UNIVERSIDAD DE GRANADA

MANUFACTURING AND CHARACTERIZATION OF ECO-FRIENDLY REFLECTIVE CERAMIC COOL ROOF TILES USING WASTE GLASS TO MITIGATE THE URBAN HEAT ISLAND (UHI) EFFECT

FABRICACIÓN Y CARACTERIZACIÓN DE TEJAS CERÁMICAS REFLECTANTES ECOLÓGICAS A PARTIR DE RESIDUOS DE VIDRIO PARA MITIGAR EL EFECTO ISLA DE CALOR URBANO (ICU)

Tesis Doctoral

Para la obtención del Grado de doctor por la Universidad de Granada

Chaimae Mourou

Programa de doctorado en Ingeniería Civil (B23.56.1)

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RESUMEN

Las consecuencias del aumento de la temperatura global debido al cambio climático continúan manifestándose, lo que lleva a temperaturas elevadas en las áreas urbanas a escala local y contribuye a la creación del efecto isla de calor urbana (ICU). Además, la implementación de materiales absorbentes de calor contribuye también a agravar este fenómeno y aumenta el consumo de energía para la refrigeración en los edificios. En este contexto, las superficies frías son una de las estrategias abordadas para reducir este efecto, para lo que la comunidad científica trabaja constantemente mediante el desarrollo de materiales reflectantes adaptables. Esta estrategia de las superficies frías consiste en la inclusión de materiales reflectantes de la radiación solar para techos, paredes y pavimentos, los cuales están diseñados para reflejar las radiaciones solares y liberar rápidamente el calor absorbido, permitiendo así reducir el calor transferido hacia las edificaciones y, por ende, reducir posteriormente el consumo de energía de refrigeración. En este mismo sentido, los aspectos de impacto ambiental y sostenibilidad relacionados con el desarrollo e implementación de estos materiales han ido ganando interés en el campo de la investigación, ya que ayudan a evitar entrar en un bucle de retroalimentación con el propio calentamiento global, pues su uso ayudaría a mitigar el agotamiento de materias primas y reducir el consumo de energía y las emisiones de gases de efecto invernadero.

En este contexto, el uso de pigmentos reflectantes en la fabricación de materiales fríos ha sido ampliamente discutido en el campo de la investigación, particularmente para techos fríos, los que están ganando una atención significativa en comparación con otras aplicaciones de superficies frías. Estos pigmentos podrían alcanzar en reflectancia solar de hasta el 95% en comparación con el TiO_2, que representa el pigmento frío estándar más utilizado. Sin embargo, cabe señalar que el proceso de fabricación de estos materiales puede requerir mucha energía. Por ello, se ha fomentado la integración de materiales secundarios en la producción de materiales reflectantes sostenibles. Este enfoque tiene como objetivo reducir el uso de materias primas naturales y reducir el consumo de energía durante las fases de procesamiento y producción, lo cual abre interesantes campos de aplicación.

Por otra parte, y como recurso secundario, el uso de materiales reciclados se considera una estrategia positiva para ahorrar recursos naturales y mitigar posibles riesgos ambientales. En este sentido, el volumen de los residuos de vidrio (RV) que se reciclan a nivel mundial se

consideran muy escasos en comparación con la cantidad desechada destinada a su eliminación en vertederos. A medida que aumenta la generación de RV y persisten los problemas ambientales relacionados con los vertederos, el reciclaje de RV se ha convertido en una alternativa atractiva para la sustitución de materias primas en la industria cerámica. Aunque se han abordado estudios sobre el uso de RV en la fabricación de materiales de construcción, en particular el uso de partículas de RV como recubrimientos para tejas, aún no se ha investigado una evaluación del rendimiento de la reflectancia solar. En consecuencia, el objetivo principal de esta investigación es la caracterización del comportamiento de tejas cerámicas utilizando RV como recubrimientos reflectantes para mitigar el efecto ICU. Esta primera prueba de concepto se evaluó siguiendo un enfoque metodológico que incluyó cuatro pasos principales: (i) análisis evaluativo de la evolución científica del campo de superficies frías; (ii) una investigación experimental introductoria de tipo práctico a través de la caracterización preliminar en términos de apariencia visual, luminosidad y comportamiento de reflectancia solar de probetas de arcilla recubiertas con RV; (iii) una investigación con un enfoque a escala real incluyendo la fabricación y caracterización física, mecánica y óptica de tejas con recubrimientos RV; y (iv) finalmente, la evaluación del impacto ambiental del uso de tejas con recubrimientos RV realizando un análisis de ciclo de vida.

Los resultados obtenidos en este trabajo mostraron que se trata de un campo de investigación en constante evolución y resaltaron la viabilidad del uso potencial sustentable de RV reciclado como recubrimientos de tejas para aplicaciones de techos frescos, lo que brinda una solución basada en el uso de un recurso secundario que contribuye al desarrollo de estrategias de mitigación. del efecto isla de calor urbano (ICU). Así, La aplicación de recubrimientos de RV para tejas incrementó la reflectancia solar hasta 90%. Como resultado de la evaluación de impacto ambiental, la solución estudiada demostró disminuir el consumo de energía de refrigeración en los edificios, particularmente en zonas con condiciones severas de verano. Las zonas climáticas B4, C3 y A4 representan los casos óptimos para esta aplicación con cubiertas planas, que permite porcentajes de ahorro del 13%, 12% y 9% respectivamente. Por último, este uso específico de RV contribuye a la mitigación del agotamiento de los recursos naturales, al presentar una alternativa a las materias primas en la fabricación de recubrimientos reflectantes y ahorrar la mayor cantidad de energía necesaria para el proceso de fabricación.

En conclusión, los resultados obtenidos representan contribuciones novedosas e interesantes en el campo científico ya que proporcionan a los futuros investigadores una referencia para la evolución científica del campo de las superficies frías y la validación de una primera prueba de concepto del uso de recubrimientos de RV para mejorar el rendimiento de la reflectancia solar de las tejas.

ABSTRACT

The consequences of global temperature rise due to climate change continue to manifest, leading to elevated temperatures in urban areas at a local scale and contributing to the creation of urban heat island (UHI) effect. In the other hand, the implementation of heat absorbing materials contributes as well to the exacerbation of this phenomenon and increases the cooling energy consumption in buildings. In this context, cool surfaces present one of the strategies approached to reduce this effect, in which the scientific community is constantly working through the development of adaptable reflective materials. This strategy consists of the implementation of solar radiation reflective materials for roofs, walls and pavements, which are designed to reflect solar radiations and exhibit effective emissivity to release the absorbed heat, allowing to reduce the heat transferred into the buildings and subsequently reduce the cooling energy consumption. Moreover, the environmental impact and sustainability aspects related to the development and implementation of these materials have been gaining interest in the research field, which help to avoid entering in a feedback loop with the global warming by reducing raw materials depletion, energy consumption, and greenhouse gas emissions.

In this context, the use of reflective pigments in the fabrication of cool materials has been widely discussed in the research field, particularly for cool roofs that are gaining significant attention compared to other applications of cool surfaces. These pigments could achieve solar reflectance up to 95% in comparison with TiO_2, that represents the most widely used standard cool pigment. However, it's worth noting that the manufacturing process for these materials may require intensive energy. On the other hand, the integration of secondary materials into the production of sustainable reflective materials has been encouraged. This approach aims to reduce the use of natural raw materials and lower energy consumption during the processing and production phases.

On the other hand, as a secondary resource, using recycled materials is considered as a positive strategy for saving natural resources and mitigating potential environmental risks. In this regard, waste glass (WG) being recycled globally is deemed insufficient compared to the discarded quantity intended for landfill disposal. As the generation of WG increases and landfill-related environmental issues persist, recycling WG has become an appealing alternative used for the substitution of raw materials in the ceramic industry. Although studies on using WG in the fabrication of construction materials have been approached, the use of WG particles as coatings for roof tiles in particular, has not been investigated yet for the solar reflectance performance assessment. Consequently, the main objective of this research is the characterisation and the evaluation of the performance of ceramic roof tiles using WG as reflective coatings to mitigate UHI effect. This first proof of concept was evaluated following a methodological approach including four main steps: (i) the evaluation of the scientific evolution of the cool surfaces field; (ii) the introduction to the practical experimentation of this work through the preliminary characterization in terms of visual appearance, lightness and solar reflectance performance of clay specimens covered with WG; (iii) the real scale approach including the fabrication and the physical, mechanical and optical characterization of roof tiles with WG coatings; (iv) finally, the evaluation of the environmental impact of using roof tiles with WG coatings performing a life cycle analysis.

The results obtained in this work showed a research field in constant evolution and highlighted the feasibility of the sustainable potential use of recycled WG as roof tiles coatings for cool roof applications, which provides a solution based on the use of a secondary resource contributing to the development of mitigation strategies of the UHI effect. The application of WG coatings for tiles increased the solar reflectance for up to 90%. As a result of the environmental impact assessment, the studied solution proved to decrease the cooling energy consumption in buildings, particularly in zones with severe summer conditions. The climate zones B4, C3, and A4 represent the optimal cases for this application with flat roofs, that allows savings percentages of 13%, 12% and 9% respectively. Moreover, this specific use of WG contributes to the decrease of natural resources depletion, by presenting an alternative for raw materials in the fabrication of reflective coatings and saving as much as possible the energy required for the fabrication process.

In conclusion, the results obtained represent interesting contributions to the scientific field as it provides future researchers with a reference for the scientific evolution of cool surfaces field and a first proof of concept of using WG coatings to enhance the solar reflectance performance for tiles.

NOMENCLATURE

ASTM	American Society for Testing and Materials
CWG1	Curved tiles with waste glass type 1
CWG2	Curved tiles with waste glass type 2
CRef	Curved tiles Reference
CRT	Cathode Ray Tubes
CTE	Código Técnico de la Edificación
FRef	Flat tiles reference
FU	Functional Unit
FWG1	Flat tiles with waste glass type 1
FWG2	Flat tiles with waste glass type 2
GHG	Greenhouse Gas
GWP	Global Warming Potential
IPCC	Intergovernmental Panel on Climate Change
LCA	Life Cycle Assessment
LS	Lead Silicate
NIR	Near Infrared Radiation
PCMs	Phase Change Materials
RR	Retro Reflective
SLS	Soda Lime Silica
UHI	Urban Heat Island
UV	Ultra Violet
WG	Waste Glass
ACV	Análisis del Ciclo de Vida
GEI	Gas de Efecto Invernadero
ICU	Isla de Calor Urbana
RV	Residuos de Vidrio

INTRODUCCIÓN, MOTIVACIÓN Y OBJETIVOS

1. Marco conceptual y regulatorio

En menos de 200 años, según el Observatorio Mauna Loa, la concentración de CO_2 en la atmósfera ha aumentado un 50% debido a actividades antropogénicas, alcanzando 421,26 ppm en junio de este año 2023 (Global Climate Change, 2023). El aumento de este gas que atrapa el calor, conocido como gas de efecto invernadero (GEI), ha planteado al mundo diversos problemas relacionados con el cambio climático, siendo el efecto isla de calor urbano (ICU) una consecuencia directa. Este fenómeno impacta en la salud pública y el equilibrio ambiental creando desafíos asociados al aumento del consumo de energía de refrigeración (Conte et al., 2020). En este sentido, la adaptación a esta situación requiere el desarrollo de estrategias que ayuden a mitigar el efecto ICU (Lassinantti Gualtieri et al., 2018).

La implementación de superficies frías en áreas urbanas se considera un enfoque eficaz para aumentar la reflectancia solar y reducir la ganancia de calor de las superficies expuestas, lo que en consecuencia disminuye la salida de radiación infrarroja térmica hacia la atmósfera (Akbari & Matthews, 2012; Gao et al, 2014). El principio de esta estrategia de mitigación es utilizar materiales reflectantes en la matriz superficial de pavimentos, techos y paredes de edificios. Entre estas aplicaciones, los techos frescos han sido ampliamente abordados en la literatura (Mourou et al., 2022a), y la ganancia de calor a través del techo representa alrededor del 50-60 % de la ganancia de calor total en el edificio (Santamouris, 2016).

La aplicación de techos fríos en los edificios reduce significativamente la demanda de energía para cargas de refrigeración y mejora el confort térmico, especialmente en regiones de clima cálido (Asadi et al., 2015; Kolokotroni et al., 2018), logrando ahorros de energía que varían del 15,0 % al 35,7 % en diferentes zonas climáticas (Rawat & Singh, 2022).

Normalmente se examinan dos indicadores cruciales para evaluar el rendimiento de un techo fresco: la reflectancia solar, que mide directamente la capacidad de reflectancia del material, o/y el índice de reflectancia solar, que según ASTM E1980-11, combina tanto la reflectancia de la radiación solar como la térmica. emisividad en un solo valor. En general, existen tres métodos ampliamente utilizados para realizar las mediciones de reflectancia solar de materiales para techos (Akbari et al., 2008), incluido el uso de un piranómetro para un área medida de 10 m^2 de acuerdo con la norma E1918 para aplicaciones in situ (ASTM E1918-16, 2016). El segundo método se caracteriza por el uso de un espectrofotómetro (UV-vis NIR) para un área de 5 cm^2 siguiendo el estándar C1549. Finalmente, y principalmente para mediciones de laboratorio, se recomienda un espectrómetro para áreas de 0,1m^2 definidas por la norma E903 (ASTM E903, 2012).

Varias entidades incluyen requisitos técnicos para la reflectancia solar de los tejados en códigos y reglamentos de construcción para promover el uso de tejados frescos para conseguir la eficiencia energética y la sostenibilidad. Organizaciones como el Consejo Europeo de Techos Fríos están trabajando en el desarrollo del conocimiento científico y la investigación para promover y facilitar la integración de los techos fríos en el sector de la construcción, ya que la variedad de productos de materiales fríos implica estándares de clasificación y etiquetado para organizar su implementación en el campo. de construcción. La viabilidad de un material de tejado fresco se considera en función de varias condiciones, incluida la pendiente del tejado, la zona climática, la densidad del producto del tejado y las especificidades del edificio. Por ejemplo, según el sistema de clasificación de Liderazgo en Energía y Diseño Ambiental (LEED), el índice de reflectancia solar de una pendiente baja debe ser mayor o igual a 78, y para una pendiente pronunciada debe ser mayor o igual a 29 (Sarkis et al., 2017). Los recubrimientos que cumplen con estos requisitos otorgan créditos de certificación LEED emitidos por el US Green Building Council. Además, según ASHRAE 90.2 (ASHRAE., 2004) para edificios residenciales de poca altura, el valor mínimo del índice de reflectancia solar es 75, y para la reflectancia y emitancia solar es 0,65 y 0,75 respectivamente.

2. Uso de residuos de vidrio reciclado

Para aumentar la reflectancia solar de los materiales utilizados en cubiertas frescas, el mercado ofrece una amplia gama de pigmentos comerciales que cumplen con especificaciones técnicas, estéticas y requisitos de reflectancia solar. El rendimiento de reflectancia solar de estos materiales puede alcanzar el 95% en comparación con el TiO_2 (Z. Li, Zhao, et al., 2017). Sin embargo, el uso de estos pigmentos comerciales podría provocar impactos ambientales. En este sentido, el uso de materiales reciclados como materia prima secundaria para mejorar la reflectancia solar de los materiales de construcción proporciona una doble solución sostenible para alcanzar un compromiso entre el ahorro de energía y la disminución de la eliminación de residuos. De hecho, la industria cerámica es una fuente importante de emisiones de GEI debido a la energía utilizada en la producción de materiales provenientes de la extracción, procesamiento y fabricación (De Carli, 2007; Kuruppuarachchi et al., 2014; B. Rossi et al., 2012; F. Silva et al., 2020). Esta situación impulsas la necesidad de considerar el uso de energías renovables, e impulsar la transición energética en este sector, y utilizar los residuos reciclados como recurso secundario.

El vidrio es un material altamente reutilizable debido a su presencia generalizada en los residuos sólidos como residuo no biodegradable (Sarkis et al., 2017). Según las Naciones Unidas, cada año se producen en todo el mundo alrededor de 14 millones de toneladas de residuos de vidrio (RV), y esta cifra está aumentando cada año (Topçu & Canbaz, 2004). Por lo tanto, se tienen muy en cuenta las opciones de reciclaje de estos residuos para evitar la última opción en la gestión del flujo de residuos, que es la eliminación, definida por la Directiva 2008/98/CE y la Directiva 2018/850 (UE., 2008; 2018).

En este sentido, la industria de la construcción es el principal sector que prueba y utiliza RV como sustitución de materia prima, la cual ha sido utilizada y probada en diferentes porcentajes y tamaños (Cardoso de Souza-Dal Bó et al., 2021; R. V. Silva et al., 2017). El uso de RV en materiales de construcción mostró resultados prometedores en la disminución del consumo de energía, la extracción de materias primas y la eliminación de residuos, además de mejorar el desempeño mecánico, físico y ambiental de los materiales. Además, demostró ser una solución eficaz para aliviar el efecto isla de calor urbano (Peng et al., 2023).

3. Evaluación de impacto ambiental

De hecho, el procesamiento de materiales reciclados genera menos cargas ambientales que la extracción y procesamiento de materias primas vírgenes (Andreola, Barbieri, Corradi, Ferrari, et al., 2007). En este sentido, la evaluación del impacto ambiental de estos materiales es imprescindible. Según (Suppa et al., 2022), la herramienta de apoyo más utilizada para alcanzar las 100 Ciudades Climáticamente Neutrales para 2030 es el análisis del ciclo de vida (ACV), cuya mayoría de indicadores se centran en el consumo de energía y las emisiones de carbono.

ACV es una herramienta cuantitativa utilizada para mejorar la sostenibilidad ambiental de un producto (Matthews et al., 2014), que se basa en una metodología estructurada definida por la norma ISO 14040 que abarca la definición de objetivos y alcance, la realización de un análisis de inventario del ciclo de vida, la realización de una evaluación del impacto del ciclo de vida y concluir con una interpretación del ciclo de vida (ISO 14040, 2006). Estos pasos se siguen para evaluar la fase de producción y/o la fase de uso de un producto o servicio, como parte de la fase de diseño de edificios para apoyar el proceso de toma de decisiones. Por tanto, la realización de un estudio ACV permite cuantificar la disminución del consumo de energía de refrigeración en los edificios como resultado de la implementación de cubiertas frías. Este análisis permite resaltar las reducciones del impacto del Potencial de Calentamiento Global asociado con el ahorro de energía y proporcionar información clave sobre el consumo energético operativo.

De acuerdo con lo anterior, los techos frescos se consideran uno de los enfoques viables de las estrategias de superficies frías para ayudar a reducir el efecto ICU y disminuir el consumo de energía de refrigeración en los edificios. Además, el uso de residuos reciclados en la fabricación de materiales frescos está siendo muy fomentado en el campo de la investigación, promoviendo aún más la valorización de los materiales de desecho, lo que se alinea con los principios de la economía circular establecidos por el acuerdo verde europeo que apunta a la transición. de la economía europea de un modelo lineal a uno circular.

4. Motivación y objetivos

Aunque se han abordado estudios sobre el uso de RV en la fabricación de materiales de construcción, el uso de partículas de RV como recubrimientos para tejas en particular aún no se ha investigado para la evaluación del rendimiento de la reflectancia solar.

En consecuencia, el objetivo principal de esta investigación es caracterizar y valorar el desempeño de tejas cerámicas utilizando RV como recubrimientos reflectantes para mitigar el efecto ICU. Para lograr este objetivo general se han definido los siguientes objetivos secundarios:

i. Analizar la evolución científica de las estrategias usadas para las superficies frías que incluye:
    - Realizar un estudio bibliométrico dual basado en el mapeo científico y el análisis del desempeño.
    - Realizar una revisión de las estrategias y materiales aplicados para superficies frías de acuerdo con los resultados del mapeo científico.

ii. Generar un estudio preliminar para la caracterización del comportamiento de reflectancia espectral de probetas de baldosas con recubrimientos de RV que incluya:
    - El procesamiento y la caracterización química de las materias primas utilizadas.

- La fabricación de las probetas de arcilla con recubrimientos de RV.
- La identificación del grado de luminosidad y reflectancia espectral especular.
- La evaluación de la influencia de diferentes parámetros sobre las probetas incluyendo la temperatura de cocción, el tiempo de mantenimiento, el tipo de RV utilizado y el espesor de los recubrimientos.

iii. Fabricar tejas cerámicas a escala real utilizando recubrimientos de RV para realizar una caracterización física, mecánica y óptica que incluye:

- La fabricación a Escala real de tejas (planas y curvas) con revestimientos de RV a escala de laboratorio.
- La caracterización de las propiedades físicas, mecánicas y ópticas de las tejas.
- La evaluación de la influencia de la forma de las tejas y del tipo de RV utilizado para los revestimientos sobre las propiedades de las tejas.

iv. Analizar el impacto ambiental de las fases de producción y uso de tejas cerámicas con recubrimientos de RV mediante ACV que incluye:

- La recogida y tratamiento de los datos relativos a la fabricación de las baldosas para la identificación del impacto medioambiental de la fase de producción.
- La evaluación del consumo energético de refrigeración mediante tejas planas y curvas con recubrimientos de RV, para cubiertas planas e inclinadas, para evaluar el impacto ambiental de la fase de uso.

5. Metodología e hipótesis

Los objetivos secundarios antes mencionados se desarrollaron en una secuencia metodológica que ha permitido perseguir el objetivo principal de este trabajo. Para llevar a cabo esta investigación, se emplearon cuatro pasos principales, como se muestra en laFigura 1. En dicha figura se muestran las principales herramientas metodológicas usadas para la consecución de los objetivos secundarios enunciados en el apartado anterior.

Los diferentes pasos han comenzando con un análisis teórico del campo de investigación utilizando las plataformas de datos Scopus y Web Of Science para los registros de la investigación y una herramienta de mapeo científico para el procesamiento de datos desarrollada en el Capítulo 1. Este paso inicial permitió identificar el status quo del campo de investigación y determinar las motivaciones que subyacen al objetivo principal de este trabajo.

El segundo paso se ha basado en la evaluación de la primera prueba de concepto introducida por esta investigación, que es el uso de recubrimientos de RV para baldosas, mediante la realización de un estudio preliminar desarrollado en el Capítulo 2. La preparación de los materiales, el proceso de fabricación, y la caracterización de las probetas se realizó en el laboratorio de materiales del Departamento de Edificación de la Universidad de Granada. Este paso permitió identificar la influencia de ciertos parámetros en la apariencia visual y propiedades ópticas de las muestras.

Posteriormente se actualizó la caracterización a escala real de las tejas para el tercer paso de la investigación desarrollada en el Capítulo 3, que incluyó la fabricación de las tejas planas y curvas, y la realización de pruebas de laboratorio utilizando diversos equipos y condiciones de ensayo. Después de la evaluación de las especificaciones técnicas de las tejas y más particularmente de la influencia de la forma de las tejas y del tipo de RV en el comportamiento de la reflectancia solar, el estudio se dirigió a la evaluación del impacto ambiental de estas tejas.

En este paso final, el avance de la investigación condujo a la evaluación ambiental de la solución propuesta desarrollada en el Capítulo 4, para incluir la valoración de otro aspecto no menos significativo que el resto de especificaciones. Por este motivo, se utilizaron herramientas de ACV y simulación de consumo energético para permitir la evaluación del impacto ambiental de la producción y el uso de tejas con recubrimientos de RV.

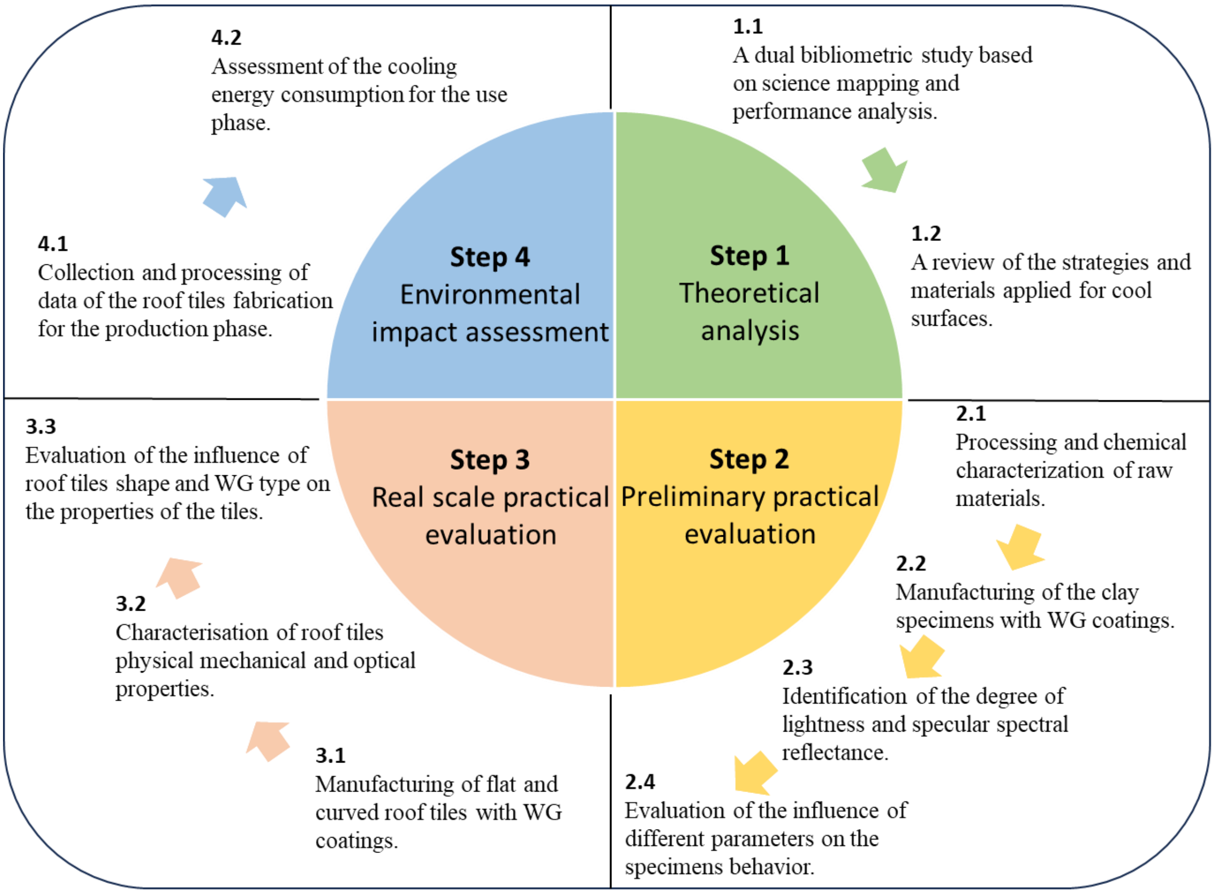


Figura 1. Los pasos del enfoque metodológico seguido en esta investigación en función de los objetivos secundarios

De acuerdo con este enfoque metodológico, y en relación con los objetivos de investigación anteriores formulados a partir del estudio preliminar del estado del arte, se planteó probar las siguientes hipótesis:

- El campo de investigación de superficies frías está en constante evolución como resultado del continuo desarrollo de los materiales empleados.
- La apariencia visual y el rendimiento óptico de las muestras de arcilla con recubrimientos RV están influenciados por varios parámetros, entre los que se consideran, la temperatura de cocción, el tiempo de mantenimiento, el tipo de RV y el espesor de los recubrimientos.
- El RV reciclado podría utilizarse como revestimiento sin afectar el rendimiento de las tejas planas y curvas.
- Las propiedades ópticas de las baldosas con revestimiento RV están influenciadas por la forma y el color de las baldosas (el sustrato).

- La aplicación de revestimientos RV sobre tejas planas y curvas mejora su comportamiento de reflectancia solar.
- El uso de RV reciclado tiene un impacto ambiental en la fabricación de tejas con revestimientos de RV, que se considera no muy elevado.
- Se espera que el consumo de energía de refrigeración utilizando tejas con revestimiento RV sea menor que en el caso convencional.

INTRODUCTION, MOTIVATION AND OBJECTIVES

1. Conceptual and regulatory framework

In less than 200 years, the concentration of CO_2 in the atmosphere has raised by 50% due to anthropogenic activities, measured at Mauna Loa Observatory, reaching 421.26 ppm in June of this year 2023 (Global Climate Change, 2023). The rise of this heat-trapping gas, known as greenhouse gas (GHG), has introduced the world to various climate change issues, with urban heat island (UHI) effect being a direct consequence. This phenomenon influences the public health and the environmental balance creating challenges associated to the increase of cooling energy consumption (Conte et al., 2020). In this sense, the adaptation to this situation claims the development of strategies that will help to mitigate the UHI effect (Lassinantti Gualtieri et al., 2018).

