

Research trends on thermochromic asphalt mixtures functionalization: Bibliometric analysis and review

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ARTICLE INFO

Keywords:

Cool pavements
Thermochromic asphalt mixtures
Thermal properties
Smart asphalt mixtures
Smart materials
Urban heat island

ABSTRACT

Asphalt mixtures absorb high levels of solar light, increasing their internal temperatures and negatively affecting their behavior. In urban areas, this leads to the urban heat island (UHI) phenomenon, where extensive asphalt coverage raises microclimate temperatures. Integrating thermochromic properties into asphalt could provide a sustainable solution to mitigate UHI while maintaining the necessary mechanical performance. This research reviews the literature, scientific advances, gaps, and future perspectives on thermochromic (TCM) asphalt mixtures through bibliometric analysis and systematic review. The analysis highlights that thermochromic asphalt has recently gained significant scientific interest, with an increasing number of publications from 2013 to 2023. Notably, Hunan University in China has emerged as a leading contributor to thermochromic-related publications. The findings identify the wet method as a prevalent form of thermochromic functionalization, with 4–6% TCM content considered most suitable for thermal performance. Adding TiO₂ improves optical characteristics due to its high reflectance in the Near-infrared solar spectrum. Studies using TCM binders typically show asphalt mixtures with better resistance to rutting and cracking, likely due to enhanced thermoregulation. The key knowledge gaps identified include the lack of consistent procedures across studies, the feasibility of scaling lab-based methods to field applications, and the need for experiments to assess their impact on road safety, surface characteristics, and the durability of thermochromic properties over the pavement's service life. This review highlights the promise of TCM for urban heat management and emphasizes the importance of additional research to achieve the ideal balance between thermal and mechanical properties.

1. Introduction

Asphalt mixtures are commonly used in road pavements, covering a large impermeable area of urban surfaces. They absorb a significant amount of solar radiation, which poses environmental, social, and economic challenges (Golden and Kaloush, 2006; Rosenfeld et al., 1996). This fact exacerbates the formation of urban heat islands (UHIs), among other adverse effects. UHI is a phenomenon where urban areas experience higher temperatures compared to surrounding and rural areas. This is mainly caused by the replacement of natural land with built surfaces. In this context, pavements with low reflectance and high heat retention may alter the local microclimate, affecting environmental conditions, human health, and the demand for electricity and water consumption to maintain thermal comfort.

Asphalt materials are sensitive to temperature changes, which affect their mechanical behavior and performance. During summer, asphalt mixtures absorb high amounts of solar radiation due to their black color, reaching temperatures up to 70 °C (Qin and Hiller, 2014). In Transportation Engineering, efforts have been made to develop sustainable materials, solutions, methods, and construction processes to low-down in-service mixture temperature, as this directly impacts their durability and accelerates deterioration. An example of such technology is the development of thermochromic asphalt mixtures (Segundo et al., 2021), which has been used to mitigate UHI effects.

The use of thermochromic materials in asphalt mixtures enhances their reflectance by undergoing reversible color changes from colored to colorless states in response to temperature fluctuations. These color changes occur at a specific transition temperature (TT), below which the

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<https://doi.org/10.1016/j.clema.2024.100273>

Received 29 June 2024; Received in revised form 8 October 2024; Accepted 8 October 2024

