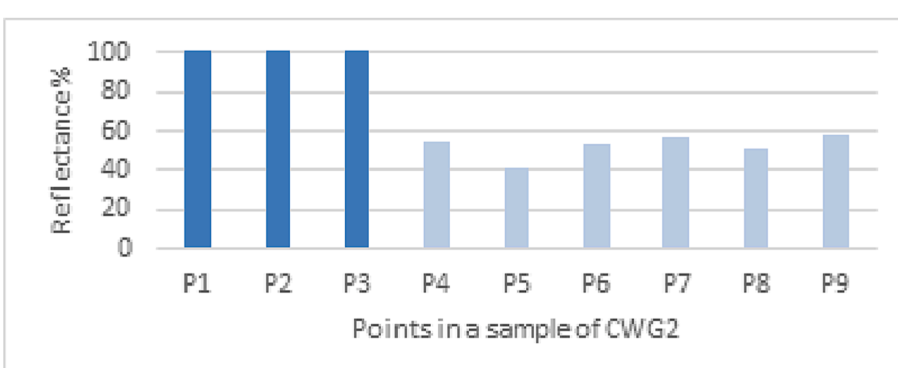

Fig. 6. Normal radiation reflectance of a curved tile sample from nine different points, the first three points (P1, P2, and P3) define the top surface line of the tile, according to Fig. 2.

WG1. This solution provides a sustainable alternative to commercial reflective materials in the market and can be considered for cool roofs applications, and subsequently contribute to the mitigation of urban heat island effect in cities. Furthermore, the use of recycled WG in the fabrication of WG coatings for roof tiles contributes to waste valorization by reducing the amount of waste discarded for landfill disposal and promoting circularity.

Once the feasibility of using WG coatings has been established, and their influence on different factors is outlined in this work, future research should be focused on performing more experiments using various WG types on an industrial scale. Additionally, further research should explore the influence of tile shape in diffuse radiation environments, analyzing global data coming from the solar radiation reflectance behavior using hemispherical distribution of detectors for different angles of radiation incidence.

CRediT authorship contribution statement

Chaimae Mourou: Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Montserrat Zamorano:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization. **Diego P. Ruiz:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization. **María Martín-Morales:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Formal analysis, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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