The implementation of cool surfaces in urban areas is considered an effective approach to increase the solar reflectance and reduce the heat gain by the exposed surfaces, which consequently decreases the outflow of thermal infrared radiation into the atmosphere (Akbari & Matthews, 2012; Gao et al., 2014). The principle of this mitigation strategy is to use reflective materials in the surface matrix of pavements, buildings' roofs and walls. Among these applications, cool roofs have been highly addressed in literature (Mourou et al., 2022a), with the heat-gain through the roof representing about 50-60% of the total heat gain in the building (Santamouris, 2016).

The application of cool roofs in buildings significantly reduces energy demand for cooling loads and improves the thermal comfort particularly in hot climate regions (Asadi et al., 2015; Kolokotroni et al., 2018), achieving energy savings varying from 15.0% to 35.7% in different climate zones (Rawat & Singh, 2022).

Two crucial indicators are typically examined to assess the performance of a cool roof: solar reflectance, which directly measures the material's reflectance capacity, or/and the solar reflectance index, which according to ASTM E1980-11, it combines both solar radiation reflectance and thermal emittance into a single value. In general, there are three widely used methods to follow for the solar reflectance measurements of roofing materials (Akbari et al., 2008), including the use of a pyranometer for a measured area of $10m^2$ according to E1918 standard for in-situ applications (ASTM E1918-16, 2016). The second method is characterized by the use of a spectrophotometer (UV–vis NIR) for an area of $5cm^2$ following the C1549 standard. Finally, and mainly for laboratory measurements, a spectrometer is recommended for areas of $0.1m^2$ defined by the E903 standard (ASTM E903, 2012).

Several entities include technical requirements for the solar reflectance of roofs in building codes and regulations to promote the use of cool roofs for energy efficiency and sustainability. Organizations such as the European Cool Roofs Council are working on developing the scientific knowledge and research to promote and facilitate the integration of cool roofs in the building sector as the variety of cool materials products implies standards for rating and labelling to organize their implementation in the construction field. The feasibility of a cool roof material is deemed by several conditions including the slope of the roof, the climatic zone, the density of the roofing product and the building specificity. For instance, according to the Leadership in Energy and Environmental Design (LEED) rating system, the solar reflectance index of a low slope must be greater than or equal to 78, and for a steep slope must be greater than or equal to 29 (Sarkis et al., 2017). Coatings that meet these requirements are giving LEED certification credits issued by the US Green Building Council. Moreover, according to ASHRAE 90.2 (ASHRAE., 2004) for low-rise residential buildings, the minimum value of solar reflectance index is 75, and for solar reflectance and emittance is 0.65, and 0.75 respectively.

2. Using recycled waste glass

To increase the solar reflectance of materials used for cool roofs, the market affords a broad range of commercial pigments fulfilling technical specifications, aesthetical and solar reflectance requirements. The solar reflectance of these materials can reach 95% compared to TiO_2 (Z. Li, Zhao, et al., 2017), that represents the most widely used standard cool pigment. However, using these commercial pigments could induce environmental impacts. In this sense, the use of recycled materials as a secondary resource to enhance the solar reflectance of construction materials, provides a twofold sustainable solution to reach a compromise between saving energy and decreasing waste disposal. In fact, the ceramic industry is a major source of GHG emissions due to the energy used in the production of materials from extraction, processing, and manufacturing (De Carli, 2007; Kuruppuarachchi et al., 2014; B. Rossi et al., 2012; F. Silva et al., 2020). This situation urged the need to consider the use of renewable energies, and encourage the energy transition in this sector, and to use recycled waste as a secondary resource.

Glass is a highly reusable material due to its widespread presence in solid waste as a non-biodegradable waste (Sarkis et al., 2017). According to the United Nations, about 14 million tons of waste glass (WG) are produced globally every year, and this number is increasing each year (Topçu & Canbaz, 2004). Hence the recycling options of this waste is highly considered to avoid the last option in the waste stream management, which is disposal, defined by Directive 2008/98/EC and Directive 2018/851 (UE., 2008; 2018).

In this regard, the construction industry is the main sector testing and using WG as a raw material substitution which has been used and tested in different percentage and size (Cardoso de Souza-Dal Bó et al., 2021; R. V. Silva et al., 2017). Using WG in construction materials showed promising results in decreasing energy consumption, extraction of raw materials and disposal of waste, in addition to improving the mechanical, physical, and environmental performance of materials. Moreover, it proved to be an efficient solution to alleviate the UHI effect (Peng et al., 2023).

3. Environmental impact assessment

In fact, the processing of recycled materials generates less environmental loads than the extraction and processing of virgin raw materials (Andreola, Barbieri, Corradi, Ferrari, et al., 2007). In this sense, the assessment of the environmental impact of these materials is a must. According to (Suppa et al., 2022), the most used support tool to achieve the 100 Climate-Neutral Cities by 2030 is the life cycle assessment (LCA) analysis, which most indicators focus on energy consumption and carbon emissions.

The LCA is a quantitative tool used to enhance the environmental sustainability of a product (Matthews et al., 2014), which relies on a structured methodology defined by ISO 14040 standard that encompasses defining goals and scope, conducting a life cycle inventory analysis, performing a life cycle impact assessment, and concluding with a life cycle interpretation (ISO 14040, 2006). These steps are followed to assess the production phase and/or the use phase of a product or a service, as a part of the design phase of buildings to support the decision-making process. Hence, performing an LCA study allows to quantify the decrease of cooling energy consumption in buildings resulting from the implementation of cool roofs. This analysis allows to highlight the reductions of the Global Warming Potential impact associated with energy saving and provide key information regarding the operational energy consumption.

According to the above, cool roofs are considered as one of the viable approaches of the cool surfaces strategies to help reduce the UHI effect and decrease the cooling energy consumption in buildings. Moreover, using recycled waste in the fabrication of cool materials was highly encouraged in the research field, promoting to the further extent the valorisation of waste materials, which aligns with the principles of the circular economy established by the European green deal that aims to the transition of the European economy from a linear to a circular model.

4. Motivation and objectives

Although studies on using WG in the fabrication of construction materials have been approached, the use of WG particles as coatings for roof tiles in particular has not been investigated yet for the solar reflectance performance assessment.

Consequently, the main objective of this research is to characterize and evaluate the performance of ceramic roof tiles using WG as reflective coatings to mitigate UHI effect. In order to achieve this objective, the following secondary objectives were defined:

i. To analyse the scientific evolution of cool surfaces strategies which includes:
   - Conducting a dual bibliometric study based on science mapping and performance analysis.
   - Conducting a review of the strategies and materials applied for cool surfaces according to the science mapping results.

ii. To conduct a preliminary study for the characterisation of the spectral reflectance performance of tiles specimens with WG coatings which includes:
   - The processing and the chemical characterization of raw materials used.
   - The manufacturing of the clay specimens with WG coatings.
   - The identification of the degree of lightness and specular spectral reflectance.
   - The evaluation of the influence of different parameters on the specimens including the firing temperature, the holding time, the type of WG used and the thickness of the coatings.

iii. To fabricate ceramic roof tiles at real scale using WG coatings in order to perform a physical, mechanical and optical characterisation which includes:
   - The manufacturing of the real shape of roof tiles (flat and curved) with WG coatings at laboratory scale.
   - The characterisation of the roof tiles' physical, mechanical and optical properties.
   - The evaluation of the influence of roof tiles shape and WG type used for the coatings on the properties of the tiles.

iv. To analyse the environmental impact of the production and the use phases of ceramic roof tiles with WG coatings through an LCA which includes:
   - The collection and processing of the data concerning the fabrication of the tiles for the identification of the environmental impact of the production phase.
   - The assessment of the cooling energy consumption using flat and curved roof tiles with WG coatings, for flat and pitched roofs, to evaluate the environmental impact of the use phase.

5. Methodology and hypothesis

The aforementioned secondary objectives were developed in a methodological sequence allowing the pursuit of the main objective of this work. To conduct this investigation, four main steps were employed that are shown in Figure 1, starting with a theoretical analysis of the research field using Scopus and Web Of Science data platforms for the investigations records and a science mapping tool for the data processing developed in Chapter 1. This initial step allowed the identification of the status quo of the research field and the determination of the motivations underlying the main objective of this work.

The second step was based on the evaluation of the first proof of concept introduced by this research, which is the use of WG coatings for tiles, through the demonstration of a preliminary study developed in Chapter 2. The preparation of the materials, the manufacturing process, and the characterisation of the specimens were carried out within the laboratory of materials of the Department of Building Construction at the University of Granada. This step allowed the identification of the influence of certain parameters on the visual appearance and optical properties of the specimens.

Subsequently the characterisation was upgraded to the real scale of roof tiles for the third step of the research developed in Chapter 3, which included the manufacturing of the flat and curved roof tiles, and the performance of laboratory tests using various equipment and tests conditions. After the evaluation of the technical specifications of the roof tiles and more particularly the influence of the tiles shape and the type of WG on the solar reflectance performance, the study was led to the evaluation of the environmental impact of these tiles.

In this final step, the research progress led to the environmental evaluation of the proposed solution developed in Chapter 4, to enclose the assessment of another aspect no less significant than the other specifications. For this reason, the LCA and energy consumption simulation tools were used to allow the evaluation of the environmental impact of the production and the use of roof tiles with WG coatings.

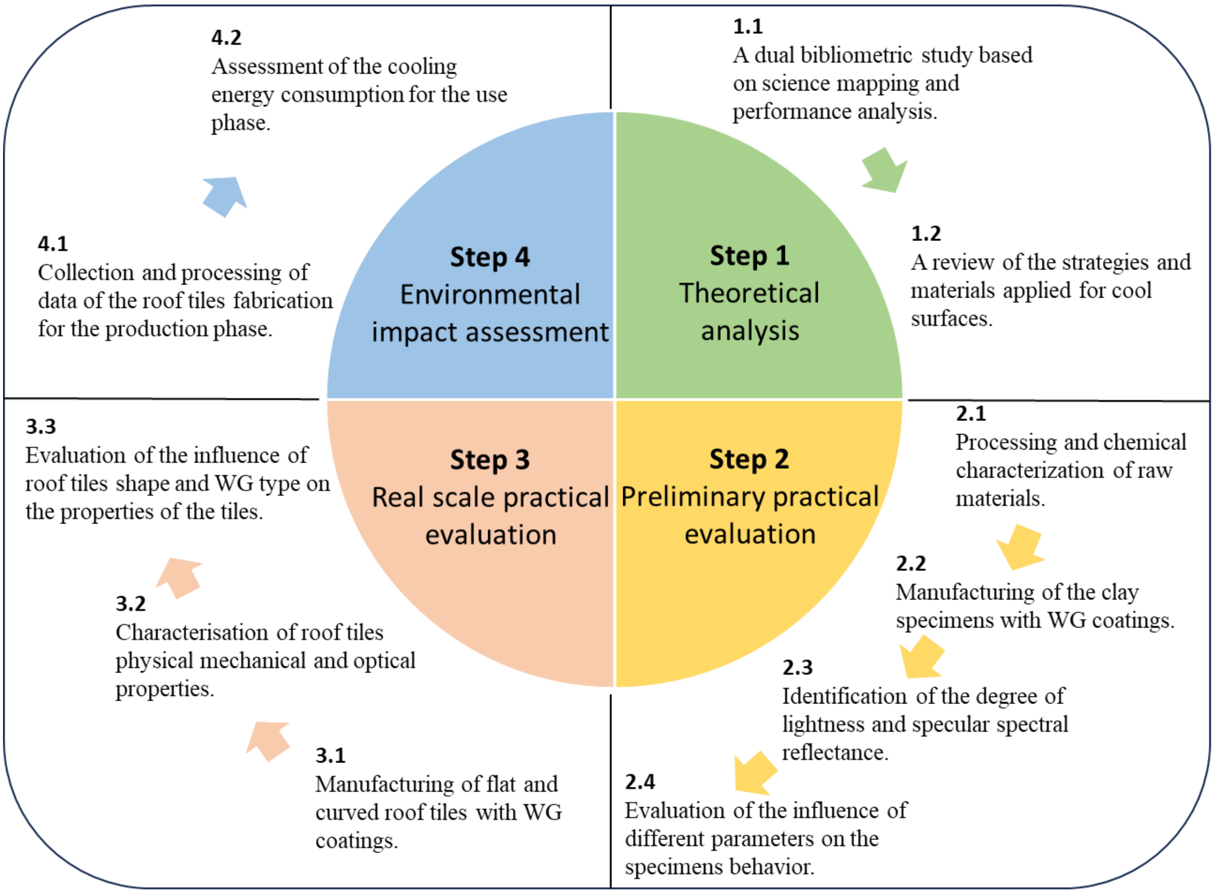


Figure 1. The steps of the methodological approach followed in this investigation based on the secondary objectives

According to this methodological approach, and related to the previous research objectives formulated from the preliminary study of the state of the art, the following hypothesis were set to be tested:

- The cool surfaces research field is constantly evolving as a result of the continual development of materials employed.
- The visual appearance and optical performance of clay specimens with WG coatings are influenced by several parameters including the firing temperature, holding time, type of WG, and thickness of the coatings.
- Recycled WG could be used as coatings without affecting the performance of the flat and curved roof tiles.
- The optical properties of tiles with WG coatings are influenced by the shape and color of the tiles (the substrate).
- The application of WG coatings on flat and curved roof tiles enhances their solar reflectance performance.
- The use of recycled WG have a small environmental impact in the manufacturing of roof tiles with WG coatings.
- The cooling energy consumption using roof tiles with WG coatings is expected to be less than the conventional case.

SECTION 1. BACKGROUND

CHAPTER 1. Analysis of the scientific evolution of cool surfaces strategies

1. Introduction

The consumption of the world's energy resources hit 75%, even though cities only cover 2% of the planet's surface (Gago et al., 2013). A portion of this energy is dissipated as heat due to anthropogenic activities, which increases the ambient temperatures in urban areas. This phenomenon was labelled as the Urban Heat Island (UHI) effect by meteorologists more than a century ago, and it is the result of the heat trapped in urban areas due to the increase of the ambient temperature, as a consequence of the high concentration of greenhouse gases in the atmosphere (Mohajerani et al., 2017; Rosenfeld et al., 1995; Zinzi & Agnoli, 2012). Climate change is the main concern occurring due to the non-balanced use of resources, leading to environmental and public health challenges (Mohajerani et al., 2017), which contributes to the increase in cooling energy consumption (Akbari & Kolokotsa, 2016; Rosenfeld et al., 1995; Zinzi & Agnoli, 2012). It can therefore be stated that the UHI effect is an environmental problem that requires theoretical and practical studies to mitigate its impact (Akbari et al., 1992).

In order to palliate the UHI effect, a growing number of studies and investigations have been conducted to develop mitigation strategies that can be implemented in urban spaces and buildings. Several solutions have been developed, including urban geometry reshaping (Gago et al., 2013), designing green and cool roofs (Chatterjee et al., 2019; Kolokotsa et al., 2013), using permeable, porous, water-retentive, and cool pavements (Santamouris, 2013), incorporating green spaces into the urban landscape, and utilizing water and wind for cooling effects (Akbari et al., 2016; Mohajerani et al., 2017; Qin, 2015). These solutions could yield a median reduction in the air temperature between 1.8 and 2.1 K (Lai et al., 2019). Furthermore, the combination of different measures could be more effective (Zhu et al., 2021), and the choice of the optimal strategy depends on the regional atmospheric and geographic specifications of the urban environment (Chatterjee et al., 2019; J. Yang et al., 2015). Among the cited UHI mitigation strategies, cool surfaces have emerged as a viable solution (Hosseini & Akbari, 2016; Roman et al., 2016), which basically refer to surfaces with reflective materials and coatings that reflect the solar energy radiation hitting buildings envelopes and urban areas (Pisello, 2017), including roofs, facades, and pavements. Cool surfaces can reduce the thermal infrared radiation outflow in the atmosphere, as well as the temperature and the solar heat gain (Akbari & Matthews, 2012; Gao et al., 2014). In fact, it has been proven that the implementation of cool surfaces to replace dark and highly absorptive materials during routine maintenance increases the albedo over time (Rosenfeld et al., 1995). These materials come in a huge variety and include natural materials, artificial cool coatings, and non-white high-albedo materials (M. Z. Chen et al., 2009; Lee Shoemaker, 2003). In addition to the fabrication process and the conditions, the thickness, the particle size, the substrate characteristics and other materials, are all key parameters that could affect the optical and thermal properties of a cool material including the albedo, permeability, conductivity, solar reflectance, and emissivity (M. Z. Chen et al., 2009; Smith et al., 2017).

The thermal performance of a material is mainly evaluated by the albedo (solar radiation) and emissivity (longwave radiation) (M. Z. Chen et al., 2009). The solar reflectance potential of cool materials initially relied on their whiteness, which promoted the use of white paints and light-color aggregates (Bretz et al., 1998). Subsequently, the research field progressed toward enhancing the near infrared (NIR) reflectance of cool materials, as it represents almost 52% of the electromagnetic spectrum of light (i.e., from 700 to 2500 nm) (Muniz-Miranda et al., 2019). Thus, novel methodologies and techniques have been considered to cover a wide range of the solar reflectance in order to enhance the performance of cool materials. The cool surface strategy has started an extensive series of studies concerning pigments that has created an industry of geoengineering and chemical solutions for the development of cool materials to enhance the solar reflectance performances. The integration of pigments in cool materials has recently been discussed in detail within research and industrial contexts (Rosati et al., 2021b). With regard to the aesthetic requirements of a design, selective pigments have been developed to maintain the optical color desired on top of the material while achieving important NIR reflectance results (Xie et al., 2019). The pigments range from organic, to complex inorganic color mixed-metal oxides (Levinson et al., 2005a, 2005b). These substances have demonstrated high solar reflectance up to 95%, compared to TiO_2 (Meenakshi & Selvaraj, 2018; Raj et al., 2015, 2017, 2019; Xiang & Zhang, 2018). In addition to cool pigments, other solutions for cool surfaces have emerged to improve urban climate conditions and energy consumption, such as retroreflective (RR) and phase change materials (PCMs). Independently of the incidence direction, retro-reflectivity refers to the capacity of a surface to reflect an incoming light beam to a surface back towards its source (F. Rossi et al., 2016). RR materials have been demonstrated to be effective in several studies in terms of the solar radiation reflectance beyond urban canyons and canopies (F. Rossi et al., 2014, 2016; Sakai et al., 2011a). On the other hand, PCMs have the ability to change their physical characteristics as a consequence of heat release or absorption (Mapston & Westbrook, 2010). In recent decades, a variety of PCMs have been investigated as dynamic components in structures (Kosny PhD & Kossecka PhD, 2013). The implementation of PCMs in the matrix of roof finishing materials decreases the flux of the roof heat gain by 54%, compared to the cool roof (Roman et al., 2016), and it helps to compensate for the effect of the thermal stress generated by the latter (Saffari et al., 2018). Moreover, PCMs can be used to regulate the indoor thermal comfort in summer and reduce the heating penalty during winter, more effectively than cool paints (Chung & Park, 2016; Pisello et al., 2017; Y. K. Yang et al., 2017, 2019). Sustainable adaptation has been a parallel concern in the development of cool material solutions. In this context, the use of recycling materials to save energy and natural resources, and to enhance the solar reflectance, presents a promising eco-friendly strategy (J. Yang et al., 2015). For instance, full body porcelain aggregate from waste tiles has been used as a cool pavement coating that exhibits important thermal performance values compared to asphalt pavement, with a solar reflectance of 0.49 at near infrared region, and a surface temperature reduction up to 6.4 ◦C during the peak periods (Anting et al., 2017a). In addition, the use of recycled glass cullet in the fabrication of a sustainable asphalt roof shingle improved the solar reflectance (Kiletico et al., 2015).

Considering the evolution of materials applied in cool surfaces, the current objective of this study is to perform a bibliometric analysis to review the scientific development of this solution in the mitigation of the UHI effect. Section 2 presents the research methodology employed. Section 3 describes the collection process for the materials and the bibliometric evaluation, provides a descriptive analysis of the findings and examines the most important contributions of the studies identified previously. Finally, Section 4 provides the most important conclusions of this study, which contribute to the existing body of knowledge by highlighting the trends in the research field of cool surfaces for building envelopes and urban areas, as well as recommending research areas for future studies.

2. Methodology

A dual bibliometric study based on science mapping and performance analysis was conducted to achieve the objective of this work. The science mapping analysis software SciMAT was used to obtain the necessary patterns and bibliometric measures (Cobo et al., 2011; Noyons et al., 1999). Science mapping visualizes, analyzes, and models a broad range of scientific and technological activities, and it follows a general workflow of data retrieval, data preprocessing, network extraction, network normalization, mapping, analysis, visualization, and finally the interpretation of the results (Cobo et al., 2011). Furthermore, the performance analysis as a complementary methodology uses different bibliometric measures and indicators to complement the visualization results and to help identify the impacts and productivities of the themes in the research field. Considering the results obtained, a number of publications were selected in order to conduct a review on cool surfaces evolution. Recent academic findings and limitations are included in the literature review section in order to enrich the discussion due to the constant evolution of the field.

2.1.Sample definition and steps for the data collection

This review addresses the intersection of the two concepts: the UHI effect and the different types of cool surfaces applied. An extensive research was carried out employing keywords linked to both concepts, and to the reflectance (Figure 2). Therefore, the first stage of the data collection was performed using the field, "Title/Abstract/Keyword", through the following keywords and search strings: "Heat island" OR "Reflect*" AND "Cool surface*", "Cool facade*", and "Cool roof*" AND "Cool pavement*", within the Web of Science Core Collection and Scopus databases. The asterisk is used at the end of keywords to broaden the research. As a result, 982 publications were found, 347 of which were excluded after the deduplicating and cleaning of the raw data. After reading the abstracts, another 121 publications were excluded because they were not aligned with the purpose of this research. Finally, the bibliometric study was performed with 514 publications.

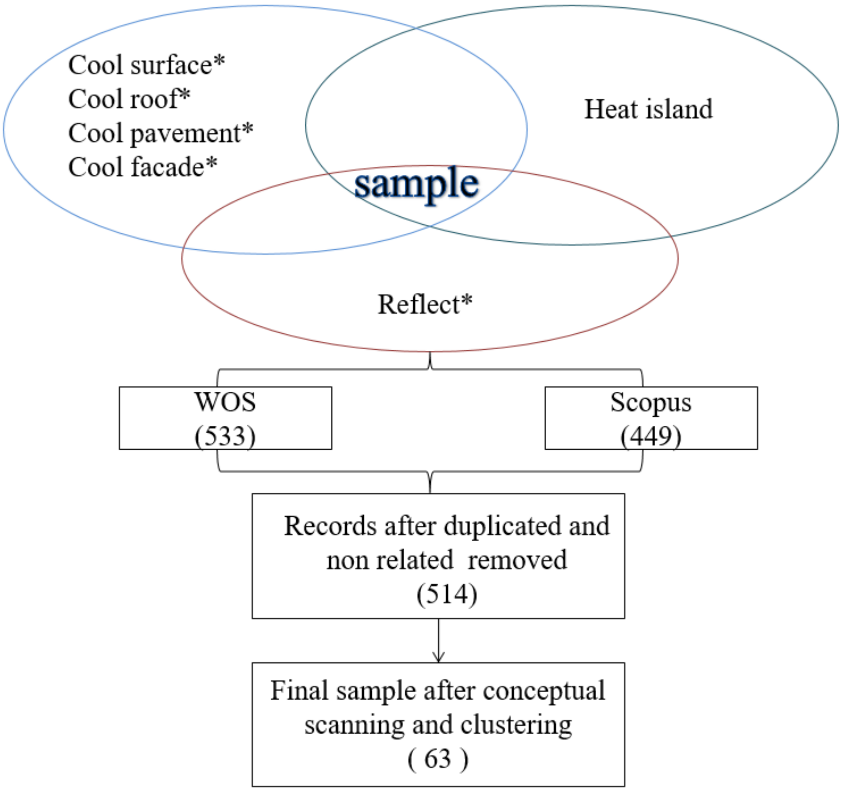


Figure 2. Data collection flowchart used in Web of Science Core Collection and Scopus database

The next step was dedicated to the restriction of the data and the refinement of the sample on the basis of a conceptual approach. According to the SciMAT visualization results, it was possible to follow the internal links in the cluster networks of the concepts with the highest potentials for reviewing, which returned 63 records. This process of data collection is described in the flowchart above (Figure 2). The analysis of these publications was the basis of the literature review of the evolution of cool surfaces through the materials applied to mitigate the UHI effect.

2.2.Systematic literature review

The analysis of the 514 documents selected for the systematic literature review resulted in the following data: total numbers and years of publications; and authors with the highest contributions to the field based on the number of publications, sources, and journals. Furthermore, strategic diagrams, thematic networks, overlay graphs, and evolution maps were used to demonstrate the links between each theme of the strategic diagrams, the keywords, and their interconnections. These were also used to identify the research motor themes, the highly developed and isolated themes, the emerging or declining themes, and the basic and transversal themes, and consequently to trace the evolution of these themes along the studied period

(Callon et al., 1983, 1991; Cobo et al., 2015). The sample of academic publications processed through this science mapping analysis was dependent on the specific input conditions, such as the unit of analysis and the keywords.

3. Results and discussions

The aim of this section is to assess the collected material that was released during the period between 1995 and 2020 using quantitative and qualitative methods. A descriptive analysis was conducted through: (i) An analysis of the evolution of the numbers of documents; and (ii) the main sources of the publications with the most prolific authors. Science mapping and visualization were subsequently conducted to obtain an assessment of the evolution of the cool-material-application domain. Finally, a literature revision of the materials applied for cool surfaces was developed, which led to five analytical sections employed for the evaluation of the materials: (i) Pigments; (ii) RR materials; (ii) PCMs; (iv) Ceramic materials; and (v) Glass.

3.1.Descriptive analysis

3.1.1. Evolution of number of documents

Since the first article identified in these databases was published in 1995, the time horizon used in this study was from 1995 to 2020. To analyze the trends and patterns in the publications, three periods were identified (1995–2001, 2002–2010, and 2011–2020) on the basis of the main turning points and milestones in the evolution of cool surfaces. The results of the contributions for the three periods identified are summarized below.

The first period (1995–2001): In this period, the contribution of the academic literature to the topic was very poor, with only five publications found (Figure 3). However, the period was marked by promoting cool roofs through building codes. In fact, starting in 1999, several energy-building standards adopted cool roof credits or requirements, such as ASHRAE 90.1, ASHRAE 90.2, the International Energy Conservation Code, and California’s Title 24.

The second period (2002–2010): This period is characterized by the rising concern with regard to the topic, which coincided with the third assessment report of the Intergovernmental Panel on Climate Change, which highlights the increase in greenhouse emissions and the global average surface temperature in the 20th century by 0.6 °C. The number of publications increased considerably (to 58) during this period (Figure 3).

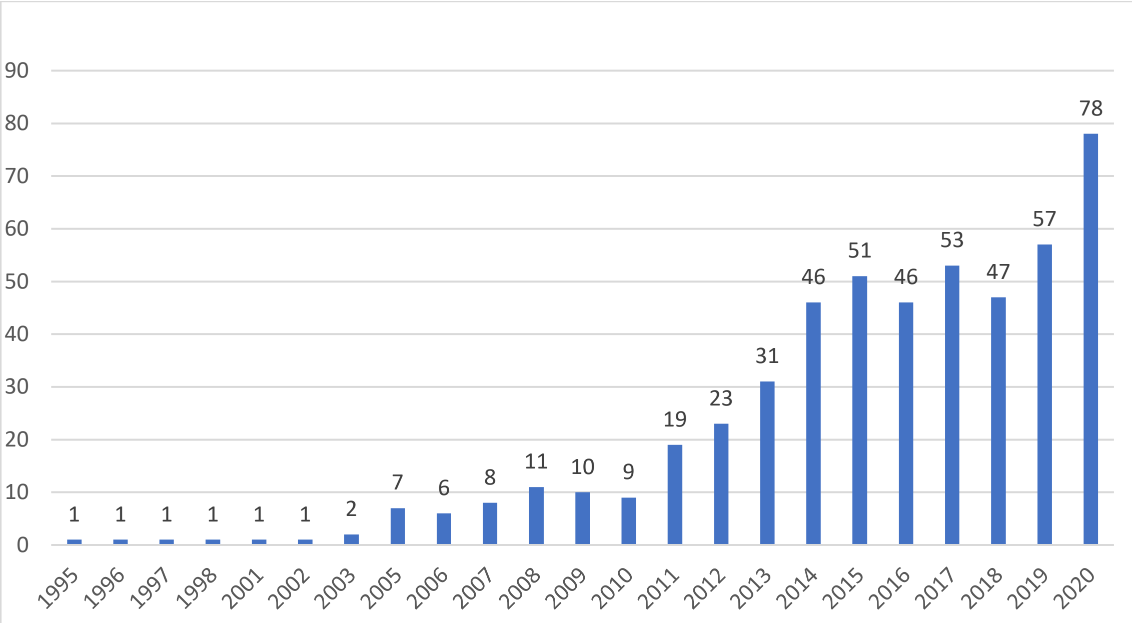


Figure 3. Number of documents per year

The third period (2011–2020): The cool surface strategy was a rapidly growing research topic during this period. In fact, it is by far the most prolific in terms of publications, with 87.74% of the records published during this time (Figure 3). This period coincides with the foundation of the European Cool Roofs Council, which seeks to develop knowledge and research regarding cool roof technology and promotes the use and implementation of this technology in Europe.

Considering the significant evolution of the number of academic documents in this field, an increasing interest in this topic has been clearly observed.

3.1.2. Main source publications

According to Figure 4, the nine most prolific journals account for 40% of the total records of the sample principally considered. It is possible to conclude that the most common aspect of the journals considered is related to energy efficiency and buildings, as well as to the science and technology of solar energy applications. In fact, *Energy and Buildings* has the dominant share, with 16.53% of the published records, followed by *Solar Energy* (6.19%), and *Solar Energy Materials and Solar Cells* (4.12%).

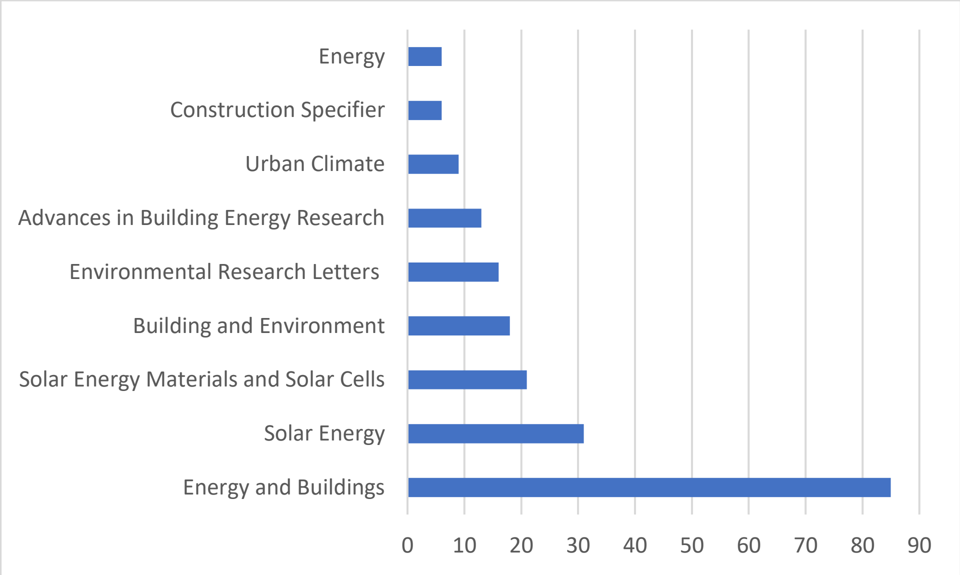


Figure 4. Distribution of journals by number of publications

Concerning the publications authorship, it is shown in Table 1 that Akbari, H., who is affiliated with United States and Canada, has made the most contributions to the field, with 36 publications, followed by Pisello, A.L. (33), and Levinson, R. (28). Most of the publications are affiliated with Italian universities (97 papers), followed by the United States and Canada, with 74 and 36 publications, respectively.

Table 1. Authors with at least 10 publications in databases and country affiliation

Order	Authors	N. Papers	Affiliation
1	Akbari, H.	36	U.States Canada
2	Pisello, A.L.	33	Italy
3	Levinson, R.	28	U.States
4	Cotana, F.	25	Italy
5	Santamouris, M.	23	Greece
6	Zinzi, M.	18	Italy
7	Muscio, A.	11	Italy
8	Rossi, F.	10	Italy
9	Li, H.	10	China
10	Berdahl, P.	10	U.States

During the last decade, with the increase of the UHI effect and the evolving climate change, it became necessary to draw attention to the development of geoengineering-based solutions, such as cool surfaces. This strategy has been implemented for roof applications, building facades, and pavements. Nevertheless, the academic research placed an emphasis on cool roofs, with a higher record of articles published on this application compared to the others

(Figure 5). Thus, the number of publications regarding cool roofs in the third period reached 251 documents, while, in the case of pavements and facades, only 58 and 22 papers were identified, respectively.

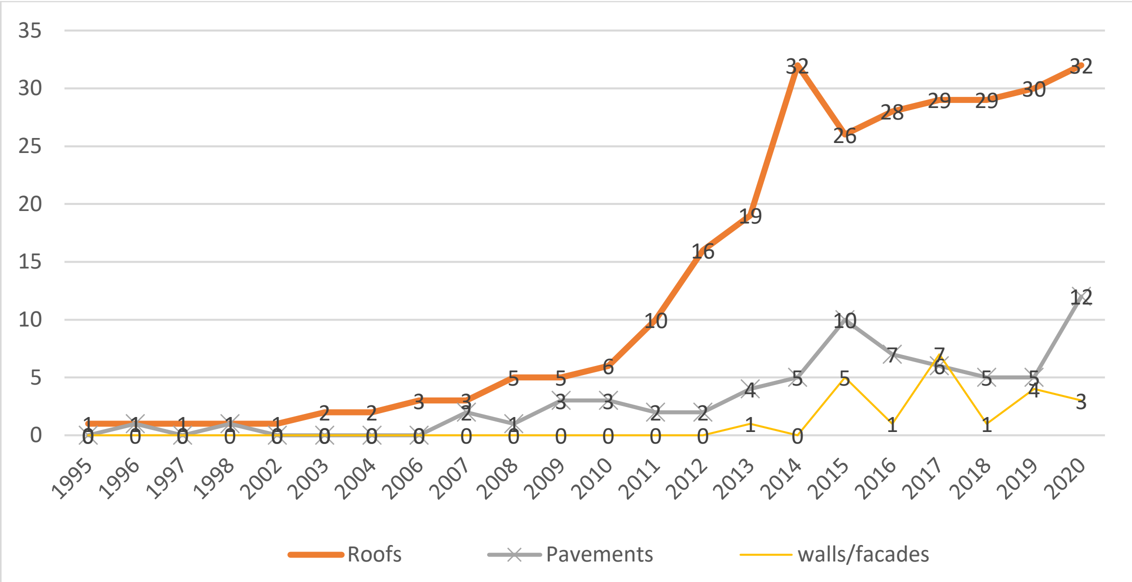


Figure 5. Evolution of cool material application domain: roofs, pavements, and walls/facades

3.2.Science mapping and visualization

Figure 6 plots the overlapping map representing the three periods of the research and the evolution of the keywords. Figure 7 presents the thematic evolution map of the research field, based on the h-index (Figure 7. a) and on the numbers of published documents for the three cited periods (Figure 7. b). The number of keywords grew substantially in the second period, with 52 units, and in the third period, with 16 more. Thus, it is possible to confirm that cool surfaces represent a growing research field. In fact, the first period was the least developed in terms of published documents, with most of them focused on the use of light-colored materials to provide high solar energy reflectance and to decrease the solar heat trapped in urban areas (Bretz et al., 1998; Rosenfeld et al., 1995). This period was characterized by the "cooling" theme (Figure 7. b), as the increase of temperature in cities urged the use of high-albedo surfaces instead of dark materials.

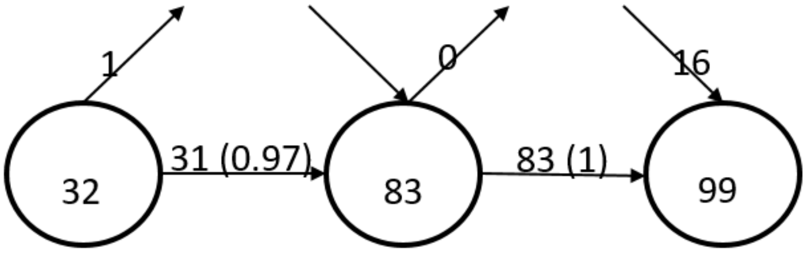
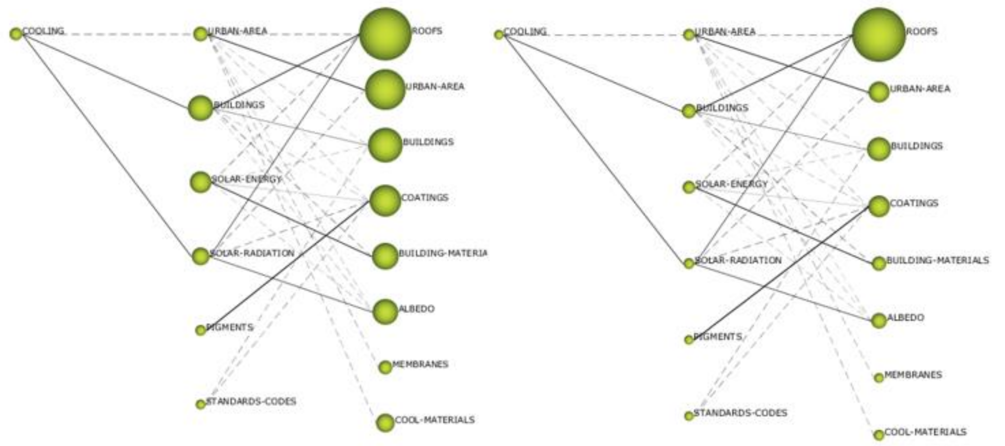


Figure 6. Overlapping map of the sample



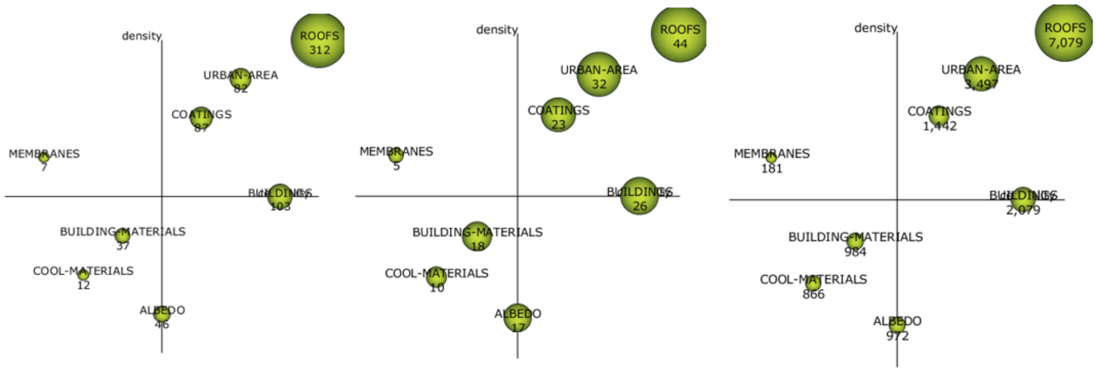
a. Thematic evolution map based on the h-index. *b. Thematic evolution map based on the number of published documents*

Figure 7. Thematic evolution map according to the h-index (a) and the number of published documents (b)

Later, several studies showed that the whiteness of materials enhances the surface albedo and increases the solar reflectance in the visible spectrum range, which reduces the cooling loads of buildings (Zinzi & Fasano, 2009). For instance, exploring alternative methods to create high-albedo concrete for pavement applications, Boriboonsomsin and Reza (Boriboonsomsin & Reza, 2007) found that replacing cement with whiter constituents (70% slag) achieves an albedo of 0.582, which is 71% higher than the conventional mix. To encourage the implementation of white reflective materials in buildings and urban areas, as well as to facilitate their integration into the construction sector, standards and product labelling were adopted and promoted on the basis of the spectral reflection examination of these materials (Akbari et al., 2001; Berdahl & Bretz, 1997; Rosenfeld et al., 1995). In this sense, some efforts have been taken to incorporate cool roofs as an effective sustainable strategy in the revised ASHRAE building standards, S90.1 (Rosenfeld et al., 1998). This approach was developing in the second period, and it coincides with the integration of building regulations to enhance energy performance, such as the first version of the Energy Performance of Buildings Directive, 2002/91/EC, as well as its subsequent update (Directive 2010/31/EU). In this second period, the research field started to receive more interest, which is highlighted by the inclusion of 52 new keywords (Figure 6) and the following six emerging themes (Figure 7): “buildings”; “solar-energy”; “solar-radiation”; “urban-area”; “pigments”; and “standards-codes”. It is observed that the h-index impact showed more emphasis for buildings, solar energy, and solar radiation, while the number of published documents is approximately the same for each theme. This rising interest was due to the need of developing cool materials with solar radiation reflectance properties not only in the visible range, but also in the NIR spectrum, to reduce energy consumption and enhance thermal comfort in buildings.

Finally, the third period increases the number of keywords to 99 (Figure 6), and it shows a link between the research field and the creation of balanced solutions in the development of coatings, membranes, and materials designed to save energy in buildings (Figure 7. b). This represents the most prolific period in terms of published documents. In this period, the conceptual evolution of the themes was developed for more specific concepts including: “roofs”; “urban-area”; “buildings”; “coatings”; “building-materials”; “albedo”; “membranes”; and “cool-materials”. Researchers started to explore more alternatives complying to the energy efficiency and aesthetic requirements, with the increasing interest for materials with appropriate thermal emissivity/absorption spectrum. For this purpose, the careful selection of nanoparticles and pigments was developed to optimize the thermal and optical performances of materials, such as radiative cooling painting. Several works were led in this sense to develop high-reflective paints known as “cool paints”, such as smart coatings with high NIR reflectance to reduce the solar heat gain and the energy consumption (Cozza et al., 2015; Kavitha & Sivakumar, 2020; Matias et al., 2015; N. Zhou et al., 2020). In addition, the coatings were developed in order to achieve the color and efficient radiative cooling requirements in a simple, low-cost, and scalable way (Y. Chen et al., 2020).

Since the third period (2011–2020) was the most prolific for this field of research, it is analyzed in detail below. Figure 8 shows a strategic diagram of a two-dimensional space that was built by plotting the themes according to their centralities and their density rank values. This includes four quadrants, each containing a specific theme: (i) Motor themes, in the upper-right quadrant; (ii) Basic and transversal themes, in the lower-right quadrant; (iii) Highly developed and isolated themes, in the upper-left quadrant; and (iv) Emerging or declining themes, in the lower-left quadrant (Cobo et al., 2012).



a. Strategic diagram of the third period based on the number of published documents.

b. Strategic diagram of the third period based on the h-index.

c. Strategic diagram of the third period based on citations.

Figure 8. Strategic diagram of the third period, the volume of the spheres is proportional to the number of documents published (a), to the h-index (b) and to the number of citations (c) in the third period associated with each theme

According to Figure 8.(a), almost 70% of the published records represent roofs, urban-area, and coatings themes, registering the highest impacts (Figure 8. b). It can be seen that roofs and urban-area are the most cited themes, with 7079 and 3497 citations, respectively, followed at a distance by coatings, with 1442 citations (Figure 8. c). For the case of roofs theme, the origin links show an association with the concepts including building, solar-radiation, solar energy, and urban areas (Figure 7), representing the most addressed application for cool materials according to Figure 5. It is noteworthy to mention that the urban-area motor theme is associated with all the specific themes of the third period as an origin theme.

Analysing the cluster network allowed the determination of the most highlighted materials applied for cool surface that are associated with the themes resulted from this study. Coatings theme showed a strong relation with the development of pigments (Figure 7. a), especially the ones performing in the NIR spectrum as reflective materials, as it can be seen in its cluster network (Figure 9. d). In terms of the isolated membranes theme (Figure 9. c), it concerns mainly the latent strategy of the thermal energy storage resumed in the PCMs. Finally, the two emerging themes of building-materials and cool-materials, discuss ceramic materials, glass, and retroreflective materials (RR materials). The most relevant contributions to these topics are analyzed in the next section for the review of materials applied for cool surfaces.

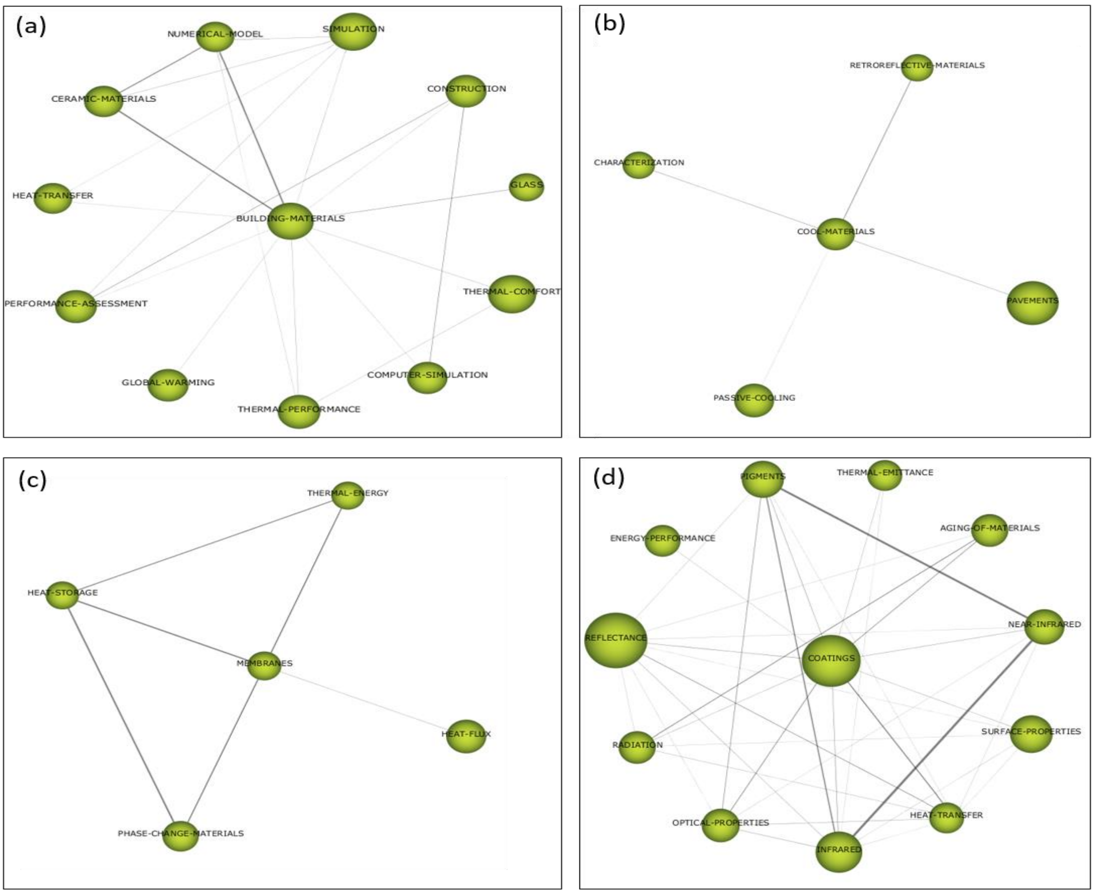


Figure 9. Cluster networks of: (a) building-materials; (b) cool-materials; (c) membranes; and (d) coatings

3.3.Literature revision of materials applied for cool surfaces

The use of cool materials for building envelopes and pavements helps to increase the solar reflectance performance, which decreases the energy use for the cooling demand and enhances the indoor and outdoor thermal comfort. The fabrication of such materials to mitigate the UHI implies a balance between the technical fabrication and use of these materials and their environmental impact to secure a sustainable production and service loop. In this section, the following most highlighted materials in the science mapping results are analyzed: (i) Pigments; (ii) RR materials; (ii) PCMs; (iv) Ceramic materials; and (v) Glass. The application of more than one of these materials to create a cool surface is considered as a complementary application strategy that contributes to the overall reflectivity performance of the surface. Moreover, the application of each one is conditioned by the specific building design, climate conditions and local regulations.

These materials were the subject of 63 publications, from which ~25% evaluate ceramic materials, 22% are dedicated to pigments, and 19, 19, and 14% evaluate PCMs, glass, and RR materials, respectively. This sample was selected by means of clustering during the third period. However, six publications were extracted from the previous periods and added to the analysis because of the impacts and contributions of the authors, such as Akbari, H, who has made the most contributions in terms of the records published related to the field of cool surfaces. The literature reviewing of these materials is included below.

3.3.1. Pigments

According to the publications presented in Table 2, pigments were evaluated through the 14 most relevant publications that are related to the theme of energy use in buildings. Most of the publications are affiliated with research entities based in India and China.

Accordingly, pigments were proven to enhance the NIR reflectance for cool coatings and to maintain a wider range of color choice, which opened the way for an industry of geoengineering and chemical solutions for cool materials (Cheng et al., 2009; Levinson et al., 2005b, 2005a; Raj et al., 2015; Xiang & Zhang, 2018). The absorption coefficient and the solar spectral backscattering measured in the range of 300–2500nm determines whether the pigment should be implemented in a cool coating (Levinson et al., 2005a). According to Levinson et al. (Levinson et al., 2005b), a pigment with a low absorptance is considered cool, whereas a pigment with high NIR transmittance will necessitate a NIR-reflective background (usually white or metallic) in order to form a NIR-reflecting coating. The developed reflective pigments allowed for the transition from lighter color to dark coating materials, often without compromising the reflective performance. Five dark-colored pigments were analyzed in a study to develop new species of high-NIR reflectance. With a formulation of a 25% weight content of rutile-type titanium dioxide, the white ceramic microspheres, with a 13% weight content, and the heavy calcium carbonate, with a 10% weight content, obtained positive results, and the back temperatures were lowered by 10–20 °C, compared to normal coatings (Cheng et al., 2009).

In general, pigments integrated in the matrix of cool materials range from organic, of which most are considered transparent, to complex inorganic color mixed-metal oxides that are often opaque (Levinson et al., 2005a; Rosati et al., 2021b). Cool pigments achieve a high solar reflectance of up to 95%, compared to TiO_2, such as bismuth titanate (Meenakshi & Selvaraj, 2018; Raj et al., 2019), barium titanate (Xiang & Zhang, 2018), and terbium-doped yttrium cerate (Raj et al., 2015, 2017). The photocatalytically active white inorganic pigment

TiO_2 represents the most widely used nontoxic pigment and is characterized by strong scattering, weak absorption, and good stability (Bettoni et al., 2012). As a result, most studies have been compared to its reflective performance (Pisello et al., 2014). The use of developed pigments was recently the most processed strategy. When incorporated into glazes, fillers, and engobes, synthesized pigments are used in coatings in order to enhance the optical and thermal performances of cool materials, as well as to respect the aesthetic requirements of a design. Several oxides were demonstrated to be able to enhance the NIR reflectance of glazes for concrete cement substrates, steel substrates, metal panels, and clay tiles (Z. Li, Yang, et al., 2017; Raj et al., 2015; Thongkanluang et al., 2013). For traditional buildings, using pigments based on sodium silicate for the tile coating improved the solar reflection in the NIR spectrum by 13% without affecting the visual appearance (Pisello et al., 2014). The integration of such pigments lowers roof overheating and reduces the energy required for cooling.

Cool pigments have become a growing field that is taking over cool surface technology, as nanoparticles are integrated into material structures to fulfil a specific design criterion and to enhance the reflective performance. In addition to their chemical stabilities, their effective thermal and optical performances have been proven in numerous studies. However, the acquisition, the production process, the synthesis methods, the high cost of the rare-earth elements used in their composition, and the hazardous environmental effect of the heavy metals may have a negative environmental impact (Cao et al., 2020; Shittu et al., 2020). On the other hand, the development of new compositions of reflective pigments continues to increase. In addition to the high reflectivity, the research field has also been evaluating the functional and long-term performances against surface contamination (Yun & Yim, 2021), thermal insulation (Ramos et al., 2021; Tian et al., 2021), and the synthesis methods in terms of the energy and raw material costs, which were directed for the sol–gel method and rare-earth compounds (Divya & Das, 2021; Kamal et al., 2021; Y. Li et al., 2021; Rosati et al., 2021a; Soranakom et al., 2021; W. Zhou et al., 2021).

Table 2. Most important publications about pigments

Title of Publ.	Authors	Journal	Year of publ.	Ref.	Country of Affiliation
Solar spectral optical properties of pigments—Part I: Model for deriving scattering and absorption coefficients from transmittance and reflectance measurements.	Levinson, R., Berdahl, P., and Akbari, H.	*Solar Energy Materials and Solar Cells*	2005	(Levinson et al., 2005a)	United States
Solar spectral optical properties of pigments—Part II: Survey of common colorants.	Levinson, R., Berdahl, P., and Akbari, H.	*Solar Energy Materials and Solar Cells*	2005	(Levinson et al., 2005b)	United States
Study of solar heat-reflective pigments in cool roof coatings.	Cheng, M., Ji, J., and Chang, Y.	*Journal of Beijing University of Chemical*	2009	(Cheng et al., 2009)	China
Bismuth titanate as an infrared reflective pigment for cool roof coating.	Meenakshi, P., and Selvaraj, M.	*Solar Energy Materials and Solar Cells*	2018	(Meenakshi & Selvaraj, 2018)	India
Pigmentary colors from yellow to red in Bi2Ce2O7 by rare earth ion substitutions as possible high NIR reflecting pigments.	Raj, A. K. V., Rao, P. P., Sreena, T. S., and Thara, T. R. A.	*Dyes and Pigments*	2019	(Raj et al., 2019)	India
A new member of solar heat-reflective pigments: BaTiO3 and its effect on the cooling properties of ASA (acrylonitrile-styrene-acrylate copolymer).	Xiang, B., and Zhang, J.	*Solar Energy Materials and Solar Cells*	2018	(Xiang & Zhang, 2018)	China
Terbium doped Sr2MO4 [M = Sn and Zr] yellow pigments with high infrared reflectance for energy saving applications.	Raj, A. K. V., PrabhakarRao, P., Divya, S., and Ajuthara, T. R.	*Powder Technology*	2017	(Raj et al., 2017)	India
Pigments based on terbium-doped yttrium cerate with high NIR reflectance for cool roof and surface coating applications.	Raj, A. K. V., Prabhakar Rao, P., Sameera, S., and Divya, S.	*Dyes and Pigments*	2015	(Raj et al., 2015)	India
Surfactant effect on titanium dioxide photosensitized oxidation of 4-dodecyloxybenzyl alcohol.	Bettoni, M., Brinchi, L., Del Giacco, T., Germani, R., Meniconi, S., Rol, C., and Sebastiani, G. V.	*Journal of Photochemistry and Photobiology A: Chemistry*	2012	(Bettoni et al., 2012)	Italy

On a cool coating for roof clay tiles: Development of the prototype and thermal-energy assessment.	Pisello, A. L., Cotana, F., and Brinchi, L.	*Energy Procedia*	2014	(Pisello et al., 2014)	Italy
Effects of added ZnO on the crystallization and solar reflectance of titanium-based glaze.	Li, Z., Yang, Y., Peng, C., and Wu, J.	*Ceramics International*	2017	(Z. Li, Yang, et al., 2017)	China
Performance of near-infrared reflective tile roofs.	Thongkanluang, T., Wutisatwongkul, J., Chirakanphaisarn, N., and Pokaipisit, A.	*Advanced Materials Research*	2013	(Thongkanluang et al., 2013)	Thailand
Environmental impact of cool roof paint: case study of house retrofit in two hot islands.	Emmanuel, S., Valentina, S., Petra, G., and Maria, K.	*Energy and Buildings*	2020	(Shittu et al., 2020)	United Kingdom
Preparation of phthalocyanine blue/rutile TiO2 composite pigment with a ball milling method and study on its NIR reflectivity	Lingyun, C., Xuening, F., Hongbin, Z., and Changliang, H.	*Dyes and Pigments*	2020	(Cao et al., 2020)	China

3.3.2. RR Materials

According to the publications presented in Table 3, the RR materials were evaluated through the nine most relevant publications that are related to the themes of sustainability, energy use, and construction engineering. The majority of these publications are affiliated with research entities based in Italy.

Independently of the incidence direction, retroreflectivity refers to the capacity to reflect the incoming light beam to a surface back towards its source (F. Rossi et al., 2014, 2016). The application of diffusive materials on building envelopes induces multiple reflections within the urban canyon patterns, therefore, in order to reduce the captured solar radiation energy, the use of RR materials presents a good alternative (Sakai et al., 2011a). In this sense, RR materials were studied to evaluate their potential with respect to diffusive (Lambertian) coatings, which allowed for the determination of a corrective parameter to enhance the comparison in terms of mitigating the UHI effect (Gambelli et al., 2019). For the retro-reflectivity measurements, Sakai et al. (Sakai et al., 2008) present a procedure to measure only the retroreflective components of RR materials, which consists of: (i) Measuring the total reflectance by thermal measurements, (ii) measuring the reflectance without retroreflection using a spectrometer; and finally (iii) the RR components are measured by subtracting the latter from the former. Several studies have proved the efficiency of RR materials to reduce the heat trapped in the building surroundings (F. Rossi et al., 2014, 2016; Sakai et al., 2011a). A RR facade with an albedo of 0.60 could reflect 55% of the incident sunlight, whereas a diffusive facade could reflect only 36% with the same albedo (Levinson et al., 2020). RR materials were also applied in pavements, where the cooling potential could reach a maximum of a 4.6% albedo increase, compared to the traditional white and beige diffusive cool materials (F. Rossi et al., 2016). The RR performance of glass beads was discussed in several investigations that show promising results, however, when comparing a base ceramic tile coated with glass spheres and clear solid barium titanate spheres, the latter had the highest global reflectance (39%), and a radiation energy that reflected up to 5% (Morini et al., 2018).

Table 3. Most important publications about RR materials

Title of Publ.	Authors	Journal	Year of Publ.	Ref.	Country of Affiliation
Analysis of retro-reflective surfaces for urban heat island mitigation: A new analytical model.	Rossi, F., Pisello, A. L., Nicolini, A., Filipponi, M., and Palombo, M.	Applied Energy	2014	(F. Rossi et al., 2014)	Italy
Experimental evaluation of urban heat island mitigation potential of retro-reflective pavement in urban canyons.	Rossi, F., Castellani, B., Presciutti, A., Morini, E., Anderini, E., Filipponi, M., and Nicolini, A.	Energy and Buildings	2016	(F. Rossi et al., 2016)	Italy
Development and evaluation of directional retroreflective materials: Directional retroreflective materials as a heat island countermeasure.	Sakai, H., Jyota, H., Emura, K., and Igawa, N.	Journal of Structural and Construction Engineering	2011	(Sakai et al., 2011b)	Japan
A normalization procedure to compare retro-reflective and traditional diffusive materials in terms of UHI mitigation potential.	Gambelli, A. M., Cardinali, M., Filipponi, M., Castellani, B., Nicolini, A., and Rossi, F.	AIP Conference Proceedings	2019	(Gambelli et al., 2019)	Italy
Reduction of reflected heat by retroreflective materials.	Sakai, H., Emura, K., and Igawa, N.	Journal of Structural and Construction Engineering	2008	(Sakai et al., 2008)	Japan
Design, characterization, and fabrication of solar-retroreflective cool-wall materials.	Ronnen, L., Sharon, C., Jonathan, S., Howdy, G., Tatsuya, H., Paul, B.	Solar Energy Materials and Solar Cells	2020	(Levinson et al., 2020)	United States, Japan
Optimized retro-reflective tiles for exterior building element.	Morini, E., Castellani, B., Anderini, E., Presciutti, A., Nicolini, A., and Rossi, F.	Sustainable Cities and Society	2018	(Morini et al., 2018)	Italy

Retroreflective façades for urban heat island mitigation: Experimental investigation and energy evaluations.	Rossi, F., Castellani, B., Presciutti, A., Morini, E., Filipponi, M., Nicolini, A., and Santamouris, M.	Applied Energy	2015	(F. Rossi et al., 2015)	Italy, Greece
Optic-energy and visual comfort analysis of retro-reflective building plasters.	B., Castellani, Alberto, M., Andrea, N., Federico, R.	Building and Environment	2020	(Castellani et al., 2020)	Italy

Besides the advantages that RR materials offer in road-sign use and visual technology, their application in building facades and pavements alleviates the heat trapped inside the buildings that is created by diffuse reflective materials. However, their retro-reflective behavior is limited to low angles of incidence, whereas, for high angles of incidence, the solar radiation is symmetrically reflected with regard to the perpendicular radiation (F. Rossi et al., 2015), which limits the performance of RR materials at all angles of incidence (Castellani et al., 2020; Levinson et al., 2020).

Recently, more research has been oriented toward the performance of RR materials for different angles of incidence that takes into account the irradiated surface geometry scenarios, the urban density, the microclimate, the durability, and the costs and benefits (Manni & Nicolini, 2021). As a solution, an angular selective behavior was discussed to overcome the limitations of RR materials, especially in summer (Anupam et al., 2021).

3.3.3. PCMs

According to the publications presented in Table 4, PCM materials were evaluated through the 12 most relevant publications that are related to the theme of energy use in buildings. Most of the publications are affiliated with research entities based in South Korea and the United States.

PCMs have the ability to change their physical characteristics during phase transition (Mapston & Westbrook, 2010). To compensate for the possible heating load increase in winter while using cool roofs, these materials can prevent the overheating of the roof surface during the summer without increasing the heating load in the winter (Yoon et al., 2018). As a consequence, they decrease the thermal stress and the annual energy load consumption, in addition to providing thermal inertia for buildings when the melting temperature is optimized (Saffari et al., 2018). The building energy performance and the thermal comfort could be improved depending on the phase change temperature adopted, according to Chang et al. (Chang et al., 2020), better results were registered with 30 °C than with 20 °C.

Table 4. Most important publications about PCMs

Title of Publ.	Authors	Journal	Year of Publ.	Ref.	Country of Affiliation
Simulating the effects of cool roof and PCM (phase change materials) based roof to mitigate UHI (urban heat island) in prominent US cities.	Roman, K. K., O'Brien, T., Alvey, J. B., and Woo, O. J.	Energy	2016	(Roman et al., 2016)	United States
Prefabricated building units and modern methods of construction (MMC).	Mapston, M., and Westbrook, C.	Materials for Energy Efficiency and Thermal Comfort in Buildings	2010	(Mapston & Westbrook, 2010)	United Kingdom
Understanding a potential for application of phase-change materials (PCMs) in building envelopes.	Kośny, J., and Kossecka, E.	ASHRAE Transactions	2013	(Kosny PhD & Kossecka PhD, 2013)	Poland, United States
Thermal stress reduction in cool roof membranes using phase change materials (PCM).	Saffari, M., Piselli, C., de Gracia, A., Pisello, A. L., Cotana, F., and Cabeza, L. F.	Energy and Buildings	2018	(Saffari et al., 2018)	Italy, Spain
Development of PCM cool roof system to control urban heat island considering temperate climatic conditions.	Chung, M. H., and Park, J. C.	Energy and Buildings	2016	(Chung & Park, 2016)	South Korea
PCM cool roof systems for mitigating urban heat island—an experimental and numerical analysis.	Yang, Y. K., Kim, M. Y., Chung, M. H., and Park, J. C.	Energy and Buildings	2019	(Y. K. Yang et al., 2019)	South Korea
Thermal Performance Test of a Phase-Change-Material Cool Roof System by a Scaled Model.	Yoon, S. G., Yang, Y. K., Kim, T. W., Chung, M. H., and Park, J. C.	Advances in Civil Engineering	2018	(Yoon et al., 2018)	Republic of Korea
Numerical analysis of phase change materials/wood-plastic composite roof module system for improving thermal performance.	Seong, J., Seunghwan, W., Hyun, M., Su-Gwang, J., Sumin, K.	Journal of Industrial and Engineering Chemistry	2020	(Chang et al., 2020)	United States, Republic Korea
How to enhance thermal energy storage effect of PCM in roofs with varying solar reflectance: Experimental and numerical assessment of a new roof system for passive cooling in different climate conditions.	Piselli, C., Castaldo, V. L., and Pisello, A. L.	Solar Energy	2019	(Piselli et al., 2019)	Italy

Effects of accelerated weathering on the optical characteristics of reflective coatings for cool pavement.	Ning, X., Hui, L., Hengji, Z., Xue, Z., Ming, J.	Solar Energy Materials and Solar Cells	2020	(Xie et al., 2020)	China
Phase change materials for pavement applications: A review	B.R. Anupam, Umesh Chandra Sahoo, PrasenjitRath	Construction and Building Materials	2020	(Anupam et al., 2020)	India
Review of current state of research on energy storage, toxicity, health hazards and commercialization of phase changing materials.	S.S.Chandel, Tanya, A.	Renewable and sustainable enrgy reviews	2017	(Chandel & Agarwal, 2017)	India

The performance of PCM-based surface technology as an UHI mitigation strategy has been evaluated through several studies. For roof application, a cool polyurethane-based membrane reduces the roof-surface temperature and the heat flux through the roof more than a traditional dark bitumen membrane, moreover, the outdoor environmental conditions and the type of PCM host material could influence the performance of the membrane (Piselli et al., 2019). Similar results were proven using PCM-doped tiles for a cool roof system in simulated summer conditions (Chung & Park, 2016). In real winter conditions, the use of PCMs maintained a higher indoor temperature compared to cool paints, and with a low surface temperature reducing the heat penalty (Y. K. Yang et al., 2019). In this sense, several types of PCMs have been tested as dynamic components in buildings in comparison to conventional cool roof materials, showing that their incorporation in the roof materials matrix decreased the heat gain flux by 54% and the sensible heat by 40% for various values of albedo compared to cool roof technology (Roman et al., 2016).

PCMs have also been tested for cool pavements. Their use is based on the temperature regulation performance of these materials, which use a lightweight aggregate with a reasonable gradation for better results. The composite PCMs incorporated into the asphalt mixtures achieved a satisfactory cooling performance, however, some of them minimized the strength reductions of the mixtures (Xie et al., 2020).

The incorporation of PCMs in cool surfaces is emerging as a growing field of research because of their ability to restore energy with a minimum change in volume, and without an increase in temperature; however, the encapsulation method may affect their performance and cause leakage (Anupam et al., 2020; Chung & Park, 2016; Kosny PhD & Kossecka PhD, 2013; Piselli et al., 2019; Roman et al., 2016; Saffari et al., 2018). Moreover, some PCMs may have environmental impacts. For instance, the PCM paraffin wax that is commonly used releases toxic vapors when burnt, which can result in severe health hazards, as it contains formaldehyde, benzene, toluene, and other toxic compounds (Chandel & Agarwal, 2017). In order to enhance the thermal energy storage in buildings, recent investigations have been oriented toward developing new techniques of encapsulation. Nano/microencapsulation methods have presented promising results while avoiding leakage (Gong et al., 2021; Naikwadi et al., 2021). Moreover, different combinations of reflective coatings and PCM applications were tested, the optimal combination required a layer of thermal insulation between the two materials, which incorporates the PCMs as a complementary technique for the energy-saving system of buildings (Ling et al., 2021).

3.3.4. Ceramic Materials

According to the publications presented in Table 5, ceramic materials have been evaluated in 16 publications that are highly related to the theme of energy use in buildings. Nearly 50% of the publications are affiliated, either implicitly or explicitly, with research entities based in Italy.

The publications showed the development of innovative solutions for a better solar reflectance index of ceramic materials, such as tiles, glazes, and engobes. The use of ceramic tiles is considered an effective component of the cool roof strategy, thanks to its durability and its solar properties, especially if it is glazed (C. Ferrari et al., 2013, 2015; Z. Li, Zhao, et al., 2017). The substrate material was tested with the application of different ceramic coatings through several studies, and the developed non white coatings enhanced the solar reflective performance and showed interesting results in terms of energy saving (Levinson et al., 2007; Rosado et al., 2014). For instance, the wollastonite–hardystonite glass–ceramic porous tiles showed high reflectance of solar radiation, coupled with low thermal conductivity in an arid environment (Marangoni et al., 2017), which highlights the complementary function with regard to the thermal insulation properties (Di Giuseppe et al., 2019; C. Ferrari et al., 2016). In the same sense, an improvement in the thermal performance of a residential building was found during the summer and the winter, as 75% of the solar radiation reflectance in the NIR spectrum was registered, i.e., 10% more with respect to traditional tiles, without altering the visible appearance (Pisello et al., 2014). The latter property is highly considered for historical buildings that are required to maintain their original aesthetic appearance. These types of buildings often exist in the center of urban areas, which are strongly affected by the UHI. Their retrofitting using innovative cool clay tile coatings increased the solar reflectance by 20% while maintaining the original color intact. This kind of retrofitting enhances the thermal responses of buildings and the urban climate in general (Pisello, 2015; Pisello et al., 2013; Takebayashi et al., 2012; Yacouby et al., 2011).

Table 5. Most important publications about ceramic materials

Title of Publ.	Authors	Journal	Year of Publ.	Ref.	Country of Affiliation
Experimental evaluation of thermal performance of cool pavement material using waste tiles in tropical climate.	Anting, N., Md. Din, M. F., Iwao, K., Ponraj, M., Jungan, K., Yong, L. Y., and Siang, A. J. L. M.	Energy and Buildings	2017	(Anting et al., 2017b)	Malaysia, Japan
On a cool coating for roof clay tiles: Development of the prototype and thermal-energy assessment.	Pisello, A. L., Cotana, F., and Brinchi, L.	Energy Procedia	2014	(Pisello et al., 2014)	Italy
Performance of near-infrared reflective tile roofs.	Thongkanluang, T., Wutisatwongkul, J., Chirakanphaisarn, N., and Pokaipisit, A.	Advanced Materials Research	2013	(Thongkanluang et al., 2013)	Thailand
Design of ceramic tiles with high solar reflectance through the development of a functional engobe.	Ferrari, Chiara, Libbra, A., Muscio, A., and Siligardi, C.	Ceramics International	2013	(C. Ferrari et al., 2013)	Italy
Design of a cool color glaze for solar reflective tile application.	Ferrari, C., Muscio, A., Siligardi, C., and Manfredini, T.	Ceramics International	2015	(C. Ferrari et al., 2015)	Italy
High-solar-reflectance building ceramic tiles based on titanite ($CaTiSiO_5$) glaze.	Li, Z., Zhao, M., Zeng, J., Peng, C., and Wu, J.	Solar Energy	2017	(Z. Li, Zhao, et al., 2017)	China
Cooler tile-roofed buildings with near-infrared-reflective non-white coatings.	Levinson, R., Akbari, H., and Reilly, J. C.	Building and Environment	2007	(Levinson et al., 2007)	United States
Measured temperature reductions and energy savings from a cool tile roof on a central California home.	Rosado, P. J., Faulkner, D., Sullivan, D. P., and Levinson, R.	Energy and Buildings	2014	(Rosado et al., 2014)	United States
New strategy to mitigate urban heat island effect: Energy saving by combining high albedo and low thermal diffusivity in glass ceramic materials.	Enríquez, E., Fuertes, V., Cabrera, M. J., Seores, J., Muñoz, D., and Fernández, J. F.	Solar Energy	2017	(Enríquez et al., 2017)	Spain
White sintered glass-ceramic tiles with improved thermal insulation properties for building applications.	Marangoni, M., Nait-Ali, B., Smith, D. S., Binhussain, M., Colombo, P., and Bernardo, E.	Journal of the European Ceramic Society	2017	(Marangoni et al., 2017)	Italy, France, Saudi Arabia, United States

A composite cool colored tile for sloped roofs with high "equivalent" solar reflectance.	Ferrari, Chiara, Libbra, A., Cernuschi, F. M., De Maria, L., Marchionna, S., Barozzi, M., … Muscio, A.	Energy and Buildings	2016	(C. Ferrari et al., 2016)	Italy
Optical properties of traditional clay tiles for ventilated roofs and implication on roof thermal performance.	Di Giuseppe, E., Sabbatini, S., Cozzolino, N., Stipa, P., and D'Orazio, M.	Journal of Building Physics	2019	(Di Giuseppe et al., 2019)	Italy
Study on the cool roof effect of Japanese traditional tiled roof: Numerical analysis of solar reflectance of unevenness tiled surface and heat budget of typical tiled roof system.	Takebayashi, H., Moriyama, M., and Sugihara, T.	Energy and Buildings	2012	(Takebayashi et al., 2012)	Japan
Development of clay tile coatings for steep-sloped cool roofs.	Pisello, A. L., Cotana, F., Nicolini, A., and Brinchi, L.	Energies	2013	(Pisello et al., 2013)	Italy
Thermal-energy analysis of roof cool clay tiles for application in historic buildings and cities.	Pisello, A. L.	Sustainable Cities and Society	2015	(Pisello, 2015)	Italy
Study on roof tile's colors in Malaysia for development of new anti-warming roof tiles with higher Solar Reflectance Index (SRI).	Yacouby, A. M. A., Khamidi, M. F., Nuruddin, M. F., Farhan, S. A., and Razali, A. E.	National Postgraduate Conference—Energy and Sustainability: Exploring the Innovative Minds	2011	(Yacouby et al., 2011)	Malaysia

Glazes present a good complementary component for cool tiles, as glass–ceramic materials they are fabricated through a controlled crystallization process for a desired microstructure. The incorporation of cool pigments in the composition of glazes yields the total reflective performance of the product up to 82.8% (Thongkanluang et al., 2013). The results of an experimental study show that a tile coated with glass ceramic material induces 20% energy savings, compared to TiO_2-based paints, and that it could be used for roofs and pavements (Enríquez et al., 2017). The application of engobes enhances the solar reflectance as well, by up to 0.90 (C. Ferrari et al., 2013). As an intermediate layer between the substrate and the glaze, it provides a high degree of adhesion while taking into account the convenience of all the coefficients of thermal expansion.

The application of developed glazes and engobes enhances the solar reflective performance of tiles (Cedillo-González et al., 2022). In recent studies, the development of cool ceramic coatings was strongly discussed in terms of the low-cost routes, the use of secondary materials, and the self-cleaning abilities (Rahayu et al., 2021). In turn, research has been focused on enhancing the compositions of glass ceramic frits, opacifiers, and pigments to reach the optimal potential for NIR reflectance. The development of coatings has been oriented toward the use of dynamic coatings, such as the passive ones: photochromic and thermochromic coatings (Khaled & Berardi, 2021). It is clear that the research field is an intersection of multiple complementary techniques and materials, which include pigments, glazes, and tiles (Divya & Das, 2021; Taallah et al., 2021), and this creates a wide range of opportunities to attain the energy-saving potential of cool materials in future studies.

3.3.5. Glass

According to the publications presented in Table 6, the use of glass is discussed through the 12 most relevant publications that are related to the themes of buildings and the cleaner production of materials. The publications are affiliated with research entities based in different countries and that are not concentrated in a specific one.

Table 6. Most important publications about glass

Title of Publ.	Authors	Journal	Year of Publ.	Ref.	Country of Affiliation
Optic-energy and visual comfort analysis of retro-reflective building plasters.	Castellani, Gambelli, Nicolini, Rossi	Building and Environment	2020	(Castellani et al., 2020)	Italy
Waste glass in civil engineering applications—A review.	Kazmi, D., Williams, D. J. and Serati, M.	International Journal of Applied Ceramic Technology	2020	(D. Kazmi et al., 2020)	Australia
Reuse of waste glass in building brick production.	Demir, I.	Waste Management and Research	2009	(Demir, 2009)	Turkey
Utilization of waste glass to enhance physical-mechanical properties of fired clay brick.	Phonphuak, N., Kanyakam, S., and Chindaprasirt, P.	Journal of Cleaner Production	2016	(Phonphuak et al., 2016)	Thailand
Properties of Fired Clay Bricks Mixed with Waste Glass.	Abdeen, H., and Shihada, S.	Journal of Scientific Research and Reports	2017	(Abdeen & Shihada, 2017)	Palestine
The role of glass waste in the production of ceramic-based products and other applications: A review.	Silva, R. V., de Brito, J., Lye, C. Q., and Dhir, R. K.	Journal of Cleaner Production	2017	(R. V. Silva et al., 2017)	Portugal, United Kingdom
Effect of waste glass on properties of burnt clay bricks.	Hameed, A., Haider, U., Qazi, A. U., and Abbas, S.	Pakistan Journal of Engineering and Applied Sciences	2018	(Hameed et al., 2018)	Canada
Thermal performance evaluation of eco-friendly bricks incorporating waste glass sludge.	Kazmi, S. M. S., Munir, M. J., Wu, Y. F., Hanif, A., and Patnaikuni, I.	Journal of Cleaner Production	2018	(S. M. S. Kazmi et al., 2018)	Australia, Pakistan, Hong Gong
Glass recycling in the production of low-temperature stoneware tiles.	Lassinantti Gualtieri, M., Mugoni, C., Guandalini, S., Cattini, A., Mazzini, D., Alboni, C., and Siligardi, C.	Journal of Cleaner Production	2018	(Lassinantti Gualtieri et al., 2018)	Italy
Effect of glass powder on the technological properties and microstructure of clay mixture for porcelain stoneware tiles manufacture.	Njindam, O. R., Njoya, D., Mache, J. R., Mouafon, M., Messan, A., and Njopwouo, D.	Construction and Building Materials	2018	(Njindam et al., 2018a)	Burkina Fasu, Cameroon
Incorporating hollow glass microsphere to cool asphalt pavement: Preliminary evaluation of asphalt mastic.	Du Yinfei, Dai Mingxin, Deng Haibin, Deng Deyi, Cheng Peifeng, Ma Cong	Construction and Building Materials	2020	(Yinfei et al., 2020)	China

Cool White Polymer Coatings based on Glass Bubbles for Buildings.	Nie, YoungjaeYoo, Hasitha Hewakuruppu, Sullivan, Krishna, Jaeho Lee	Scientific Reports	2020	(Nie et al., 2020)	South Korea, United States

Recently, numerous solutions for cool materials have been evaluated through the academic research in terms of sustainability, which have paved the way for substituting raw materials and using secondary ones. Waste glass was introduced as a secondary raw material contributing to the efficiency of waste management principles.

Using glass as a secondary material in the fabrication of ceramic materials enhances the mechanical behavior and the sintering action due to its amorphous structure (D. Kazmi et al., 2020). Different percentages of waste glass have been tested in different studies for the fabrication of clay bricks. Results showed that the mechanical and thermal behaviors of these materials were affected by the percentage of the substitution, the size of the waste glass particles, and the chemical composition of each type (Abdeen & Shihada, 2017; Demir, 2009; Hameed et al., 2018; S. M. S. Kazmi et al., 2018; Phonphuak et al., 2016; R. V. Silva et al., 2017). For the fabrication of tiles, 41 wt.% of waste glass demonstrated good flexural strength and abrasion resistance when using the boron-rich waste glass as a sintering promoter (Lassinantti Gualtieri et al., 2018). However, Njindam et al. (Njindam et al., 2018b) demonstrated that the addition of high amounts of glass (>30 wt.%) into ceramic bodies is undesirable because of its negative effect on the physical properties. The integration of glass into the matrix of cool materials was mostly in its finest structure, in general, good results were obtained when using small-sized particles of glass. For instance, hollow glass microspheres were integrated into an asphalt mixture, which resulted in a 40% decrease in the thermal conductivity, and a 60% increase in the infrared reflectance (Nie et al., 2020; Yinfei et al., 2020). In the same sense, it was demonstrated that glass spheres incorporated in RR materials showed good results in terms of the energy reflected beyond the canyon for the building envelopes, in addition to the road-traffic-marking efficiency (Castellani et al., 2020). In the other hand, using waste glass cullet as secondary raw materials showed a good impact on the reflectance performance, improving the solar reflectance with 25% for the fabrication of a sustainable asphalt shingles (Kiletico et al., 2015).

Although the evaluation of the solar reflectance of ceramic materials incorporating waste glass has been little discussed in the academic research, according to the aforementioned studies, it was showed that for the fabrication of materials integrating waste glass, the solar reflectance performance depends in particular on the chemical composition and the particle size of the waste glass, in addition to the other technical conditions.

4. Conclusion

On the basis of a bibliometric analysis using SciMAT software, a sample of 982 academic records in the field of cool surfaces was processed for three periods, from 1995 to 2020. In particular, the most prolific period between 2011 and 2020, with 87.74% of the total records, was the main subject of the discussions, more recent studies were added. It was shown that cool surfaces is a developing research field that aims to create sustainable cool materials and coatings with efficient solar reflectance for applications including pavements, facades, and notably roofs, and which conform to the technical requirements of new and existing buildings,

The results of the analysis have placed emphasis on the materials perspective, as most materials highlighted in the research field of cool surfaces are pigments, RR materials, PCMs, ceramic materials, and glass. The majority of heat energy falls within the NIR wavelength spectrum, hence more concern was given to the development of NIR reflective materials. According to the literature, the efficiency of these materials is influenced not only by their thermal and optical performances, but also by the environmental impact resulting from the manufacturing and the use phase.

In the following is a summary of the major findings obtained:

- Developing pigments with high NIR reflectance is a growing industry, which provides up to 95% of the solar reflectivity performance. However, the acquisition, the production process, and the methods of synthesis of these nanoparticles may induce environmental burdens.

- Retroreflective materials, such as backscattering materials, present good performances by reflecting the incident solar radiation beyond the urban canyon and easing the heat trapped in the urban canopy with respect to the diffusive materials. However, their retroreflective behavior occurs mainly for low angles of incidence, therefore limiting their performance at large angles of incidence. More studies should discuss alternative solutions for the angular selective behavior.

- PCMs represent a good solution as cool-surface dynamic switch materials, and their implementation in the reflective materials matrix enhances the solar reflectivity performance. However, some PCM-based materials fabrication may impact the environment, moreover the encapsulation methods should be further addressed in future studies for better potential use.

- Ceramic materials are complementary effective materials for cool materials and coatings that enhance the total solar reflectance performance, with a broad range of reflective glazes and engobes.

- The integration of glass in the fabrication of ceramic materials as fine particles has been discussed in literature. The use of glass in these materials, with the optimal dosage and particle size, enhances their physical, mechanical, and thermal properties. Moreover, waste glass showed a good potential performance as a secondary raw material for solar reflectance purposes.

- The strategies discussed in this review have a common aim, which is contributing to the energy efficiency using sustainable raw materials and methods, these materials could be used as complementary strategies in buildings envelope for an energy saving system.

- The use of secondary raw materials as substitutes for primary raw materials is highly encouraged for the fabrication of sustainable cool materials. The incorporation of glass generated from waste in the manufacturing of cool materials is poorly discussed in the academic literature; thus, more research on the solar reflectance performance of waste glass particles as raw material should be further investigated.

This study presents a scientific contribution to the cool surfaces research field in terms of the materials and strategies it presents to counteract the urban heat island effect.

SECTION 2. MANUFACTURING AND CHARACTERIZATION OF COOL ROOF TILES WITH WASTE GLASS COATING

CHAPTER 2. Spectral reflectance characterization of waste glass coatings for tiles specimens: a preliminary study

1. Introduction

The cool surfaces strategies discussed in the first section provides a status quo of the evolution of this field, more recently in the context of developing new strategies that is encouraging the sustainability principles, and promoting the use of secondary raw materials for the fabrication of reflective materials. In the other hand, the increase of WG generation rises the need to develop sustainable techniques in terms of waste management and recycling possibilities. According to Blengini et al., the adaptation of efficient WG management implies the decrease of environmental impacts related to landfilling avoidance, recovery of co-products, and eco-friendly use of energy through the production process (Blengini et al., 2012). Therefore, the integration of such recyclable material as an alternative for raw materials in the matrix of cool coatings will help reduce the energy consumption and consequently the potential environmental burdens.

The non-biodegradable nature of glass encourages the development of new options to recycle it in order to alleviate the pressure from disposal procedures and raw materials extraction (Shao et al., 2000). According to the academic literature, promising results were obtained from recycling WG in the production of eco-friendly ceramic materials regarding the physical, mechanical, and thermal properties (Abdeen & Shihada, 2017; Conte et al., 2020; Demir, 2009; Lassinantti Gualtieri et al., 2018; Phonphuak et al., 2016; R. V. Silva et al., 2017). However, solar reflectance evaluation of coating materials containing WG was poorly discussed, especially in the visible and near-infrared radiation (NIR) spectrum (Dal Bó et al., 2014; Kalirajan et al., 2016).

In general, WG was either integrated in substrate materials, such as bricks and tiles, or coating materials, such as glazes, engobes, and binders. For the production of new designs of glazes, the substitution procedure was conducted in a total replacement of raw material (Anggono et al., 2019), or in a partial replacement, such as in the frits composition, for instance with a feasible amount of 8% mass in the case of a laminated WG (Cardoso de Souza-Dal Bó et al., 2021). Significant results were achieved regarding the cost of the new glazes with WG in their composition (Gol et al., 2021). Among WG origins, soda–lime glass, also called soda–lime–silica (SLS)glass, is the most abundant source of glass (Zimmer & Bragança, 2019), which explains the growing interest in the research field for their potential use for radiation-shielding applications. The addition of several oxides, such as antimony (III) oxide (Sb_2O_3), improves the radiation-shielding ability of SLS WG network (Kurtulus & Kavas, 2020), and it decreases the X-ray transmission (Kurtulus et al., 2020). Moreover, the addition of both lanthanum oxide (La_2O_3) and gadolinium oxide (Gd_2O_3) can increase the linear attenuation coefficient (LAC) values and, improve its gamma-rays-shielding characteristics (Kurtulus et al., 2021). Similar results were obtained with the addition of MoO_3 (Kurtulus et al., 2021). Furthermore, it is frequently found in WGs for their shielding applications and their optical properties the flint glass or lead silicate (LS) glass that contains a minimum of 24% (by weight) the lead (II) oxide (PbO). A broad range of scientific studies used chelating treatments to recycle LS WG resulting from cathode ray tubes (CRT) (Hu & Hui, 2018; Lv et al., 2016), since the amounts of CRT wastes have increased around the world after gradually replacing it with liquid crystal displays (LCD) (Meng et al., 2016). CRT glass is considered as a substitute for non-plastic materials, in particular, ceramic frits obtained from mixtures of silicates and carbonates to produce ceramic glazes (Singh et al., 2016). On the other hand, LS WG requires more caution in recycling and in disposal measures, since it may contain some toxic metal oxides.

The objective of this work is to establish a first proof of concept to identify the influence of using WG on the specular spectral reflectance and the degree of lightness of the WG coated tile specimens, considering the type of WG materials and the manufacturing process characterized by the parameters of temperature of burning, thickness of WG coating, and time of burning. The characterization of spectral reflectance performance of WG coatings, especially in the near infrared spectrum, was poorly discussed in the literature, and even less evidence on the use of waste glass as coatings on tiles. Therefore, a testing was performed for the relevance of variables, such as the WG composition and other manufacturing features including the holding time and temperature of burning in the degree of lightness and spectral reflectance. To achieve this goal, laboratory measurements were used to determine the spectral reflectance and degree of lightness of the specimens in the wavelength range of 350–1100 nm. The process was conducted in four steps: characterization of raw materials, manufacturing of the specimens, firing stage, and finally the measurement of the degree of lightness and the specular spectral reflectance. In this study, three types of WG were used in the preparation of coating for clay tiles specimens: two types of SLS WG with different compositions and a LS WG derived from CRT.

2. Materials and methods

In the experimental setup for the evaluation of clay tiles specimens coated with WG regarding the spectral reflectance and degree of lightness, several specimens were manufactured by using three values of WG thickness (0.5, 0.75, and 1 mm), with different burning temperatures (700, 850, and 1000 ◦C), using the laboratory kiln with different holding times (20, 40, and 60 min). In this section, the materials and their manufacturing based on the temperature of burning, thickness of WG coating, and time of burning are described. Moreover, the experimental setup used to measure the spectral reflectance and the degree of lightness is explained.

5.1.Origin of materials

Three types of WG were used in this study for the specimens' coatings. WG material was provided by the company "Camacho Recycling", the glass-collection plant located in Albacete, Spain. 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. Regarding the preparation of the substrate (ceramic body), it was carried out using a clay powder provided by the local company "Ladrillos Suspiro del Moro S.L" in Granada, Spain, under the instructions and supervision of the research team. Both companies have a long history in the collection and recycling of waste glass, and the manufacturing of bricks or other construction materials.

2.2.Preparation of the specimens

According to the dosage provided by the company, 81 clay substrates of 3.2 cm × 3.2 cm × 1.5 cm were fabricated by mixing clay powder with 15 wt.% of water ("wt." stands for percentage of weight per unit volume). The production process is shown in Figure 10, and it can be summarized as follows:

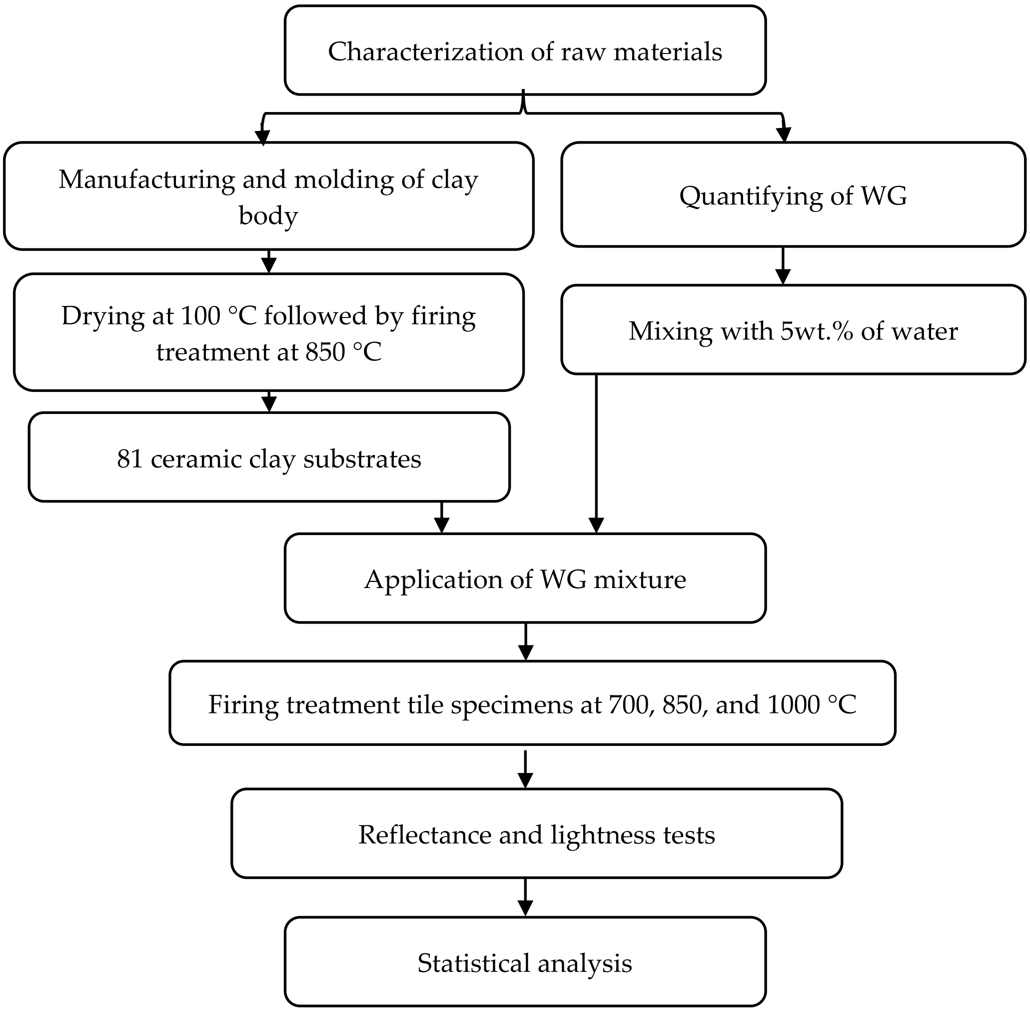


Figure 10. Flowchart of the research procedure

Initially, clay powder was processed and treated using a drying process until the weight was stabilized. The resulted treated clay powder was mixed homogenously with water using an electrical mixer. The following step was the molding of the substrates, using a mold (Figure 11) fabricated according to the dimensions required for the further tests' measurements. With the help of a compressive test machine, uniaxial pressing was applied on the mold to shape the clay body at 1000 MPa. This process was followed by a drying cycle of 48h under the temperature of 100ºC at the laboratory furnace. Finally, a firing treatment was conducted for the specimens during 1h under the temperature of 850ºC at the laboratory kiln. In this work, we opted for a double firing process, the biscuit state was obtained by drying the 81 clay substrates for 48h at 100ºC, using the laboratory furnace, followed by a firing treatment during 1 h at 850ºC using the laboratory kiln.

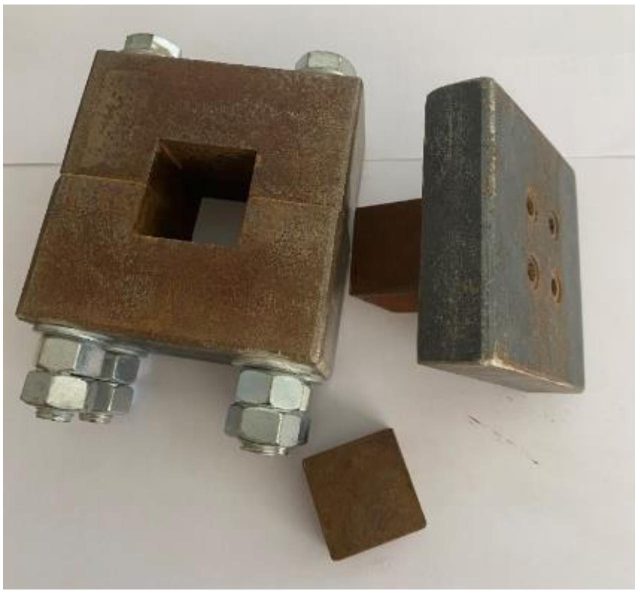


Figure 11. Mold used in the laboratory for the specimen shaping

In a second step, the WG coatings were applied to the clay substrate. According to the varying parameters of the study, for the preparation of WG coatings, three quantities of each WG type representing the three values of thickness were mixed with 5 wt.% of water (Table 7) in order to obtain a mixture to be spread evenly. After the application of the coatings, the firing treatment was processed according to the following variables: holding time and temperature, i.e., 20, 40, and 60min for each of the temperatures 700, 850, and 1000ºC.

Table 7. Designation of WG specimens

Waste Glass (WG) Type	Particle Size (mm)	Identification code	No. of Specimens
1st SLS WG	(0-1)	WG1_Qx [1]_y [2]	27
2nd SLS WG	(0–3)	WG2_Qx_y	27
LS WG	(0–4)	WG3_Qx_y	27

[1] Thickness of WG coating (Q1 = 0.5, Q2 = 0.75 and Q3 = 1 mm).

[2] Time of burning: 20, 40, and 60 min.

2.3.Chemical characterization of raw materials

The characterization of the three WG types and the clay powder was conducted through the chemical composition obtained by X-ray fluorescence (XRF) analysis as a non-destructive test method used to analyze chemical composition. In the XRF-based analysis, a primary X-ray beam is directed at a sample, which allows the measurement of the secondary X-ray emitted from a sample (called fluorescence) when it is excited by the primary X-ray source. Every

element in a sample produces a set of unique characteristic fluorescent X-rays that allows to determine the chemical composition of materials. The equipment used is a Philips MagiX 2400. The equipment was calibrated with the corresponding standard sample. The analysis of the majority elements was carried out by preparing a bead mixing 0.3 g of sample and 5.5 g of Lithium Tetraborate. Quantification was carried out by using the quantitative analysis curve for silico-aluminous materials. When the concentration of the elements was low, i.e., they were present in trace form, the pressed tablet or pellet method was used.

2.4.Spectral reflectance and degree of lightness

In this study, a spectrometer was used to perform the measurement of the specular spectral reflectance of the specimens. For this type of reflectance measurements, the StellarNet miniature spectrometers family members are suitable, since they are a portable and compact fiber optic instruments for ultraviolet, visible (VIS), and near infrared (NIR) measurements offering CCD 2048 and PDA 512/1024 detectors with the required accuracy for the objectives of this research. The experimental setup was described in Figure 12. For the measurements, a StellarNet BLUE-Wave Spectrometer of STN–BW–VIS type was used, it is a fiber-optic-coupled instrument for measurements in the range of 350–1150 nm wavelength. It uses a 16-bit digitizer via high speed USB-2, and each unit contains a USB-2 interface with a snap shot memory to provide instantaneous spectral image from the highly sensitive CCD or Photo Diode Array detectors. The reflectance probe used was a STN-R600-8-VisNIR type for VIS and NIR (400–2200 nm wavelength) measurements. This probe was assembled in a reflectance probe holder for 90◦ angle measurements and this strand fiber optic cable or probe assembly delivers input via standard SMA 905 fiber optic connector. The experimental setup also contains a light source STN-SL1 type, which is a 10,000 h Tungsten and Halogen lamp, 2800 Kelvin color temperature, 350–2500 nm (Figure 12). This spectrometer equipment was calibrated with NIST (National Institute of Standards and Technology) traceability.

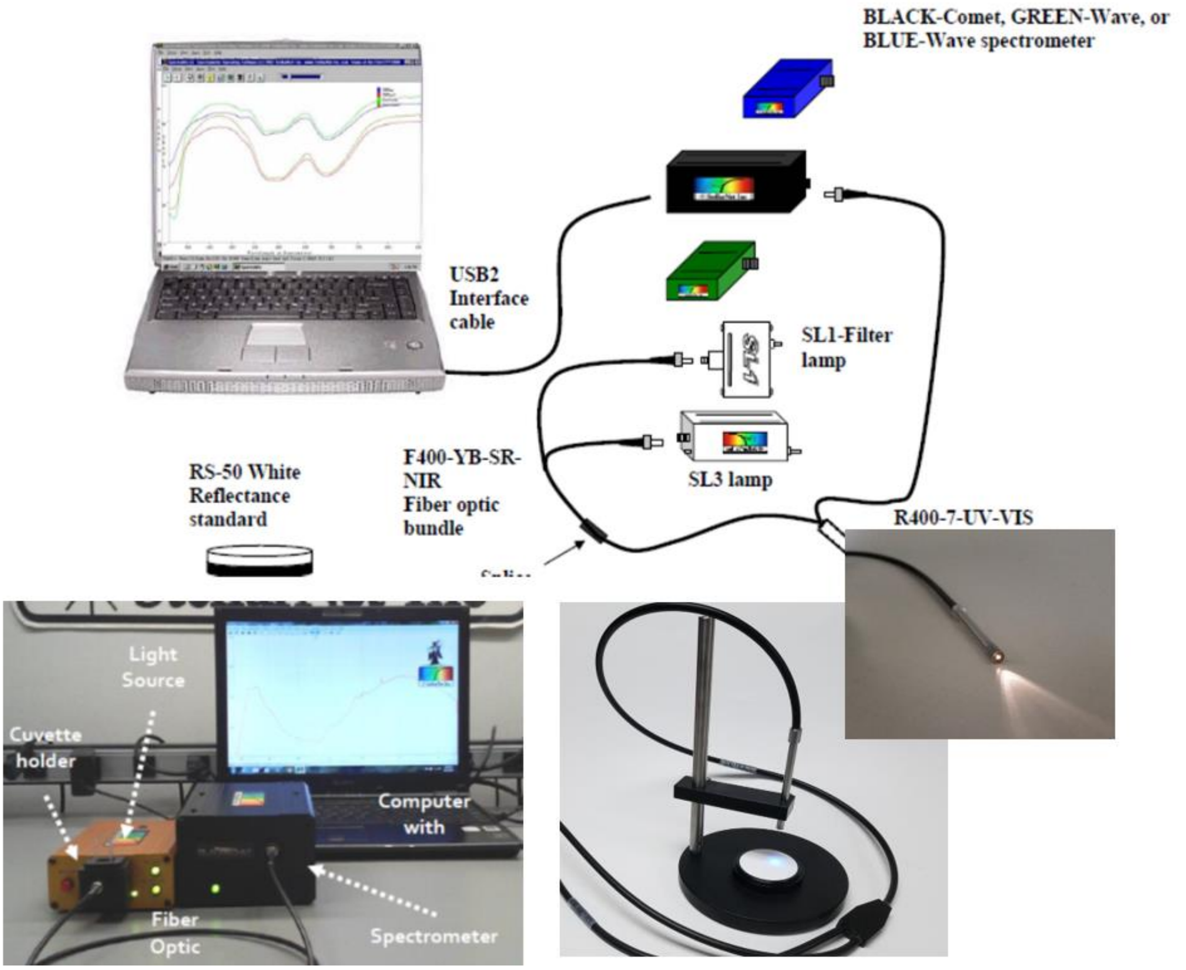


Figure 12. StellarNet BLUE-Wave Spectrometer STN–BW–VIS with the reflectance probe and probe holder for the light spectral reflectance measurements

The equipment also contains the STN-RS50 reflectance standard, which is a 50 mm diameter white reflectance standard made of Halon. It is used to take reference measurements by using the R600-8 Reflectance Probe. The white standard will reflect >97% of the light from 300 to 1700 nm. Data were recorded by using the SpectraWiz software to accurately measure the light reflected intensity and perform other spectral calculations. Once the WG-coated tile specimen was placed in the sample holder in a dark lab room, the experimental reflectance data-collection procedure became as follows:

- Dark spectrum measurement: it records the background noise with the source turned off. Dark spectrum is subtracted from measurements.

- Reference spectrum measurement: it records the reference spectrum with the STNRS50 white reflectance standard.

- Sample spectrum measurement: it records the quotient between the sample reflectance spectrum and the reference spectrum of the RS50 standard.

For each reflectance measurement of the WG coated samples, the number of spectra to signal averaging was set. This option provides a smoothing effect, thus increasing the system signal-to-noise ratio by the square root of the number of scans being averaged. The rule is to set the averaging to the highest number tolerable when there is sufficient light signal, keeping the detector integration time short but out of saturation. In our measurements, the integration time was kept above 30 ms and at least 10 scans were averaged. Since we manufactured three WG-coated tile samples with the same characteristics of holding time, temperature, and thickness, as stated in Section 2.2, the experimental data collection was performed for the 27 pieces, and each measurement was repeated three times with each sample to ensure the quality of acquired data. For all the samples, we measured the specular spectral reflectance in the visible spectral range that extends from 400 to 700 nm and the NIR spectral range from 700 to 1100 nm.

Regarding the degree of lightness denoted as L*, the measurements were conducted according to the CIELAB D65 reference of the French-based international Commission on Illumination (CIE). This CIE lightness degree refers to the relative degree of near white materials under specific lighting conditions, and it correlates the visual ratings of whiteness for certain surfaces compared to the white-surface standard in the visible spectrum range. L* increases with whiteness, reaching the maximum value of 100 for the perfect white sample (Westland, 2014). Finally, with the aim of gaining some insight of data, some measurements were processed through statistical analysis, using SPSS software, details are shown in the Annex. The statistical analysis focused on analyzing the existence of differences between the probability distributions for the different spectral reflectance measurements and lightness degree measurements of the WG coated tiles. The results were interpreted following the specific statistical analysis.

3. Results and discussion

In this section, the results obtained, and the discussion associated are described for the behavior evaluation of WG coated specimens in terms of chemical and visual characterisation, lightness degree and spectral reflectance, that is based on the effect of variables including thickness of WG coating, holding time of burning, WG type, and the temperature of the firing treatment.

3.1. Chemical characterization of raw materials

WG particles used in this study were classified into three types, based on their origin, grain size, and chemical composition (Table 8). The XRF chemical characterization of the WG highlights the presence of silica (SiO_2) that is the main mixed oxide in the composition of glass as well as in the composition of the clay powder that is obtained by mixing two types of raw constituents 40% grea and 60% lime, in addition to the fluxing oxides Na_2O, K_2O, PbO, and stabilizing elements including Al_2O_3, CaO, BaO, and MgO. The first type, denoted as WG1, is a hollow green glass collected from recycled bottles. According to its composition, it is a SLS glass characterised by high concentration of silicon, sodium and calcium oxides. The second type denoted as WG2, contains some flat glass, and it is a SLS glass as well that is characterized with the presence of a small quantity of stones and ceramic materials. The third type denoted as WG3, is a LS glass mainly coming from CRT TV monitors and characterized by high concentrations of silica and lead oxide.

Table 8. Average chemical composition in wt.% of WG and clay obtained from FRX analysis. Please note that they are average values and do not necessarily add up to 100% for each element

	SiO_2	**Na_2O**	**CaO**	**MgO**	**Al_2O_3**	**Fe_2O_3**	**K_2O**	**TiO_2**	**P_2O_5**	**BaO**	**PbO**
Clay	44.46	0.69	11.21	3.63	16.08	5.48	3.34	0.66	0.14	-	-
WG1 [1]	73.2	11.4	10.8	1.35	2.03	0.31	0.88	0.066	<0.04	-	-
WG2 [2]	72	13	9	2	1.75	<0.1	0.55	-	-	-	
WG3 [3]	52.5	6	3	1.75	2.25	0.15	7.5	0.075	-	2	20.5

[1] SLS WG with a particle size of between (0 and 1) mm.

[2] SLS WG with a particle size of between (0 and 3) mm.

[3] LS WG with a particle size of between (0 and 4) mm.

3.2. Qualitative visual characterization of the specimens

As a result of the firing treatment, none of the WG types reached the melting point at 700ºC. At 850ºC, WG1 and WG2 types created a dense coat that did not adhere to the substrate, whereas WG3 showed more densification, however, it did not cover the entire surface. At 1000ºC, all types of WG reached the melting point, and a dense coat structure was formed. The tiles specimens fired at 1000ºC showed better properties in terms of material adhesion when compared to 700 and 850ºC. Accordingly, the 27 specimens prepared at 1000ºC shown in Figure 13 were selected for further tests. All the samples had a homogeneous visual appearance to ensure the reproducibility of data.

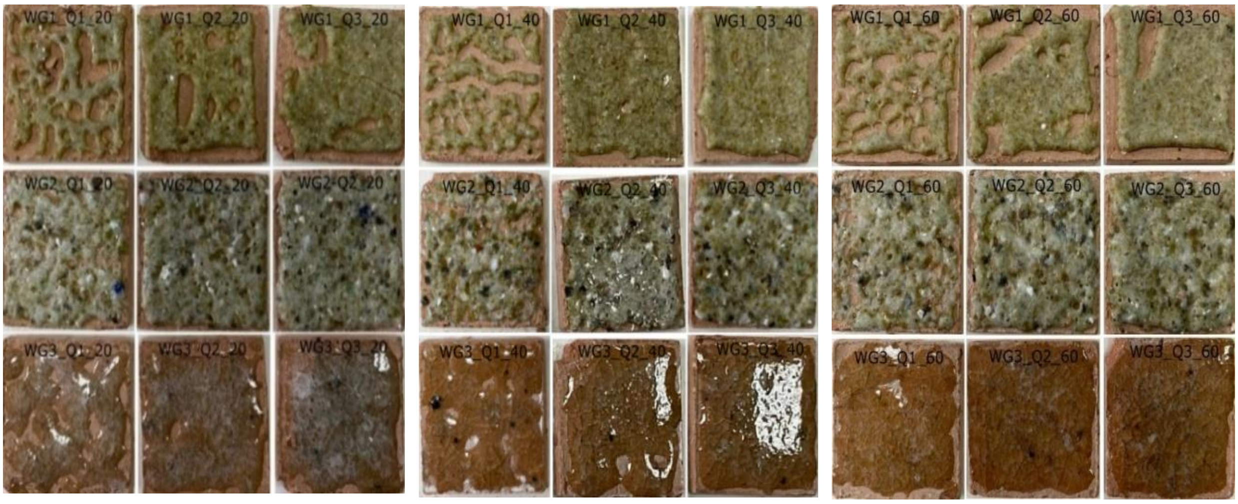


Figure 13. Tiles specimens fired at 1000°C during 20 min, 40 min, and 60 min. In each figure, the first row shows WG1, the second row WG2, and the third row WG3 coated samples respectively. In addition, in each figure, the first column shows Q1 samples, the second column shows Q2 samples, and the third column shows Q3 samples.

The surface coating structure of the three types of WG was different. According to Figure 13, regarding specimens cooked during 20min, WG1 presented a weak transition zone characterized by some cracking, and irregular distribution of melted WG due to particle shrinkage for small quantity of WG that improved with the increase of WG quantity. WG2 showed a porous surface and more roughness than the other types, these qualities were notably reduced at a holding time of 60 min for the second set of specimens (Figure 13). WG1 and WG2 coatings had the appearance of an opaque surface on the substrate, and WG3-coated samples had a glossy surface that is notably transparent, in fact, some breaks or rifts can be observed at the transition zone between the substrate and the coating. Moreover, WG3 coating contained air bubbles and some pinholes occurred due to the release of gases during the chemical reactions. These effects decreased with the increase of the holding time.

the chemical and qualitative visual characterizations of the WG coated specimens showed clear distinguishing features, demonstrating the capability to showcase different behavior for the lightness degree and the spectral reflectance of the specimens.

3.3. Lightness and spectral reflectance characterization

Table 9 shows the values L* describing the degree of lightness for the three WG coated types of tiles specimens. In this table, WG1Q1 stands for specimens with a coating thickness of Q1, and so on for the other cases with WG2 and WG3. According to the results described, lightness degree ranged from 50.45 to 52.93 with an average of 51.78 for WG1, and from 49.24 to 55.23 with an average of 52.93 for WG2. While specimens with WG3 have the highest degree of lightness with an average of 84.49 with regard to the other cases.

Table 9. Degree of lightness (L*) of the 27 specimens

Burning Time	**L***								
	WG1Q1	**WG1Q2**	**WG1Q3**	**WG2Q1**	**WG2Q2**	**WG2Q3**	**WG3Q1**	**WG3Q2**	**WG3Q3**
20 min	51.76	50.69	50.56	55.23	52.56	51.28	61.39	97.33	100
40 min	52.93	52.18	52.16	53.86	54.8	53.25	71.46	83.22	100
60 min	52.41	50.45	52.88	49.24	54.78	51.45	54.54	92.5	100

According to the statistical analysis, the two variables, holding time and thickness, do not have a significant influence on the lightness degree of the specimens. However, concerning the type of WG, the results showed that the choice of WG significantly influences the response of the degree of lightness. According to these results, the composition of WG had a significant influence on the lightness degree with WG3 specimens registering the highest mean value (84.5) compared to the other types, the different concentration of the oxides in the composition of WG influences the whiteness of the specimens. According to Samoilenko et al., the whiteness indicator of an engobe containing WG was influenced by the ratio of kaolin, alumina and zirconium. Moreover, the smooth surface of the WG3 specimens contribute to the decrease of the light scattering that increases the lightness.

Regarding the spectral reflectance measurements, Figures 14 to 16 show the different light spectra of reflectance obtained for the three types of WG-coated specimens. Each figure represents the relative specular spectral reflectance of the WG1, WG2, and WG3 coated specimens with different holding times of burning. For instance, WG1T20 stands for the WG1 coated specimens with a holding time of 20 min (Figure 14), and so on for the other cases of 40 min (Figure 15) and 60 min (Figure 16).

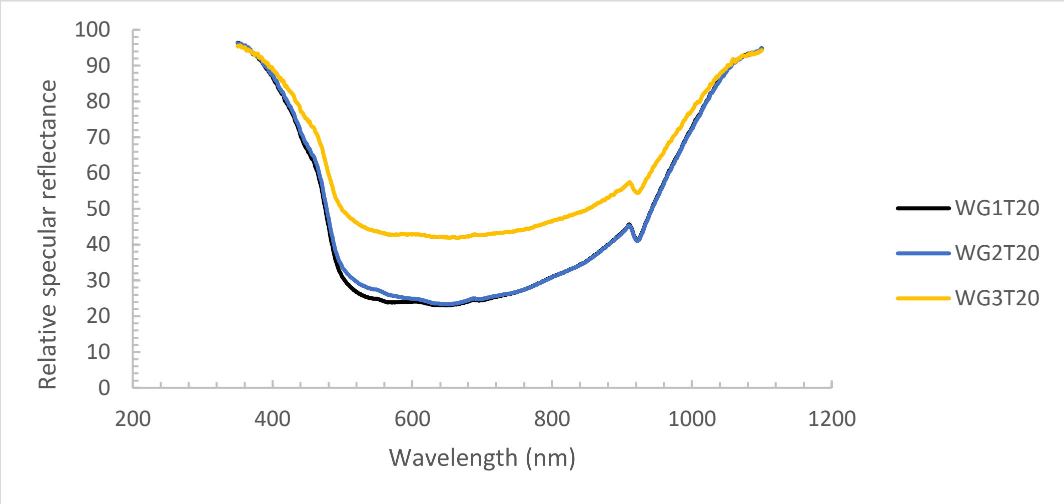


Figure 14. Specular reflectance of the first set of specimens fired at 1000°C during 20 min

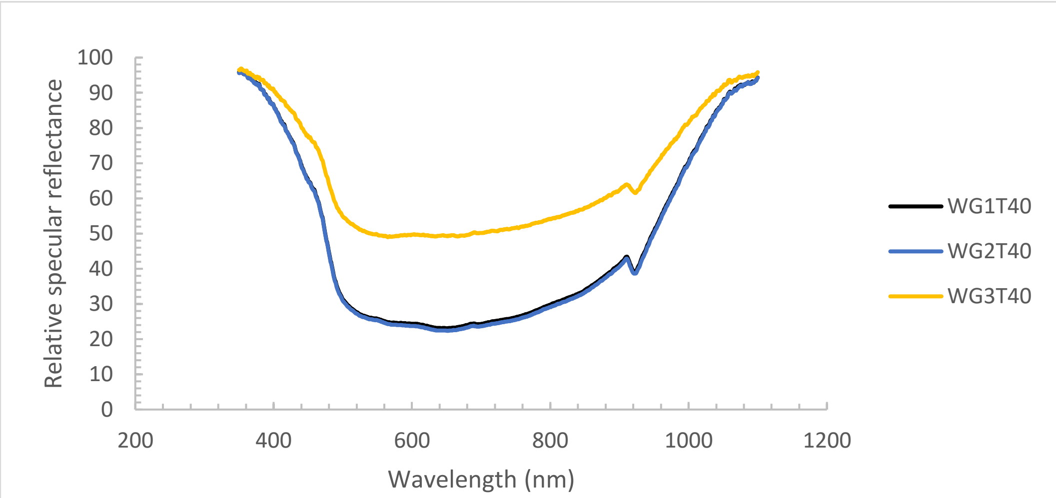


Figure 15. Specular reflectance of the first set of specimens fired at 1000ºC during 40 min

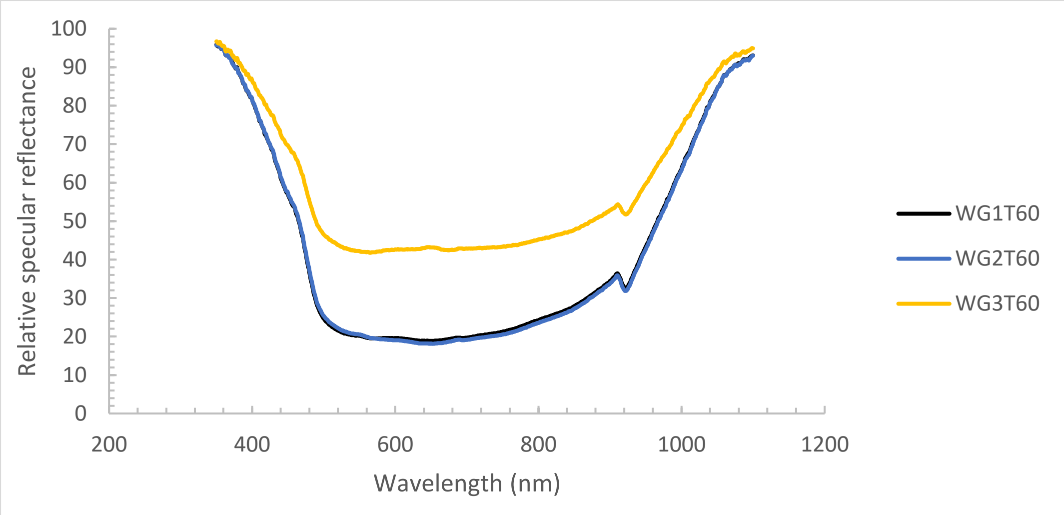


Figure 16. Specular reflectance of the first set of specimens fired at 1000ºC during 60 min

As shown in Figures 14, 15, and 16, all specimens reached a minimum light spectral reflectance in the range between 500–900 nm, which tends to increase through the rest of the visual range and gradually in the NIR portion. According to the results, WG3 specimens showed a high spectral reflectance with regard to the other types which is in accordance with the lightness results analysis, and WG1 showed the least spectral reflectance. The spectral range between 500–900 nm, which majorly corresponds to the visible range portion (yellow, orange, and red colors) and a small portion of the near infrared, is the one showing almost constant and stable values of the relative spectral reflectance, considering the extremities of the spectrum data as representative of background noise that should be discarded. Therefore, this range present the most interesting part to fulfil the aim of this evaluation which is the identification of the potential influence of the WG type and the holding time on the spectral data through the statistical analysis. According to the results of the nonparametric Mann–Whitney U tests analysis for the spectral reflectance behavior of the specimens, a significant difference was found between all types of WG coatings (WG1, WG2, and WG3) regardless of the holding time ($p < 0.001$). Regarding the holding time influence, there was a significant difference between the groups of 20 and 60 min and between 40 and 60 min for WG1 with $p < 0.001$, unlike between the two groups of 20 and 40min. For WG2 and WG3 specimens, a significant difference was found between all the holding times with $p < 0.001$. Regarding the thickness of WG coatings, the statistical results showed that the spectral reflectance of the specimens is not influenced by the thickness, for all types of WG used.

This proof of concept about the spectral reflectance behavior of the WG coating showed that the spectral reflectance is highly influenced in the spectrum range that extends from 500 to 900 nm due to the composition of the three types of WG used in this study. Moreover, the holding time can influence the response of the solar reflectance as well, while the parameter of thickness for the WG coatings has no impact on the solar reflectance. The refractive index of the chemical compositions of the specimens may influence the reflectance behavior of the specimens depending on the wavelength range studied, in fact this effect was reported in other applications where the use of WG in the production of engobes and glazes compositions increased the refractive indexes (Bloomfield & Rothschild, 1999; Dal Bó et al., 2014; Kalirajan et al., 2016). Moreover, the difference of the spectral reflectance performance of the specimens could be explained by the percentage of the crystalline phase developed through the sintering process of each type of WG, according to the visual characterisation, the holding time of 60 min promotes particles bounding and densification which provides a relevant adherence of the coatings to the substrate.

4.Conclusions

This work presents a preliminary study for the evaluation of the spectral reflectance performance of waste glass (WG) coatings for tiles specimens. An optical characterization in terms of lightness degree and spectral reflectance properties of the WG coated tiles specimens is provided by testing the relevance of variables including the WG types and other manufacturing features, such as the holding time, temperature of burning and thickness of the coatings. The chemical composition of the three types of WG used and the temperature of the firing treatment had noticeably a significant influence on the degree of whiteness and spectral reflectance of the specimens.

The holding time influences the sintering behavior of the specimens and the solar reflectance as well, in this study the holding time of 60min showed the optimal results with the firing temperature of 1000ºC that provided the appropriate adherence between the coatings and the substrate.

Coatings with LS WG3 had the highest mean value of the lightness degree and the spectral reflectance compared to the SLS WG1 and WG2, it is noteworthy to mention that WG1 provided the least feasibility as a coating in terms of adhesion, lightness and reflectance performance. Moreover, the varying parameters including the holding time and thickness of WG coatings have no significant influence on the lightness degree.

This work provides a reference for future studies as a first proof of concept of the use of SLS and LS waste glass as coatings for clay tile specimens. The results of this study concerning the spectral reflectance behavior aligns with the literature results that promotes using secondary materials to enhance the spectral reflectance performance, as several studies investigated the use of WG for the replacement of raw materials used in the production of glazes which showed promising results reducing the extraction of raw materials and consequently decreasing energy consumption. Accordingly, an extended study for the real shape of tiles should be conducted, to further evaluate the spectral reflectance performance using LS WG for cool roof applications.

CHAPTER 3. Characterization of roof tiles with waste glass coatings for cool roof applications

1. Introduction

Due to progressive urbanization worldwide, the construction industry is facing challenges in producing materials with a sustainable life cycle and quality performance. As aforementioned in the Background part, 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. The development of such materials with solar radiation reflective properties showed promising results in the mitigation of urban heat island. The most discussed application is cool roof (Mourou et al., 2022a), as the heat-gain through the roof represents about 50-60% of the total heat gain in the building (Santamouris, 2016), while the cooling energy represents 2.9% to 6.7% of the world energy consumption (Rawat & Singh, 2022). This cooling strategy significantly reduces energy demand for cooling loads in air-conditioned houses in hot climate regions (Asadi et al., 2015; Kolokotroni et al., 2018). Recent study indicates that buildings using cool roofs achieve energy savings varying from 15.0% to 35.7% in different climatic zones (Rawat & Singh, 2022). 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 (Pisello et al., 2014).

On the other hand, the world economic growth has caused an increase in the waste production (Chunfa et al., 2007), and the scientific community is in 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 (Ferdous et al., 2021). 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 (Bhavsar & Panchal, 2022; Cardoso de Souza-Dal Bó et al., 2021; Costa et al., 2009; Darweesh, 2019; de Azevedo et al., 2020; Dondi et al., 2009; Fedaoui-Akmoussi et al., 2023; Grdić et al., 2022; Hamada et al., 2022; Kadhim et al., 2022; Pitarch et al., 2021; Shayan & Xu, 2004; R. V. Silva et al., 2017). In this regard, according to 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, in a complete or a partial substitution for different percentages and size (Cardoso de Souza-Dal Bó et al., 2021; R. V. Silva et al., 2017). 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 (Darweesh, 2019). In terms of pozzolanic properties, the use of WG in concrete as for partial substitution for cement or sand indicated favorable results in terms of rigidity and durability (Fedaoui-Akmoussi et al., 2023; Hamada et al., 2022; Shayan & Xu, 2004), it improves the compressive strength of cement mortar cubes in an ultrafine material form with 10% of cement replacement (Balamuralikrishnan & Saravanan, 2021). More specifically regarding the type of WG used, the one derived from spent fluorescent lamps was successfully used to replace Portland cement in mortars (Pitarch et al., 2021), Dusan et al. (Grdić et al., 2022) 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 showed significant benefits in constructing environmental friendly pavements (Huang et al., 2007; C. Li et al., 2023; Ming et al., 2022; Peng et al., 2023).

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 (Andreola, Barbieri, Corradi, & Lancellotti, 2007). 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 (Gol et al., 2021). According to Andreola et al. (Strecker & Costa, 2014), up to 30% of recycled WG powder could be incorporated into glazes. More recently, Cardoso et al. (Cardoso de Souza-Dal Bó et al., 2021) 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. (Andreola et al., 2007) found that the use of CRT glass as a substitute for ceramic glaze frits reduces the environmental impact of the ceramic glaze production process, moreover, it decreases the production expenses with a substitution of 25 wt.% (Karaahmet & Cicek, 2019). In this regard, Revelo et al. (Revelo et al., 2018) found that the optimal amount to maintain the properties expected for ceramic glazes is 20 wt.%. In all cases, the potential use of WG in ceramic-based materials showed in the aforementioned studies encourages the recycling of WG which helps alleviate the pressure on 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 the shape of the roof tiles and the 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.

This study builds on the previous findings of the preliminary study in Chapter 2, where a characterization of clay specimens 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 optimal results obtained from the characterization of these different samples clay tiles reported in Chapter 2, i.e. using the two types of WG that showed feasible results which are the lead silicate (LS) WG and the second type of soda lime silica (SLS) WG and, considering the firing treatment temperature of 1000ºC and a holding time of 60 min, the thickness of the coatings was chosen to be less than 1mm .

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.

1.1.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: SLS (WG1) derived from recycled hollow glass and LS (WG2) derived mainly from CRT of television sets. The chemical composition of WG is shown in Table 10. The used particle size of WG was reduced and passed through a 0.63mm 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.

Table 10. 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 .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

1.2.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, keeping the shape and scale. This does not truly present a limitation as the measurements remain unaffected by the actual size of the specimens, given that they are scaled accordingly. Table 11 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 (Figure 17). 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 hours; the second stage was for the WG particles application with a thickness of coating less than 1mm and a quantity of 450g/m² on the exterior surface of the tiles after applying an intermediate engobe coat to ensure the adhesion of WG particles to the tiles, the tow coats were 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 60min with a heating rate of 1.22ºC/min that takes up to 1.5h to get to 1000ºC. 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 1mm 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 this work.

Table 11. Identification code of the configurations of tiles based on the shape of tiles and type of WG

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**

Figure 17. 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

2.3. Tiles characterizations

For the experimental part, the technical characteristics described in the standard UNE EN 1304 (UNE-EN 1304, 2020) which includes geometrical characteristics, flexural strength, permeability, frost resistance and fire behavior were measured. In addition, a leaching test (EN 12457-1, 2003), and the colorimetric and normal solar radiation reflectance characterization through the VIS and NIR solar radiation spectrum (Standard, 1996) were performed. Table 12 shows the tests conducted according to the above standards and the samples used for each one.

Table 12. 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 (UNE-EN 1304, 2020)	All Tiles
Flexural strength	UNE-EN 538 (UNE-EN 538,1995)	3 samples of each tile
Permeability	UNE-EN 539-1(UNE-EN 539-1, 2007)	3 samples of each tile
Frost resistance	UNE-EN 539-2 (UNE-EN 539-2, 2013)	3 samples of each tile
Fire behavior	UNE-EN 1304 (UNE-EN 1304, 2020)	All Tiles
Leaching test	UNE-EN 12457-1 (EN 12457-1, 2003)	Ratio of 2l/kg
Colorimetric characteristics	CEI EN 60335-2-27	3 samples of each flat tile
Solar reflectance	ASTM E903 (Standard, 1996)	3 samples of each tile

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.

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 350-1100nm, taking measurements for the VIS and NIR radiation according to ASTM E903 standard for laboratory measurements (Standard, 1996). All the experimental set up is presented in Chapter 2 (Mourou et al., 2022b). 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 tile: 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 (Figure 18). 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 at all points where the beam hit the surface. Subsequently, the data were processed using the Spectrawizz OS v5.33 software.

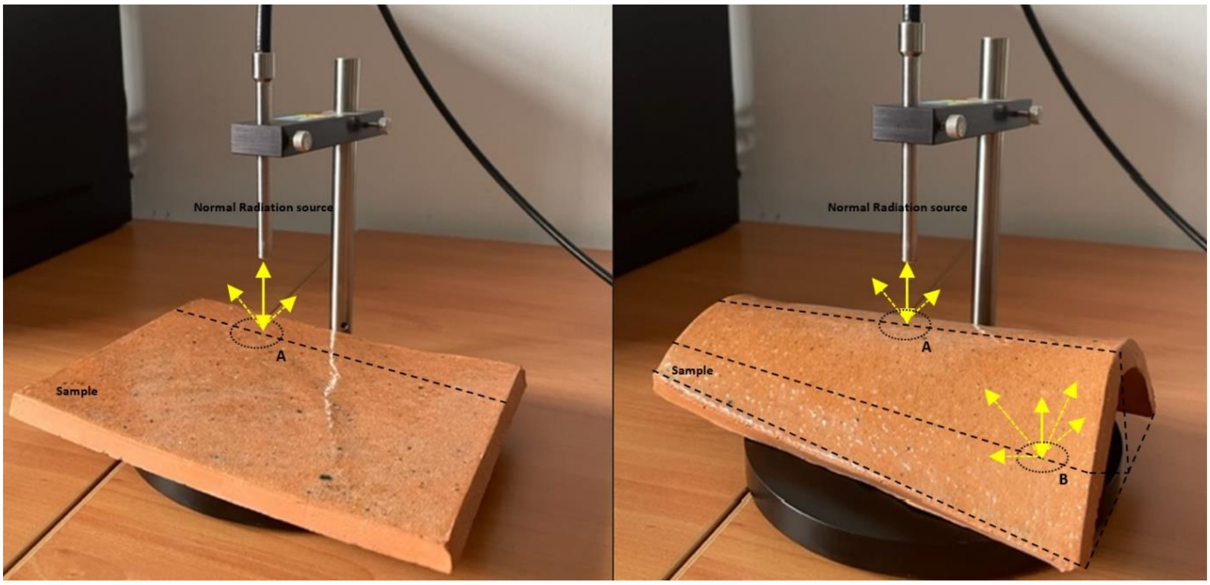


Figure 18. Normal radiation reflectance experimental set-up using the Stellarnet spectrometer at different points of a curved and a flat tile

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:

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

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 13. 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.

Table 13. 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

For the analysis of the specimens, it is interesting to mention the difference in the visual appearance of the tiles with WG1 and WG2 coatings and the reference. As it can be seen in Figure 17, 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.

5.2.Geometrical characteristics

The geometrical characteristics for all the tiles used in this study conform to the standard. More specifically, tiles covered with WG 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 use does not impact the geometrical characteristics requirements.

5.3.Flexural strength test

According to the results of the flexural strength test (Table 13), 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 WG in the fabrication of ceramic construction materials generally increases the flexural strength, while decreasing the particle size of glass (Bernardo et al., 2012; Furlani et al., 2011; Lu et al., 2014; R. V. Silva et al., 2017). In a study conducted by Dusan et al. (Grdić et al., 2022), 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.

5.4.Permeability

For the permeability test, all the specimens showed high impermeability (Table 13), 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 noticeable 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 (Lazareva et al., 2018).

5.5.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 13). 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 (Christogerou et al., 2021; Raimondo et al., 2009), it can be affirmed that the tiles studied are able to withstand adverse climatic conditions, and subsequently presents a positive durability potential.

5.6.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.

5.7.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, 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 2l/kg (water/solid) was adopted.

According to the ASTM D3987 (ASTM D3987, 2012), lead ratio in the sludge sample obtained from tiles must be lower than 5 mg/l (UNE-EN 1304, 2020), As it can be seen in Table 14, 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 with LS WG2 are lower than the SLS 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 14, 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 LS WG was null. In general, the low toxicity levels obtained when using LS 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 (Dondi et al., 2009; Raimondo et al., 2007; R. V. Silva et al., 2017).

Table 14. 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	5

5.8.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 Figure 19 and Table 15. These results indicate that tiles covered with WG2 have the highest lightness which increased by 41% 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 Figure 17, 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 (Cardoso de Souza-Dal Bó et al., 2021), 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 Chapter 2 (Mourou et al., 2022b), 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 (Arunraj et al., 2020; Bretz et al., 1998; Kiletico et al., 2015; Xie et al., 2019), which makes the application of WG2 coatings on clay roof tiles the better potential candidate with regard to WG1 for cool roofs applications.

Table 15. 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 (41%)	+8.723	-1.598	8.87	-10.38

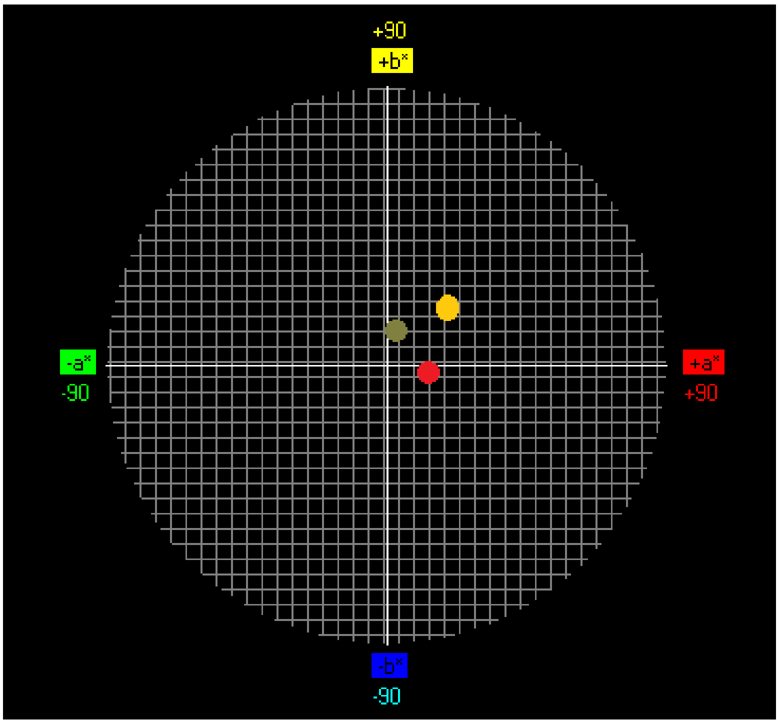


Figure 19. Lightness coordinates of the three configurations: tiles with WG1 (green), tiles with WG2 (Red) and tiles without WG for reference (Yellow)

5.9.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 Figure 20 and 21 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 1000nm in the NIR range.

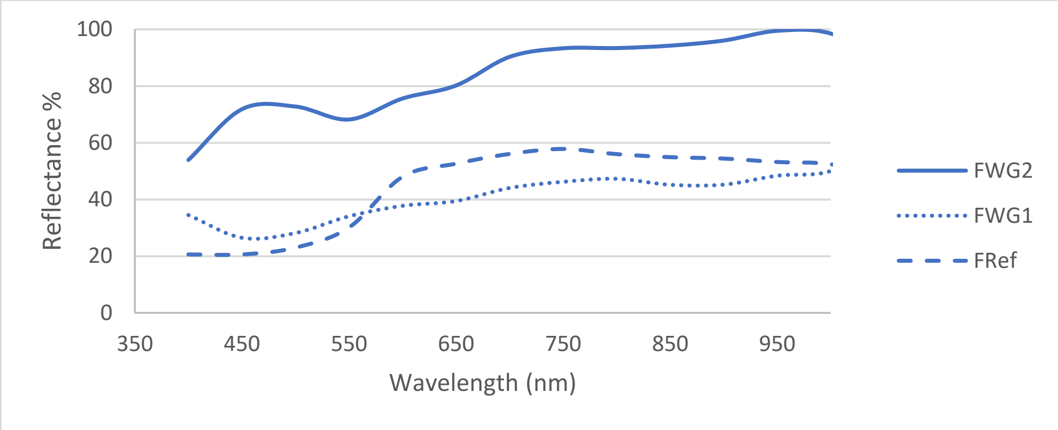


Figure 20. The spectral solar reflectance profiles of FWG1, FWG2 and FRef tiles

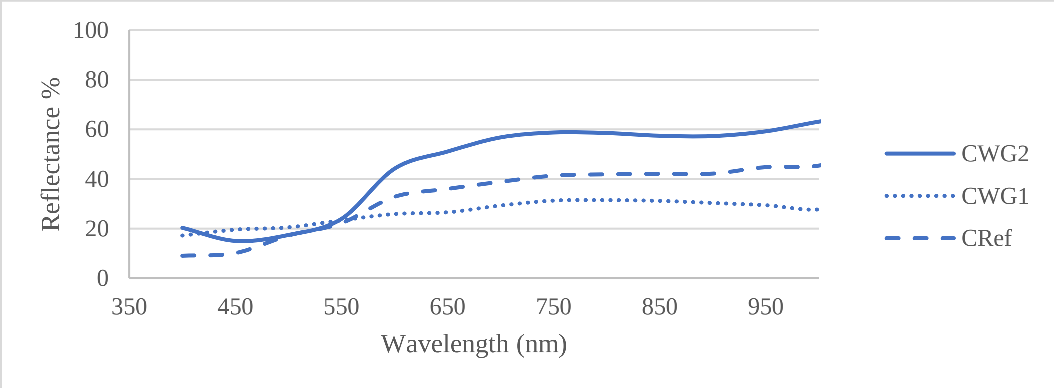


Figure 21. The spectral solar reflectance profiles of CWG1, CWG2 and CRef tiles

In the range between 400-550nm 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 700-1100nm 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 1000nm, which represents the relevant solar spectral range between 700nm and 1100nm in the NIR range. The results are reported in Table 16, 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 16, 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 90% with regard to FRef, whereas the solar reflectance of FWG1 decreases by 7% with regard to FRef. For the curved tiles, the application of WG2 registered the highest solar reflectance as well with an increase of 36% 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.

Table 16. 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 (90%)	0.3	0.26 (-13%)	0.41 (36%)
Reflectance at 1000nm	52	50 (-4%)	98 (88%)	45	28 (-38%)	63 (40%)

Conversely to flat tiles, the spectral profiles of curved tiles show almost the same behavior for much of the spectral range, according to Figure 20, all samples overlap around 550nm 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 400-1000nm, the U Mann-Whitney statistical test was performed for the averages of the spectrum range (Table 8). According to Table 17, 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.

Table 17. Mann-Whitney U to identify the difference in the overall distribution of reflectance across groups of tiles through the spectrum range 400-1000nm 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)	.000	.168	.000	.024	.035	.022

The application of WG2 significantly increases the solar reflectance of tiles in accordance with the results found in Chapter 2, 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 13, 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 (Cardoso de Souza-Dal Bó et al., 2021). According to Xie et al. (Xie et al., 2019), 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. (Kiletico et al., 2015) 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 (Arunraj et al., 2020; Bretz et al., 1998). 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, a detailed analysis was conducted for the spectral reflectance readings of one curved tile in nine different points. Taking into consideration the fact that curved and flat tiles have the same type of WG still flat tiles reflects more, by evaluating the response of each point of a curved tile (Figure 22); it was found that at the highest points (P1, P2 and P3 in Figure 22) of the curved tile where the radiation hit normally as it is shown in Figure 18 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 Figure 18 (P4, P5, P6, P7, P8 and P9 in Figure 22), 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. (F. Rossi et al.,

2015) presented an example of the reflectivity behavior of a white diffusive material according to different angles of incident radiation.

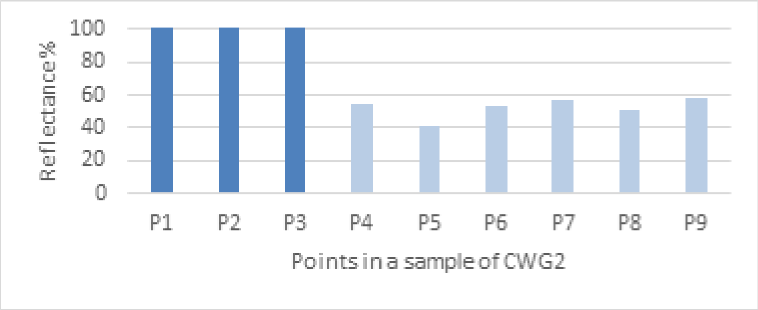


Figure 22. Normal radiation reflectance of a curved tile sample from nine different points, the first three points (P1, P2, and P3) define the top surface line of the tile, according to Figure 18

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 surface's structure and color, in addition 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 (Cao & Sendur, 2019), the surface roughness significantly affects 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 (Johannes A. Lercher, 2007). Xie et al. (Xie et al., 2019) 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.

6. 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 work was based on the characterization of a novel utilization of WG for 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 0,63mm and a thickness less than 1mm. Soda lime silica waste glass (WG1) derived from recycled hollow glass and lead silicate 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:

1. 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.

2. 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.

3. 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 LS 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 41%.

- 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 350-1100nm, with an increase of 90% and 36%, for flat and curved tiles respectively, with regard to the reference.

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 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.

CHAPTER 4. Evaluation of the environmental impact of roof tiles with waste glass coatings through an LCA approach

1. Introduction

For the fabrication of construction materials, the environmental burdens associated with the energy consumption and raw materials extraction are main concerns highly addressed by the scientific community, in an attempt to provide sustainable solutions and alternatives for the stakeholders and actors in the decision making and policy processes. According to the Intergovernmental Panel on Climate Change (IPCC) (IPCC, n.d.), the increase of greenhouse gas (GHG) concentration in the atmosphere has reached 417.1 ppm CO_2eq in 2022, which should be decreasing to help limiting the increase of temperature to 1.5 Cº by 2100. In this context, the ceramic industry represents one of the major responsible of GHG emissions due to the energy used in kilns for firing and drying processes (Mezquita et al., 2017; Quinteiro et al., 2022; Ros-Dosdá et al., 2018; Wang et al., 2020). According to Mohaddes et al., the fabrication of glazes used for roof tiles requires high energy consumptions due to the melting process of raw materials which is considered the most impacting process (Mohaddes Khorassani et al., 2020). Moreover, the application of engobes and glazes increases the global warming potential up to 7.2% for the fabrication of ceramic facing bricks (Silvestri et al., 2021).

In that sense, to evaluate the impact of different materials and services on the environment, health and wellbeing, several studies have used the Life Cycle Assessment (LCA) methodology as a part of the design phase of buildings to support the decision-making process (Abd Rashid & Yusoff, 2015; Atılgan Türkmen et al., 2021; Suppa et al., 2022). It was developed, starting from an energy analysis, to then evolve into an environmental analysis tool, and it subsequently expanded its scope of application to include economic and social aspects in a comprehensive Life Cycle Sustainability assessment (Guinée et al., 2011). The objective of an LCA instrument is to help reduce the environmental burden and provide all insights of the processes to identify the contribution associated with the environmental burdens (Fnais et al., 2022). According to Matthews et al. (Matthews et al., 2014), LCA is a quantitative tool used to enhance the environmental sustainability of a product. By using several indicators that focus on energy consumption and carbon emissions, it is considered as a valuable knowledge supporting stakeholders to ensure the carbon neutral transition by predicting the operational carbon emissions during the early-design stages (Suppa et al., 2022).

According to ISO 14040 standard (ISO 14040, 2006), LCA is based on a methodological framework including goal and scope definition, life cycle inventory analysis, life cycle impact assessment and life cycle interpretation. These steps are followed to assess the production phase or the use phase of a product or a service. Most studies consider a cradle to gate analysis and base their LCA on the production phase, mainly characterized by the embodied energy of the materials used. The use phase is characterized by the operational energy resulted mainly from the heating and cooling processes (Kulatunga et al., 2020; Kuruppuarachchi et al., 2014; Quinteiro et al., 2022). According to Rossi et al. (B. Rossi et al., 2012), the operational phase produces the largest environmental impact. However, the LCA may present some limitations associated with the collection of appropriate and representative inventory data, as well as with the time considered for the analysis (Fnais et al., 2022; F. Silva et al., 2020).

In this work, an LCA analysis was conducted to assess the environmental impact of using waste glass (WG) coatings for roof tiles, and the potential of this solution in decreasing the cooling loads. Spain has been chosen as the study location, due to its diverse climate, enabling the findings and conclusions to serve as a reference in other regions for the implementation of sustainable coatings, that have a two-fold potential in waste valorisation and decreasing energy consumption. In this context, according to literature, the implementation of cool roofs in Spain helps decrease the cooling loads of buildings depending on local climatic conditions (De Carli, 2007; Dominguez-Delgado et al., 2020). In fact, buildings using cool roof achieve energy savings varying from 15.0% to 35.7% in different climatic zones (Rawat & Singh, 2022). Hence, the solution discussed in this work could contribute to the cooling energy savings in buildings (decreasing the operational energy), and present a sustainable product that promotes recycling WG which minimise landfill disposal. Moreover, it is expected that a slight disparity will be observed between the embodied energy resulting from the production of roof tiles with WG coatings and conventional roof tiles without WG coatings. On the other hand, according to literature, the recycling processes and the use of WG as a partial substitute for virgin materials reduces the CO_2eq emissions of the whole manufacturing processes (Aslani et al., 2023; Blengini et al., 2012; Tushar et al., 2023), which makes WG a feasible alternative for raw materials partial substitution. However, this work will perform a new aspect of using WG for roof tiles.

The objective of this study is conducting an LCA analysis that is based on the comparison between three scenarios: (i) conventional roof tiles without recycled WG coating (Scenario 0), (ii) roof tiles with first type WG1 coating (Scenario 1), and (iii) roof tiles with second type WG2 coating (Scenario 2). In order to fulfill the aim of the study, the LCA analysis was performed using Simapro software following the principles, requirements and guidelines stated by the standards ISO 14040, and 14044(ISO 14040, 2006; ISO 14044, 2006). The study was identified and developed through the four LCA steps: scope and definition, inventory analysis, impact assessment and interpretation of the results through the discussion of the findings. Those steps include the production phase of the roof tiles in a gate-to-gate system. Finally, a complementary annual cooling energy simulation was conducted for the use phase of the roof tiles implementation in a model building, highlighting the potential use of these WG coatings for cool roof applications.

2. Materials and methods

In this section, the methodological steps and tools employed in conducting the environmental LCA study are outlined. These include defining the study's goal and scope, preparing an inventory analysis, and performing the impact assessment in addition to the cooling energy simulation. These steps collectively enable the interpretation of the study's findings.

2.1.Goal and scope definition

The analysis of the environmental impact of using WG coatings for flat roof tiles has been conducted through an LCA study, based on the comparison between conventional roof tiles without recycled WG coatings representing scenario 0, and roof tiles with recycled WG coatings, considering two types WG1 and WG2, representing scenario 1 and scenario 2 respectively (Figure 23). The comparison will help to identify the environmental impact of each case, showing the factors that will contribute to the decision making for the optimal solution. The functional unit of the study, which represents the performance characteristics of the system defined in a unit, is defined as $1m^2$ ceramic tile, commonly adopted in buildings, as stated in many studies (Ferrari, 2019; Kulatunga et al., 2020; Peiris et al., 2017; Quinteiro et al., 2022). For the system boundaries, the production phase of the study is a gate-to-gate system, as it is shown in Figure 23 and Table 18, including the processes of the production of roof tiles with and without recycled waste glass. For the use phase analysis of the roof tiles, the annual cooling energy simulation of a common building is considered. The application process of WG coatings has been discarded, as well as the procedures related to transport, considered as non-significant limitation of the study (Atılgan Türkmen et al., 2021).

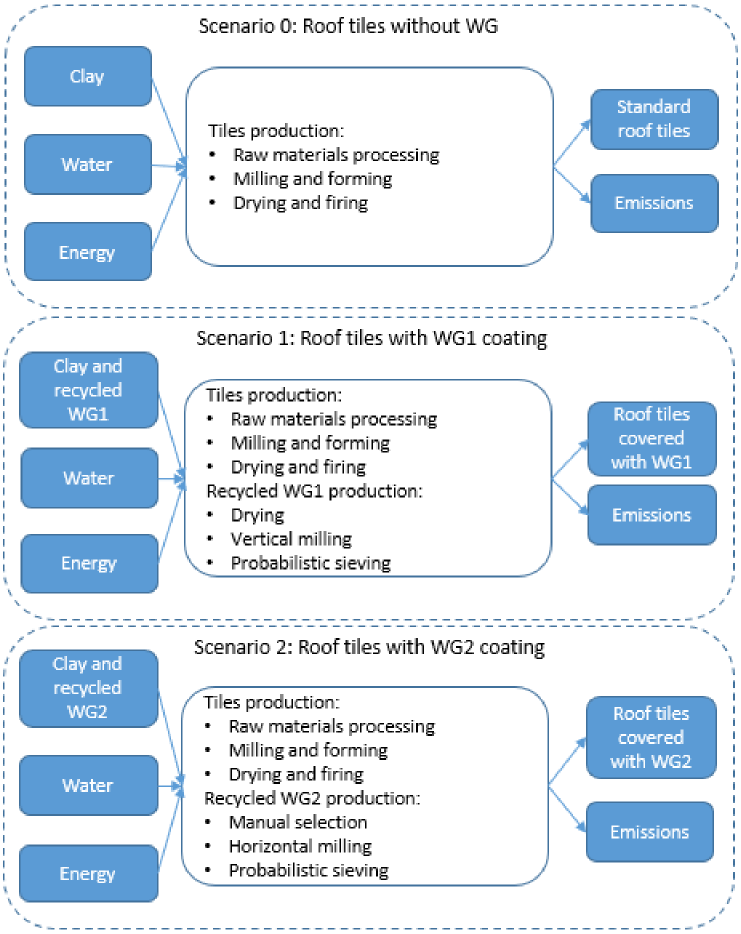


Figure 23. The LCA boundaries of the three scenarios of roof tiles

2.2.Inventory analysis

This step of the LCA study is dedicated to data collection and all the inputs and outputs parameters definition. The study has been articulated in two major processes at two different plants: the production of roof tiles and the production of recycled WG. As shown in Table 18, the stage (P1) of the production phase of roof tiles includes three major steps: raw materials processing, milling and forming, drying and firing. The related data were based on literature data (Kulatunga et al., 2020; Peiris et al., 2017). Regarding the WG recycling stage (P2), it relies on the primary data provided by the glass-collection plant "Camacho Recycling" located in Albacete, Spain. The company has developed glass-collection systems for all types of glass, regardless of their origin, composition, and quantities generated by factories or homes. The treatment of WG undergoes a line that consists of a manual selection of impurities, drying, milling and a sieving process to obtain the required size of WG particles. For the emissions of this process, the data was based on the Ecoinvent database.

The stage (P3), defining the use phase, is described in the following section "Cooling energy simulation".

Table 18. LCA processes and subprocesses description considered for the study

Process	Designation	Product stage	Sub-process
Production phase	P1	Flat roof tiles	Raw materials processing Milling, forming Drying, firing
	P2	Recycled WG	Drying, milling, sieving
Use phase	P3	Tiles covered with WG	Operational energy simulation

The production of roof tiles, which process is based on literature data (Atılgan Türkmen et al., 2021; Kulatunga et al., 2020; Kuruppuarachchi et al., 2014; Peiris et al., 2017; Quinteiro et al., 2022), consists at first stage on the collection and processing of raw materials, mainly clay and water. For this study 51,7 kg/m² of clay and 21kg/m² of water per FU were considered. On the second stage, the clay is treated through milling and mixed with water to form the required shape of tiles. Finally, the third stage consists of the drying process of tiles and subsequently the firing treatment that is based on a single firing process for the substrate. The total energy consumed during this process is estimated by 3.6 kWh/m² (Electricity) and 4.2 kg/m² (LP Gas). Scenario 0 (Figure 23) ends at this point, whereas for scenario 1 and scenario 2, before firing treatment, the recycled WG coatings are added. For this LCA study, the application process is not considered.

For the recycled WG particles used in scenario 1 (Figure 23), the process includes the use of a soda lime silica WG derived from recycled hollow glass (WG1). The treatment of this type follows three stages starting with a gas-based drying process, then a vertical axis milling followed by a sieving process for a particle size of (0.0-3.0 mm). The production rate was estimated by 11t/h and all the treatment process was provided by an energy consumption of 122kW/h. For scenario 2 (Figure 23), it was based on the use of a lead silicate WG (WG2) derived from CRT television sets. For the treatment of this type a manual selection of impurities was necessary to avoid the parts requiring special chemical treatments, this step was followed by a horizontal axis milling, unlike the vertical mill used for WG1, the horizontal mill is used for cutting heavier and deeper grooves into the material. Subsequently, the sieving process was performed to obtain a particle size of (0.0-4.0 mm). The production rate was estimated by 7t/h and the energy needed for the treatment by 73kW/h. Per FU, a quantity of 0.450kg of WG was considered and the particle size was reduced to less than 0.63mm.

2.3.Impact assessment

The impact assessment method represents the environmental footprint of the products, according to ISO 14040/14044 standards (ISO 14040, 2006; ISO 14044, 2006), and it is basically structured as follows: Characterization, Damage assessment, Normalization and Weighting of the impact. The software Simapro provides different impact assessment methods. For this study, the method CML-IA baseline V3.08 was considered (Guinee, 2002)(Gomes et al., 2019), in accordance with the European standard recommendations EN 15804:2012 + A1 2013 that defines the following impact categories for the assessment of the environmental impact of construction products: Abiotic depletion, abiotic depletion (fossil fuels), global warming (GWP100a), ozone layer depletion (ODP), photochemical oxidation, acidification, eutrophication, in addition to human toxicity, fresh water aquatic ecotox, marine aquatic ecotoxicity, and terrestrial ecotoxicity.

2.4.Cooling energy simulation

In order to identify the effect of using roof tiles with WG coating on the energy cooling consumption in buildings, an energy simulation was performed using Design-builder v7.0.2.006, representing the stage (P3) of the study (Table 18). This simulation tool provides several analysis within one modelling environment, and allows the assessment of the performance of a new or an existing building. Several features could be used during the study and provides sustainable building certifications such as LEED and BREEAM (Building Research Establishment Environmental Assessment Methodology). For the objective of this study, the software used the energy simulation engine Energy Plus 9.4. This study is focused on the effect of roofing materials on the cooling energy consumption; hence a simple model HVAC (heating-ventilating-air conditioned) was considered.

Solar reflectance measurements were conducted for the tiles at a previous stage of the research (Mourou et al., 2023). The use of WG2 coatings for flat tiles presents a better option for the tile's normal solar reflectance with regard to WG1, which increases the solar reflectance by 90%. Regarding the emissivity of the tiles, an estimation based on laboratory measurements was conducted. Data acquisition of ten tiles of each scenario was carried out using a T450bx FLIR infrared camera for the measurements. The test procedure was based on the method developed by Bauer and Moldenhauer (Bauer & Moldenhauer, 2004). The results details of the test are shown in the Annex.

To compare the effect of the three scenarios on the cooling energy consumption in different weather areas, the location of the model building is assigned to different climate zones in Spain. According to the Spanish Technical Building Code (CTE), there are 15 climate zones (α3, A2, A3, A4, B2, B3, B4, C1, C2, C3, C4, D1, D2, D3, E1) identified by letters corresponding to the climatic severity in winter and numbers corresponding to the summer value (Código Técnico de la Edificacion, 2022). Due to data availability limitation, in this study, 13 locations out of 15 were selected to represent 13 climate zones of the Spanish territory according to their elevation from the sea level (Table 19).

Table 19. Location assigned representing thirteen climatic zones of the CTE

Site	Elevation (m)	Climatic zone
Málaga	7	A3
Almeria	21	A4
Murcia	62	B3
Sevilla	31	B4
La Coruna	67	C1
Barcelona	6	C2
Valencia	62	C3
Cáceres	405	C4
Pamplona	453	D1
Salamanca	794	D2
Madrid	582	D3
San Sebastian	259	E1
STA. Cruz de Tenerife	36	α3

The characteristics of the model building adopted for the cooling energy simulation of the study were based on the work conducted by Carpio et al. (Carpio et al., 2015), considering a single housing type with a ground floor (135.70m²) and first floor (88.12 m²). The thermal transmittance limit (U) used fulfilling the requirements of the CTE in its basic document of energy savings (DB-HE1) are: external wall (0.54 W/m^2 K), ground floor (0.65 W/m^2 K), wood door (2.823 W/m^2 K), garage door (3.20 W/m^2 K), and windows (2.47 W/m^2 K). For the structure of roofs, a typical reference pitched and flat roofs for medium weight were considered based on design-builder data, characterized with a thermal insulation of MW stone wool (conductivity of 0.04 W/mK and a thickness of 0.182m) that was set according to the limit permissible thermal transmittance for roofs defined by the CTE DB-HE1, and three scenarios were adopted characterized with different parameters for the roof tiles including conductivity, solar reflectance and emissivity values that are reported in Table 20. The second block of the building was considered for the simulation.

Table 20. Roof tiles characteristics of the three scenarios for the simulation study

Scenarios	Configurations	Solar reflectance	Conductivity (W/mK)	Emissivity
Scenario 0	Flat reference tiles	0.42	0.64	0.76
Scenario 1	Flat tiles with WG1	0.39	0.55	0.83
Scenario 2	Flat tiles with WG2	0.80	0.67	0.76

3. Results

As mentioned before, this work was based on two main stages to reflect the feasibility of using WG coatings for roof tiles: the production phase, as a first stage including the fabrication of roof tiles and recycling of WG, and the second stage, the use phase, mainly affected by the cooling energy savings that this solution can provide. The results of the environmental impact of both phases for the comparison of the three scenarios are reported in this section.

3.1. Production phase of roof tiles

Table 21 and Figure 24 show the contribution of each scenario to the environmental impact of the production phase as it is shown by the result of the life cycle impact assessment. Accordingly, all the impact categories considered for the assessment present the same values for scenarios 1 and 2. In terms of CO_2 emissions, the "Global warming potential" impact category shows a value of 7.79 kg CO_2 eq for scenario 1 and 2, with a slight increase of 0.26% compared to Scenario 0 (the reference). Moreover, the difference in the impact between scenarios 1 and 2 and scenario 0 is very small, with the exception of "Abiotic depletion" (fossil fuels) and "Acidification" impacts that show no difference.

Figure 25 shows that the contribution of P1 is much higher than P2 to the environmental impact for all the impact categories, with a value of 95% or more. Whereas the contribution of

P2 to the environmental impact is slightly highlighted with the impact categories "Fresh Water aquatic ecotox" and "Marine aquatic ecotoxicity" with 1.73% and 4.46% respectively.

Table 21. Life cycle impact assessment results for each scenario

Impact category	Unit	Scenario 0	Scenario 1	Scenario 2
Abiotic depletion	kg Sb eq	3.08E-05	3.09E-05	3.09E-05
Abiotic depletion (fossil fuels)	MJ	108	108	108
Global warming (GWP100a)	kg CO_2 eq	7.77	7.79	7.79
Ozone layer depletion (ODP)	kg CFC-11 eq	9.03E-07	9.04E-07	9.04E-07
Human toxicity	kg 1.4-Db eq	2.31	2.33	2.33
Fresh water aquatic ecotox	kg 1.4-Db eq	1.71	1.73	1.73
Marine aquatic ecotoxicity	kg 1.4-Db eq	3.43E-03	3.59E-03	3.59E-03
Terrestrial ecotoxicity	kg 1.4-Db eq	0.0064	0.0065	0.0065
Photochemical oxidation	kg C2H4 eq	0.0013	0.0013	0.0013
Acidification	kg SO2 eq	0.0183	0.0183	0.0183
Eutrophication	kg PO4--- eq	0.0044	0.0044	0.0044

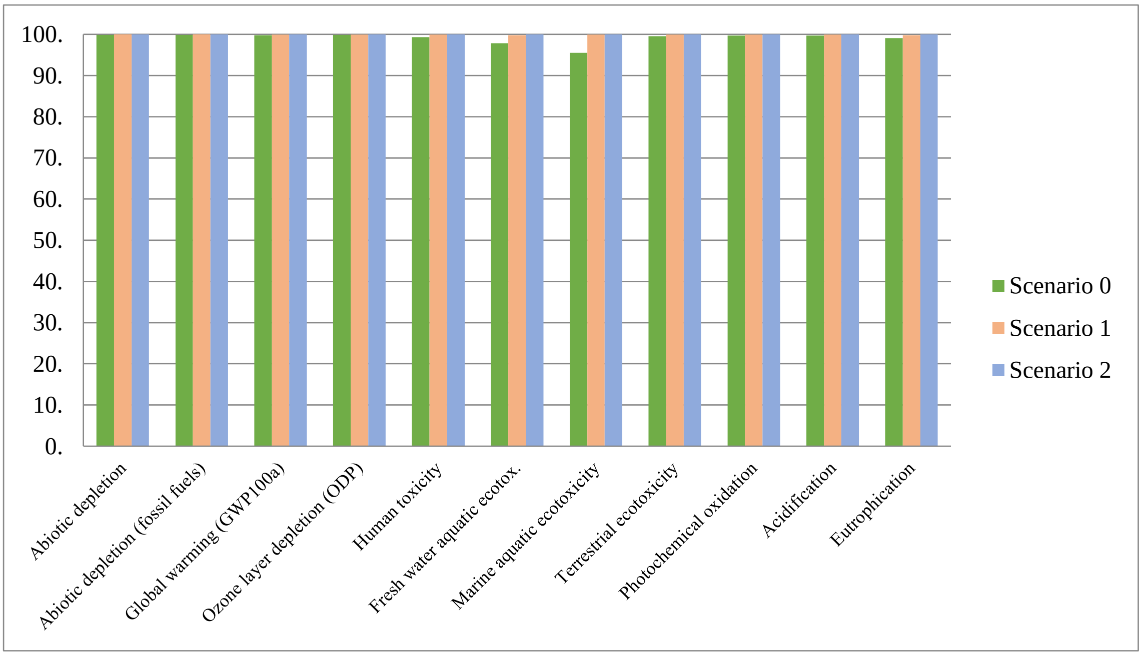


Figure 24. Percentage contribution of each scenario of the production phase of roof tiles for the environmental impact

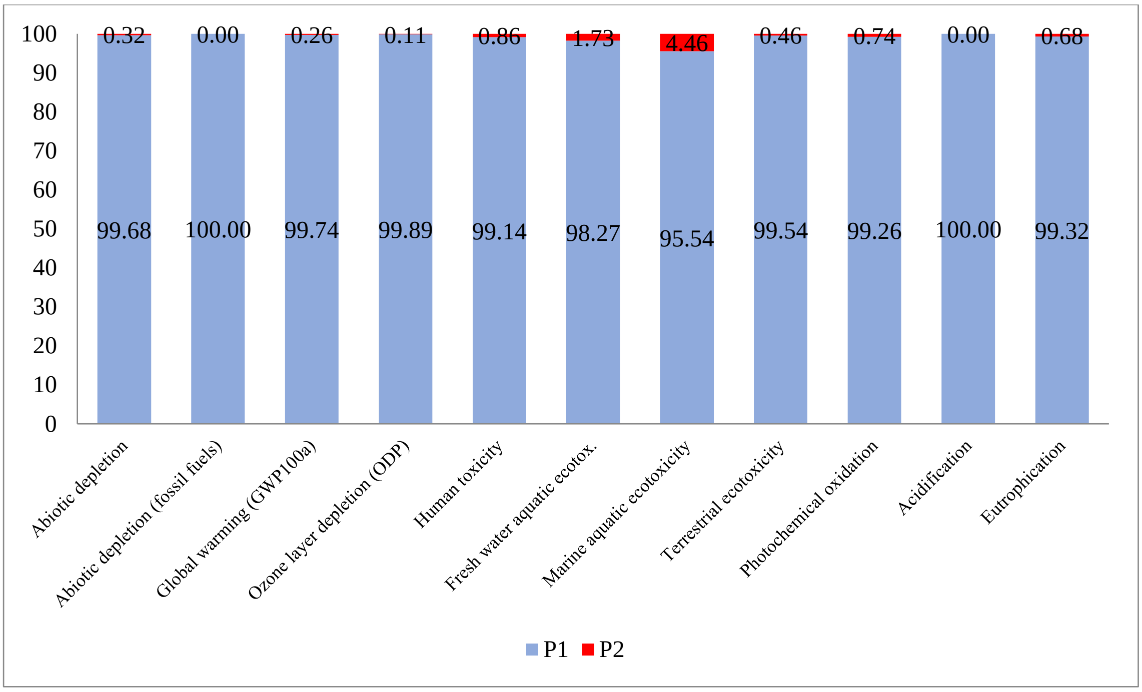


Figure 25. Percentage contribution of the processes of roof tiles fabrication (P1) and recycling WG (P2)

3.2.Use phase of roof tiles

The results of the cooling energy simulation, using Design-builder, are shown in Table 22 and Figure 26. According to Table 22, it is observed that the cooling energy consumption in general is lower for pitched roofs with regard to flat roofs. More specifically, the cooling energy consumption assigned to the climatic zones C1 (0.55kWh and 2.34kWh for pitched and flat roofs respectively in scenario 0) and E1 (0.95kWh and 2.73kWh for pitched and flat roofs respectively in scenario 0) for this study is very low with regard to the other climatic zones. Moreover, scenario 2 provides more cooling energy savings than scenario 1, the climate zones including B4 (285kWh), C3 (205kWh), A4 (154kWh), A3 (138kWh), and C4 (123kWh) show the highest cooling energy savings in the following order from highest to lowest, respectively.

Table 22. Cooling energy simulation per year results of the three scenarios for flat and pitched roof

	Cooling energy consumption (kWh)					
	Flat roof			Pitched roof		
Climatic zone CTE 2022	**Scenario0 (Reference)**	**Scenario1**	**Scenario2**	**Scenario0 (Reference)**	**Scenario1**	**Scenario2**
A3	1278.02	1256.83	1140.32	1193.83	1179.95	1100.29
A4	1646.77	1623.29	1492.34	1557.46	1540.52	1452.91
B3	1073	1057	966	1003	991	933
B4	2215	2156	1930	2076	2044	1906
C1	2.34	1.89	0.64	0.55	0.45	0.09
C2	902.33	879.69	780.48	826.31	813.38	755.35
C3	1726	1686	1521	1611	1589	1489
C4	1224.49	1204.34	1101.82	1162.64	1148.15	1075.98
D1	127	122	92	99	95	76
D2	208	199	156	170	165	138
D3	1210	1169	1015	1194	1180	1100
E1	2.7	2.4	1.1	0.9	0.8	0.3
α3	1046	1025	914	967	953	880

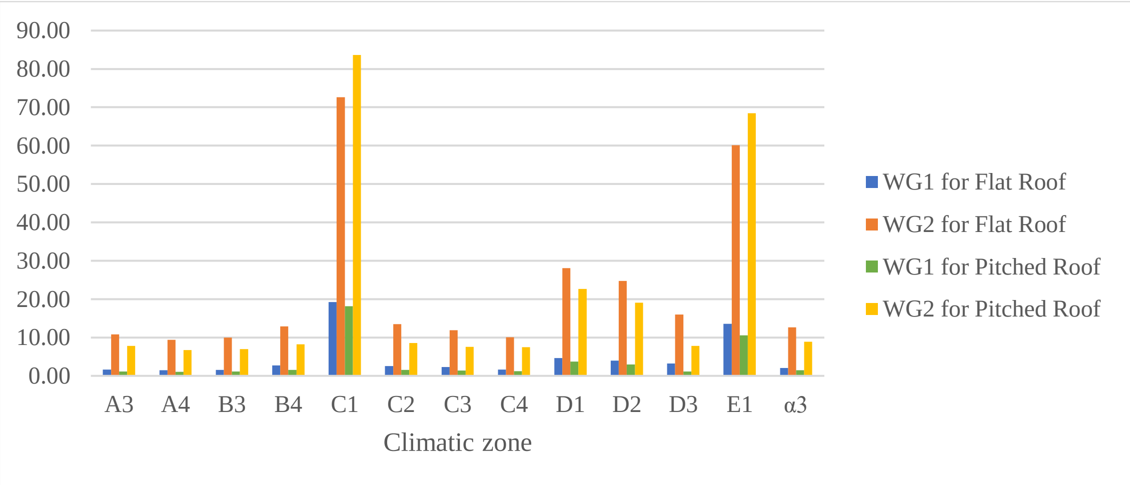


Figure 26. The percentages of savings in cooling energy for scenarios 1 and 2 in flat and pitched roofs for different climatic zones

Figure 26 shows the different percentages of cooling energy savings for scenarios 1 and 2 with regard to the reference. For the thirteen climate zones considered, scenario 1 provided law cooling energy savings percentages that are varying from 1.09% (A4) to 3.71% (D1) for pitched roof and 1.43% (A4) to 4.61% (D1) for flat roof, whereas scenario 2 showed the highest cooling energy savings percentages varying from 6.71% (A4) to 22.63% (D1) for pitched roof and 9.38% (A4) to 28.06% (D1) for flat roof. It is observed that the lowest the cooling energy consumption is, the highest is the percentage of savings using scenario 1 and 2 which is not feasible for the total energy loads of the buildings. It is noteworthy to mention that the cases of

C1 and E1 are not included in this comparison for the cooling energy savings percentages which will be further explained in the discussion section.

4. Discussions

The production phase highlighted the effect of using WG coatings on the overall environmental impact through a comparison between the three scenarios. The impact categories demonstrate that the processing of recycled WG, regardless of the type of WG used in scenarios 1 and 2, shows a very low environmental impact compared to the roof tiles fabrication process (Figure 25). The contribution of the impact categories "Fresh Water aquatic ecotox" and "Marine aquatic ecotoxicity" was slightly higher in scenarios 1 and 2 compared to the other impact categories due to emissions related to the recycling process of WG.

Regarding the CO_2 eq emissions generated during the production phase, the "Global warming potential" impact category shows a low increase of 0.26% for scenarios 1 and 2 compared to scenario 0. This demonstrates that recycling WG process generates a low impact on greenhouse gas (GHG) emissions. Compared to commercial glazes fabrication of roof tiles, this option eliminates the energy consumption related to frit production, which is considered the most impacting process as stated by S. Mohaddes Khorassani et al. (Mohaddes Khorassani et al., 2020). Moreover, the application of WG coatings for roof tiles is part of the single firing process of the tiles, inducing less energy consumption (Zanatta et al., 2021). The findings obtained in this phase are attributed to the difference in embodied energy of each process, where the roof tiles fabrication process requires significantly higher energy compared to the recycling of WG process. In other words, the recycling of WG process in scenarios 1 and 2 has a minor effect on the overall process of roof tiles production that is known as an energy intensive sector (Quinteiro et al., 2022), which primarily relies on high energy consumption in kilns, dryers, and the processing of raw materials. These factors subsequently contribute to most of the fuel-related emissions which aligns with Gomes et al. (Gomes et al., 2019) findings, who reported that the processing of materials, fabrication processes, and extraction contribute to 73% or more for the most of the environmental impact categories.

For the cooling energy simulation, the results reported in Table 22 show that the case of pitched roof in general registers lower cooling energy consumption, which could be explained by the structure of the roof that includes the attic and contributes to a natural ventilation. Moreover, in terms of the amount of solar heat gain that influences the cooling energy consumption, compared to pitched roofs, flat roofs have a larger surface area exposed to the sun, according to Sumol et al. (Pisitsungkakarn & Jirakulsomchok, 2023), less energy consumption was obtained for roofs installed at an angle of 45 compared to those at an angle of 15. On the other hand, climate zones C1 and E1 exhibit the lowest cooling energy consumption less than 1% of the total cooling energy consumption assigned to the climate zone A4. Which is explained by the high severity of winter and milder summer conditions of these areas that do not require improving the cooling measures. These results indicate that these locations are not presenting a relevant element in the comparison to highlight the contribution of WG coatings in the cooling energy savings. In other terms, scenarios 1 and 2 are not feasible for these areas and the application of these scenarios will increase the heating loads in a way to impact the total loads savings.

In this study, four major parameters were considered for the assessment of cooling energy savings including climate zones, reflectance, conductivity, and emissivity of the roof tiles used. According to the results, scenario 2 registered the highest cooling energy savings, which is considered as a feasible solution for flat and pitched roofs that is characterized by the highest solar reflectance of 0.8 (Table 19). This result demonstrates the contribution of solar reflectance of WG2 coatings to the cooling energy savings, moreover, in the case of scenario 1 that is characterized with a solar reflectance lower than scenario 0, the overall impact on the cooling energy was affected by the emissivity (Table 19), which is considered as a significant parameter in the thermal radiation characteristics of a material thus in the buildings' energy saving and insulation (Guo et al., 2019; Tang et al., 2019).

The climate zones including B4, C3, A4, A3 and C4 were characterized by the highest values of cooling energy savings with scenario 2 in the case of flat roofs, with the energy saving percentages 13%, 12%, 9%, 11% and 10% respectively. These climate zones are characterized by severe summer conditions based on the Spanish CTE classification. This result demonstrates that scenario 2 is considered as a sustainable solution applicable in these locations for flat and pitched roofs, with better feasibility for flat roofs. However, this selection could be refined to the climate zones including B4, C3, and A4, where the applicability of scenario 2 allows the decrease of the annual loads without having a great impact on the heating ones. In this context, using WG2 coatings represented by scenario 2 in this study, can be considered as a sustainable alternative for commercial cool roof materials, such as cool paints that was demonstrated by Antonio et al. (Dominguez-Delgado et al., 2020), to decrease the annual total loads by 32% for a solar absorptivity equal to 0.1 in southern Spain, these results are in accordance with the case of B4 and A4 assigned to Sevilla and Almeria that provides cooling energy savings of 13% and 9% respectively. Moreover, it was found that significant sensible cooling energy savings up to 42%, were obtained using cool roof materials in the Mediterranean area (De Carli, 2007), which proves that cool coatings are dependent on the locale climate conditions. The annual thermal loads savings can be improved as well by finding the optimal combination between reflectivity and thermal insulation (Pisitsungkakarn & Jirakulsomchok, 2023; Yuan et al., 2016), in this work, the thermal insulation was set according to the limit permissible value defined by the Spanish CTE, using MW stone wool.

The environmental impact assessment of the three scenarios considered, show that the results are complementary which highlight the benefits of using WG2 coatings for cool roof applications as it was expected. The embodied energy represented by the production phase in the recycling process of WG has a low environmental impact with regard to the entire production process of tiles. At the same time, the operational energy represented by the use phase demonstrates the advantages of implementing roof tiles with WG2 coatings in terms of cooling energy savings, and the feasibility of implementing this solution in hot climate regions characterized with severe summer conditions where cooling load dominates most of the year (Arunraj et al., 2020; Kolokotroni et al., 2018).

Analyzing the results of the LCA in this study, several factors contribute to the sustainability of this ecofriendly solution, including the use of recycled raw materials for the fabrication of reflective coatings which decrease the primary raw materials consumptions. In fact, less environmental loads occur related to virgin raw materials extraction and processing. Another factor is the replacement of commercial reflective glazes that consume huge amounts of energy for the fabrication process. Moreover, using recycled WG induces less landfill disposal, which complies to the circular economy and waste management strategies recommended by the European green deal. In addition to the decrease in cooling loads that alleviates the pressure from consuming energy and consequently lower the concentration of GHGs in the atmosphere. The potential reduction of CO_2 emissions was estimated using a Coefficient of Performance (CoP) of 1.32 for the air conditioning system, for Spain the electricity CO_2 emission factor was set as 0.343 $kgCO_2/kWh$ (Brander et al., 2011). Hence for the case of the climate zone B4 using scenario 2 for flat tiles, a saving approximation was estimated up to 74.05 $kgCO_2$. These results emphasize the importance of the use phase in this

LCA assessment study which presents a compromise between CO_2 emissions in the production phase and the use phase associated with using WG coatings for roof tiles.

5. Conclusion

The environmental impact analysis of using WG coatings for roof tiles was conducted through two main complementary stages, production phase using Simapro software for the environmental impact assessment according to a life cycle perspective and use phase utilizing Design-Builder software for the cooling energy simulation. The following findings have been allowed.

For the analysis of roof tiles using WG, the "Global warming potential" impact was slightly increased by 0.26%. However, the recycling process of WG coatings has a small contribution to the overall environmental impact of the LCA with less than 5% for all impact categories regardless of the WG type considered, with regard to the conventional production process of the roof tiles without WG. In the other hand, the implementation of scenario 2 using WG2 coatings proved to be a viable solution used to decrease the cooling energy consumption in buildings due to the solar reflectance performance, indicating the highest cooling energy savings for up to 285 kWh/year and 170 kWh/year for flat and pitched roofs respectively. Moreover, the cooling energy savings were proved to be dependent on the climate zones, in that sense, it is recommended the implementation of scenario 2 in hot climate regions characterized by severe summer conditions where cooling loads dominates most of the year for better efficacity to avoid the increase of the annual loads. In this study, the climate zones B4, C3, and A4 represent the optimal cases for this application with flat roofs, which allows savings percentages of 13%, 12% and 9% respectively.

This study highlighted the significant role of the use phase in the assessment of the environmental impact of this solution, which allows to demonstrate the compensation for the potential environmental burdens resulted from the production phase by the energy savings in the operational phase. These findings permit to consider scenario 2 as a sustainable solution for cool roofs applications which has a twofold potential in saving raw materials using recycled glass wastes and decreasing cooling energy consumption.

This study provides a reference and a starting point for future investigations to analyse the environmental impact of using WG coatings for roof tiles from various perspectives. It is recommended to enlarge the system boundaries including as much as possible relevant steps from cradle to grave to broaden the research results for the application of this solution.

CONCLUSIONES

Siguiendo el enfoque metodológico descrito previamente en este estudio, se considera que los objetivos de esta investigación se lograron a través de las diferentes etapas de este trabajo. Las principales aportaciones obtenidas se resumen en los cuatro apartados siguientes que representan los objetivos formulados de esta investigación:

(i) Analizar la evolución científica de las estrategias de superficies frías.
(ii) Realizar un estudio preliminar para la caracterización del comportamiento de reflectancia espectral de muestras de baldosas con recubrimientos de RV.
(iii) Fabricar tejas cerámicas a escala real utilizando recubrimientos de RV para realizar una caracterización física, mecánica y óptica.
(iv) Analizar el impacto ambiental de las fases de producción y uso de las tejas cerámicas con recubrimientos de RV mediante un análisis del ciclo de vida.

A continuación, se detallan las principales conclusiones y aportaciones en relación con los 4 objetivos anteriores.

En cuanto a la primera aportación, es decir, al análisis de la evolución científica de las estrategias de superficies frías, se realizó un estudio bibliométrico basado en mapeo científico y análisis de desempeño que permitió establecer una revisión de las estrategias y materiales aplicados en superficies frías. Los resultados obtenidos al lograr el objetivo inicial mostraron que se trata de un campo que está en constante evolución siguiendo las cambiantes especificaciones de las edificaciones, lo que valida con la hipótesis planteada para esta investigación. Los resultados de esta investigación muestran que :

- El campo de las superficies frías ha ido ganando atención a lo largo de los años y el período más prolífico comenzó en 2011, representando el 88% de los registros generales de esta investigación.
- Las estrategias de superficies frías evolucionan continuamente, con el objetivo de producir materiales sostenibles que se alineen con los requisitos técnicos de

los edificios nuevos y existentes, y para diversas aplicaciones, incluidos pavimentos, fachadas y, en particular, tejados.

- Los temas más destacados en esta investigación para aplicaciones de superficies frías son los pigmentos, los materiales retror reflectantes, los materiales de cambio de fase, los materiales cerámicos y el vidrio.
- La aplicación de pigmentos diseñados, materiales retror reflectantes, materiales de cambio de fase y materiales cerámicos puede producir un rendimiento de reflectancia de la radiación solar de hasta el 95%; sin embargo, el proceso de producción puede inducir cargas ambientales; además, estas estrategias presentan algunas limitaciones de rendimiento.
- Los RV mostraron un potencial prometedor como materia prima secundaria utilizada con fines de reflectancia solar.

En cuanto el Segundo objetivo, el estudio preliminar para la caracterización del comportamiento de reflectancia espectral de probetas de baldosas con recubrimientos de RV, se realizó a través de la fabricación de las probetas y la realización de mediciones ópticas que permitieron evaluar la influencia de diferentes parámetros sobre las probetas. La hipótesis inicial planteaba valorar el impacto de varios parámetros, pero no todos los parámetros considerados impactaron el desempeño de los especímenes en términos de las características ópticas y apariencia visual, lo que permitió las siguientes conclusiones:

- Se obtuvo una mejor densificación como se desprende del aspecto visual de las probetas, utilizando una temperatura de cocción de 1000ºC y un tiempo de mantenimiento de 60min, lo que proporcionó una mejor adherencia entre los recubrimientos de RV y el sustrato.
- El recubrimiento de RV de silicato de plomo registró el valor medio más alto del grado de luminosidad y de la reflectancia espectral en comparación con el RV de sílice soda cálcica.
- El tiempo de permanencia y el espesor de los recubrimientos de RV no tienen una influencia significativa sobre el grado de luminosidad y la reflectancia espectral de las muestras. Sin embargo, se observó una excepción en el RV de

silicato de plomo, donde la reflectancia espectral se mejoró con un tiempo de retención de 60 minutos.

En referencia al tercer objetivo de la investigación, consistente en la fabricación de tejas cerámicas a escala real utilizando recubrimientos de RV y su caracterización física, mecánica y óptica, el objetivo se logró mediante la fabricación de tejas planas y curvas y la realización de las correspondientes pruebas de laboratorio, lo que permitió evaluar la influencia de la forma de las tejas y el tipo de RV. Los resultados obtenidos en esta etapa de la investigación estuvieron alineados con la hipótesis inicial, y se resumen a continuación:

- La aplicación de RV reciclado para revestimientos de tejas se alinea con los estándares y no tiene ningún impacto adverso en el desempeño técnico de las tejas.
- Los recubrimientos de RV mejoraron la resistencia a la flexión de las tejas, especialmente con el uso de recubrimientos de RV de silicato de plomo que la aumentaron hasta un 6,06% y un 16,37%, para las tejas planas y curvas, respectivamente.
- Los recubrimientos de RV mostraron buenas características físicas que permiten el uso de las tejas en climas húmedos, además, la posibilidad de lixiviación del plomo contenido en el RV de silicato de plomo resultó nula.
- Según las características colorimétricas, el color del soporte y la estructura superficial influyen en la luminosidad de las baldosas. La aplicación de RV de silicato de plomo incrementó la luminosidad hasta 41%.
- La forma de las tejas influye fuertemente en el comportamiento de la reflectancia solar. Las baldosas planas reflejan más radiación que las curvas para las mediciones normales de reflectancia solar. Además, la aplicación de RV de silicato de plomo mejora la reflectancia solar normal de las tejas a lo largo del rango del espectro de 350-1100 nm, con un aumento del 90% y 36%, para tejas planas y curvas respectivamente, con respecto a la referencia.
- La aplicación del tipo de RV de silicato de plomo para tejas planas se considera la opción óptima en este estudio para la reflectancia solar normal de la teja, lo que proporciona una alternativa sustentable a los materiales reflectantes

comerciales en el mercado para aplicaciones de techos frescos, y consecuentemente contribuye a la mitigación de Efecto ICU en las ciudades.

Finalmente, para el último objetivo ligado al análisis del impacto ambiental de las fases de producción y uso de las tejas cerámicas con recubrimientos de RV mediante un ACV, este objetivo se logró mediante la recolección y el procesamiento de datos relacionados con la fabricación de tejas y la simulación. del consumo de energía de refrigeración. Los resultados obtenidos confirmaron las hipótesis planteadas para esta investigación, y permitieron obtener las siguientes conclusiones:

- Para el análisis de tejas utilizando RV, el impacto del "Potencial de calentamiento global" se incrementó ligeramente en un 0,26%. Sin embargo, el proceso de reciclaje de los recubrimientos de RV tiene una pequeña contribución al impacto ambiental general del ACV con menos del 5 % para todas las categorías de impacto, independientemente del tipo de RV considerado.
- La implementación de tejas con recubrimientos de RV de silicato de plomo demostró ser una solución viable utilizada para disminuir el consumo de energía de refrigeración en los edificios debido al rendimiento de la reflectancia solar, lo que indica el mayor ahorro de energía de refrigeración de hasta 285 kWh/año y 170 kWh/año. para tejados planos e inclinados respectivamente.
- Los ahorros de energía de refrigeración dependen de las zonas climáticas, para las cuales se recomienda la implementación de recubrimientos de RV de silicato de plomo en regiones de clima cálido caracterizadas por condiciones severas de verano donde las cargas de refrigeración dominan la mayor parte del año, para una mejor eficacia y evitar el aumento de la cargas anuales. Las zonas climáticas B4, C3 y A4 representan los casos óptimos para esta aplicación con cubiertas planas, que permite porcentajes de ahorro del 13%, 12% y 9% respectivamente.
- El estudio ACV destacó el importante papel de la fase de uso en la evaluación del impacto ambiental, que permite demostrar la compensación de las posibles cargas ambientales resultantes de la fase de producción mediante el ahorro de energía en la fase operativa.

Los resultados destacados en esta investigación demuestran que las tejas con recubrimientos de RV de silicato de plomo en particular se consideran una solución sostenible para aplicaciones de techos frescos que tiene un doble potencial en el ahorro de materias primas, tanto por el uso de desechos de vidrio reciclado como por la disminución del consumo de energía de refrigeración. Esta aplicación contribuye a la cadena dirigida a la mitigación del fenómeno del efecto ICU. Además, los hallazgos mencionados anteriormente contribuyen al conjunto de conocimiento existente en el campo de las superficies frías, presentando una referencia teórica para la evolución científica de este campo de investigación. En definitiva, se considera que la presente Tesis Doctoral presenta una evaluación práctica de una primera prueba de concepto que muestra el rendimiento de la reflectividad solar de tejas con revestimientos de vidrio reciclado, que permite mayores profundizaciones y desarrollos futuros, como se recogen en el siguiente apartado.

CONCLUSIONS

Following the methodological approach described previously in this study, the objectives of this research were achieved through the different stages of this work. The major contributions obtained are outlined in the four following sections representing the objectives of this research:

(i) To analyse the scientific evolution of cool surfaces strategies.

(ii) To conduct a preliminary study for the characterisation of the spectral reflectance performance of tiles specimens with WG coatings.

(iii) To fabricate ceramic roof tiles at real scale using WG coatings in order to perform a physical, mechanical, and optical characterisation.

(iv) To analyse the environmental impact of the production and the use phases of ceramic roof tiles with WG coatings through a life cycle assessment.

Concerning the first section, the scientific evolution analysis of cool surfaces strategies, the bibliometric study based on science mapping and performance analysis was conducted which allowed the establishment of a review for the strategies and materials applied in cool surfaces. The results obtained from achieving the initial objective showed a field that is constantly evolving following the changing specifications of buildings, which aligns with the hypothesis set for this investigation. For this part of the research:

- The findings indicated that the cool surfaces field has been gaining attention over the years with the most prolific period starting from 2011, accounting for 88% of the overall records in this research.
- Cool surfaces strategies are continuously evolving, with the objective of producing sustainable materials that align with the technical requirements of new and existing buildings, and for various applications including pavements, facades, and notably roofs.
- The most highlighted themes in this research for cool surfaces applications are pigments, RR materials, PCMs, ceramic materials, and glass.
- The application of designed pigments, RR materials, PCMs and ceramic materials can yield the solar radiation reflectance performance up to 95%, however the production process may induce environmental burdens, moreover, these strategies present some performance limitations.

- WG showed a promising potential as a secondary raw material used for solar reflectance purposes.

Regarding second section representing the preliminary study for the characterisation of the spectral reflectance performance of tiles specimens with WG coatings, the objective was conducted through the manufacturing of the specimens and the performance of the optical measurements which allowed the evaluation of the influence of different parameters on the specimens. Contrary to the initial hypothesis, not all the parameters considered impacted the performance of the specimens in terms of the optical characteristics and visual appearance, which allowed the following conclusions:

- Better densification was obtained as evident from the visual appearance of the specimens, using a firing temperature of 1000ºC and a holding time of 60min, which provided better adherence between the WG coatings and the substrate.
- Coating with lead silicate (LS) WG registered the highest mean value of the lightness degree and the spectral reflectance compared to the soda lime silica (SLS) WG.
- The holding time and thickness of WG coatings have no significant influence on the lightness degree and spectral reflectance of the specimens. However, an exception was noted for the LS WG, where the spectral reflectance was enhanced with a holding time of 60 minutes.

For the third section , which is the fabrication of ceramic roof tiles at real scale using WG coatings for the performance of a physical, mechanical, and optical characterisation, this objective was accomplished through fabricating flat and curved roof tiles and performing laboratory tests which allowed the evaluation of the influence of the tiles shape and WG type. The results obtained at this stage of the research were aligned with the initial hypothesis, and are outlined in the following:

- The application of recycled WG for roof tiles coatings aligns with the standards and have no adverse impact on the technical performance of roof tiles.
- WG coatings enhanced the flexural strength of roof tiles, notably with the use of LS WG coatings that increased up to 6.06% and 16.37%, for flat and curved tiles respectively.

- WG coatings showed good physical characteristics which allow the use of the tiles in wet climates, moreover, the possibility of lead leaching contained in the LS 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 LS WG increased the lightness by 41%.
- The shape of tiles strongly influences the solar reflectance behavior; flat tiles reflect more radiation than curved tiles for the normal solar reflectance measurements. Besides, the application of LS WG enhances the normal solar reflectance of the tiles along the spectrum range of 350-1100nm, with an increase of 90% and 36%, for flat and curved tiles respectively, with regard to the reference.
- The application of LS WG type for flat roof tiles is considered the optimal option in this study for the tile’s normal solar reflectance, which provides a sustainable alternative to commercial reflective materials in the market for cool roofs applications, and subsequently contributes to the mitigation of UHI effect in cities.

Finally, for the last section which is the analysis of the environmental impact of the production and the use phases of the ceramic roof tiles with WG coatings through an LCA, this objective was achieved through the collection and the processing of data related to the fabrication of tiles, and the simulation of the cooling energy consumption. The results obtained were confirmed the hypothesis set for this investigation, and allowed the following conclusions:

- For the analysis of roof tiles using WG, the "Global warming potential" impact was slightly increased by 0.26%. However, the recycling process of WG coatings has a small contribution to the overall environmental impact of the LCA with less than 5% for all impact categories regardless of the WG type considered.
- The implementation of roof tiles with LS WG coatings proved to be a viable solution used to decrease the cooling energy consumption in buildings due to the solar reflectance performance, indicating the highest cooling energy savings for up to 285 kWh/year and 170 kWh/year for flat and pitched roofs respectively.
- The cooling energy savings are dependent on the climate zones, for which it is recommended the implementation of LS WG coatings in hot climate regions

characterized by severe summer conditions where cooling loads dominates most of the year, for better efficacity to avoid the increase of the annual loads. The climate zones B4, C3, and A4 represent the optimal cases for this application with flat roofs, that allows savings percentages of 13%, 12% and 9% respectively.

- The LCA study highlighted the significant role of the use phase in the assessment of the environmental impact, which allows to demonstrate the compensation for the potential environmental burdens resulted from the production phase by the energy savings in the operational phase.

The findings highlighted in this research demonstrate that roof tiles with LS WG coatings in particular are considered a sustainable solution for cool roofs applications, which has a twofold potential in saving raw materials using recycled glass wastes and decreasing cooling energy consumption. This application contributes to the chain addressed for the mitigation of the UHI effect phenomenon. Moreover, the findings stated above contribute to the existing body of knowledge in the cool surfaces field, presenting a theoretical reference for the scientific evolution of this research field. Finally, it is concluded that this Doctoral thesis presents a practical evaluation of a first proof of concept testing solar reflectivity performance of roof tiles with WG coatings.

LÍNEAS FUTURAS DE INVESTIGACIÓN

A partir del trabajo desarrollado en esta investigación se han observado ciertos aspectos que requieren un análisis más detallado, y se han identificado nuevos campos interesantes de desarrollo que se consideran susceptibles de futuros progresos. En consecuencia, a continuación, se proponen las siguientes futuras líneas de investigación:

- La caracterización y evaluación del comportamiento de la reflectancia solar de tejas que incorporan RV en el propio cuerpo del sustrato.

- La consideración del factor de impacto del envejecimiento en la evaluación de la reflectancia solar de tejas con recubrimientos de RV y su durabilidad.

- La evaluación de la influencia de la forma de la baldosa sobre la reflectancia solar considerando diferentes ángulos de incidencia de la radiación y utilizando detectores hemisféricos.

- Valorar y ampliar la caracterización de las tejas con recubrimientos de RV ensayando diferentes tipos de RV y considerando una fabricación industrial de las tejas, mejorando así la información sobre la caracterización de las mismas.

- Para el ACV, sería muy interesante ampliar los límites del sistema incluyendo más datos primarios para obtener resultados completos sobre el impacto ambiental de la producción y el uso de tejas con recubrimientos de RV. Además, se podrían abordar otros aspectos complementarios como los impactos económicos y sociales de esta solución.

FUTURE LINES OF RESEARCH

From the work developed in this research, certain aspects have been observed that require more detailed analysis. Consequently, the subsequent future lines of research are proposed below:

- The characterisation and the evaluation of the solar reflectance behavior of roof tiles incorporating WG in the substrate body.

- The consideration of the aging impact factor on the solar reflectance evaluation of roof tiles with WG coatings and their durability.

- The evaluation of the influence of the tile's shape on the solar reflectance considering different angles of radiation incidence and using hemispherical detectors.

- Upgrade the characterisation of roof tiles with WG coatings by testing different types of WG and considering an industrial manufacturing of the roof tiles.

- For the LCA analysis, it is recommended to broaden the system boundaries including more primary data for comprehensive results regarding the environmental impact of the production and the use of roof tiles with WG coatings. Other complementary aspects could be approached such as the economic and social impacts of this solution.

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ANNEX A

Statistical Analysis for the lightness degree and spectral reflectance

Table A1. ANOVA for the lightness of the specimens.

Factor	Model	Sum of Squares	df	Mean Square	F	Sig.
WG type	Linear	6201.564	2	3100.782	28.708	0.000
holding time	Linear	15.205	2	7.603	0.021	0.979
thickness	Linear	729.768	2	364.884	1.086	0.354

Table A2. Mann–Whitney U test for specular solar reflectance, according to the holding time of 20 min.

	Reflectance		
	WG1T20_WG2T20	WG1T20_WG3T20	WG2T20_WG3T20
Mann–Whitney U	137.786500	0.000	0.000
Asymp. Sig. (2-tailed)	0.000	0.000	0.000

Table A3. Mann–Whitney U test for specular solar reflectance according to the holding time of 40 min.

	Reflectance		
	WG1T40_WG2T40	WG1T40_WG3T40	WG2T40_WG3T40
Mann–Whitney U	146.304000	0.000	0.000
Asymp. Sig. (2-tailed)	0.000	0.000	0.000

Table A4. Mann–Whitney test for specular solar reflectance, according to the holding time of 60 min.

	Reflectance		
	WG1T60_WG2T60	WG1T60_WG3T60	WG2T60_WG3T60
Mann–Whitney U	150.144500	0.000	0.000
Asymp. Sig. (2-tailed)	0.000	0.000	0.000

Table A5. Mann–Whitney U test for specular solar reflectance of WG1 during the three holding times.

	Reflectance		
	WG1T20_WG1T40	WG1T20_WG1T60	WG1T40_WG1T60
Mann–Whitney U	172.056000	9385.000	7424.000
Asymp. Sig. (2-tailed)	0.046	0.000	0.000

Table A6. Mann–Whitney U test for specular solar reflectance of WG2 during the three holding times.

	Reflectance		
	WG2T20_WG2T40	WG2T20_WG2T60	WG2T40_WG2T60
Mann–Whitney U	116.399500	4878.500	13.262500
Asymp. Sig. (2-tailed)	0.000	0.000	0.000

Table A7. Mann–Whitney U test for specular solar reflectance of WG3 during the three holding times.

	Reflectance		
	WG3T20_WG3T40	WG3T20_WG3T60	WG3T40_WG3T60
Mann–Whitney U	2599.000	18.817000	0.000
Asymp. Sig. (2-tailed)	0.000	0.000	0.000

Table A8. Mann–Whitney U test for specular solar reflectance of WG1 with the three thickness values.

	Reflectance		
	WG1Q1_WG1Q2	WG1Q1_WG1Q3	WG1Q2_WG1Q3
Mann–Whitney U	19.000	23.000	19.000
Asymp. Sig. (2-tailed)	0.482	0.848	0.482

Table A9. Mann–Whitney U test for specular solar reflectance of WG2 with the three thickness values.

	Reflectance		
	WG2Q1_WG2Q2	WG2Q1_WG2Q3	WG2Q2_WG2Q3
Mann–Whitney U	20.000	17.000	15.000
Asymp. Sig. (2-tailed)	0.565	0.568	0.391

Table A10. Mann–Whitney U test for specular solar reflectance of WG3 with the three thickness values.

	Reflectance		
	WG1Q1_WG1Q2	WG1Q1_WG1Q3	WG1Q2_WG1Q3
Mann–Whitney U	18.000	17.000	20.000
Asymp. Sig. (2-tailed)	0.406	0.568	0.886

Emissivity test results

The atmospheric temperature is 22°C, the reflected apparent temperature is 22°C, and the black body emissivity is taken as 0.95 (spot 2 in the Figure A1 below).
The mean emissivity values of FRef, FWG1 and FWG2 are respectively 0.76, 0.83, and 0.76.

Table A11. Emissivity values of the ten specimens in two different spots with regard to the black body emissivity.

Tile	FRef					FWG1					FWG3				
	Temperature °C			Emissivity		Temperature °C			Emissivity		Temperature °C			Emissivity	
	Spot 1	Spot 2	Spot 3	Spot 1	Spot 3	Spot 1	Spot 2	Spot 3	Spot 1	Spot 3	Spot 1	Spot 2	Spot 3	Spot 1	Spot 3
1	35,6	37,2	35,4	0,76	0,74	36,9	37,2	36,1	0,84	0,81	35,6	37,2	35,2	0,77	0,75
2	35,7	37,4	36,7	0,74	0,77	35,8	37,3	37,6	0,78	0,87	35,8	37,4	35,7	0,79	0,76
3	34,7	37,2	35,7	0,74	0,76	35,5	37,2	35,7	0,73	0,75	34	37,1	34,5	0,71	0,73
4	35,3	37,5	35,6	0,75	0,74	35,6	36,5	36,5	0,82	0,84	34,4	37,6	34,6	0,72	0,74
5	34,9	37,1	36	0,75	0,78	36	37,1	35,5	0,83	0,8	36,5	37	35,5	0,78	0,76
6	35,6	37,1	35,8	0,77	0,77	35,7	36,7	36,7	0,83	0,87	35,2	37,3	35,4	0,77	0,78
7	35,7	37,3	35,5	0,77	0,76	35,8	37,4	36,7	0,82	0,85	35,5	37,2	36,2	0,79	0,81
8	35,5	37,4	36,7	0,77	0,78	36,9	37,1	36	0,88	0,84	35,4	37,6	34,5	0,78	0,73
9	35	37,3	35,6	0,76	0,77	36,7	37,4	36,9	0,84	0,86	36,2	37,4	35,9	0,77	0,76
10	35,3	37,2	35,5	0,73	0,74	36	37,4	36,6	0,82	0,85	35,7	37,1	34,6	0,74	0,72

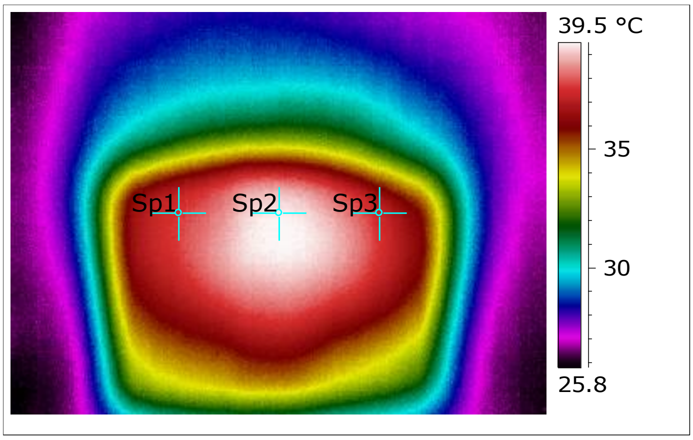

Figure A1. thermal image of the tile pointing the three different spots