Available online 10 October 2024

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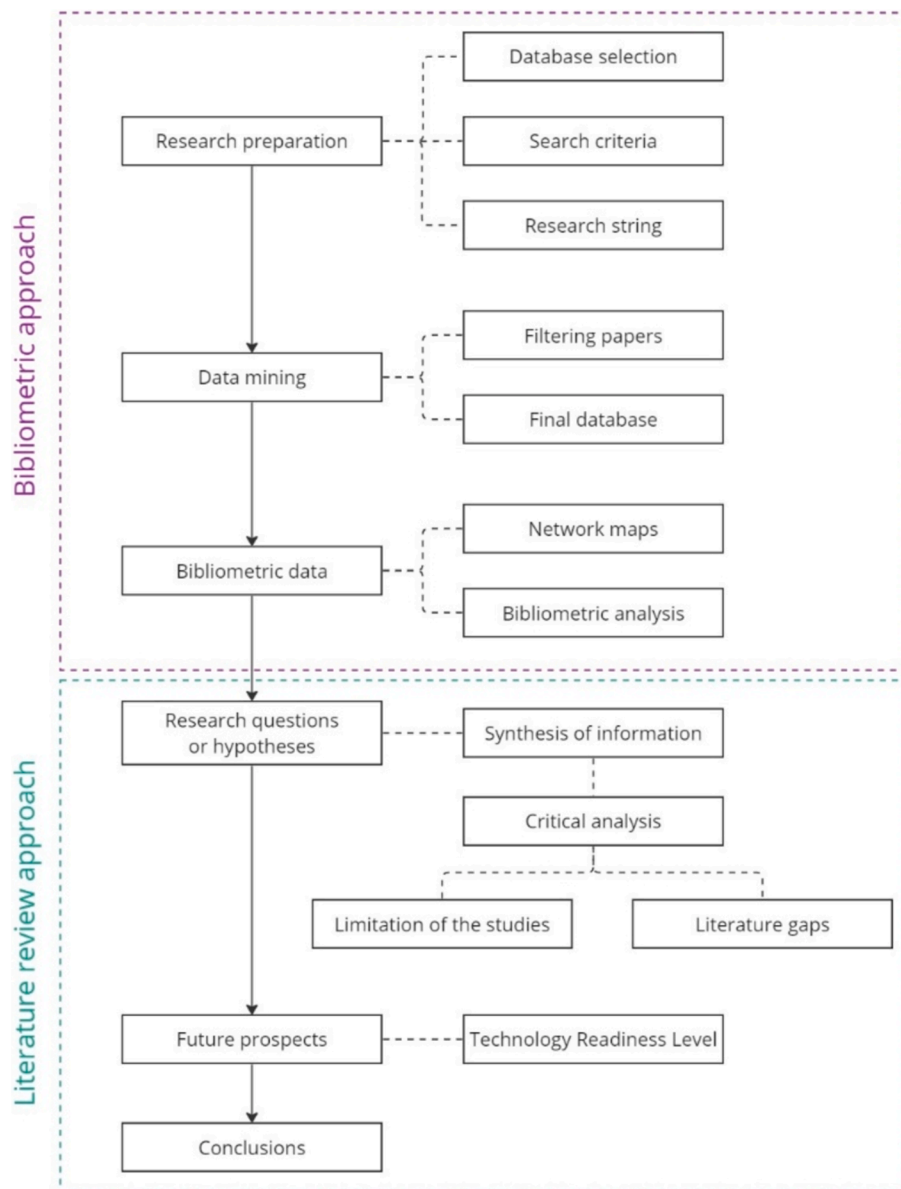


Fig. 1. Flowchart of the schematic structure.

material is colored and above which it becomes colorless, resulting in a modification of the material's optical properties. The temperature coefficient of the TT is typically consistent with that of the construction sector (Berardi et al., 2020). This makes it suitable for use in the production of paints, roofs, and external coatings of buildings, among others and additionally tends to reduce the material's deterioration.

Although thermochromic capacity is a relatively new subject in the transportation infrastructure area and still little explored in the literature (Segundo et al., 2021), the number of publications on this theme has shown an increasing trend in the last decade (Rocha Segundo et al., 2022; Cheng et al., 2018). A few examples are the functionalization of asphalt pavements through the use of modified asphalt binder or mixtures to minimize temperature variation (Hu and Yu, 2020; Li et al., 2020; Xie et al., 2023), resulting in materials with greater mechanical performance and better resistance to aging and the proposed of innovative road engineering applications using thermochromic sensors through functionalized road markings to improve road safety (Lima et al., 2022). This has been a widely researched topic (Hu and Yu, 2016).

The investigations have focused on controlling temperature variation and reducing heat absorption by the pavement. According to Hu

(Hu and Yu, 2013), the surface temperature of a thermochromic asphalt binder can be reduced by up to 6.6 °C compared to the base binder (Hu and Yu, 2013). Similarly, thermochromic hot mix asphalt (HMA) can keep surface temperatures an average of 5 to 15 °C cooler than base HMA during summer (Hu and Yu, 2020). Despite these promising results, there is significant variation in the TCM content, methods, and procedures used for functionalization, which complicates the standardization and comparison of thermal improvement results in these scientific studies and hinders the practical application of thermochromic technology in road engineering.

Few studies have explored the combination of different cool pavement techniques to mitigate UHI effects. Zhang et al. (Zhang et al., 2022) initiated an analysis combining open-graded asphalt layers with a thermochromic spray coating on the surface. Their key findings showed a reduction in surface temperature by 10.57 °C in an experimental indoor solar radiation simulation and a maximum air temperature decrease of 1.5 °C at a height of 1.5 m when a CFD simulation applied the thermochromic coating across all paved areas of Shanghai, China (Zhang et al., 2022). These findings highlight the importance and necessity of using combined techniques, such as permeable and reflective

layers, to improve the thermal balance of pavement. It also emphasizes the need for further research into obtaining thermodynamic parameters of materials and predicting the interactions within the pavement-atmosphere system and the feasibility of scaling lab-based methods to field applications.

In addition to thermal benefits, researchers have also investigated the mechanical and rheological performance of thermochromic asphalt. Most findings suggest that thermochromic powders can enhance the high-temperature stability of asphalt binder, as evidenced by improvements in softening point, viscosity, complex modulus, penetration, phase angle, and dynamic shear rheological tests (Li et al., 2020; Zhang et al., 2017; Chen et al., 2019). According to (Li et al., 2018; Zhang et al., 2018), the additions of thermochromic powders had only a slightly impact on the physical and rheological properties of SBS-modified asphalt compared to the base binder. Furthermore, Du et al. found that the use of thermochromic microcapsules improved the fatigue and aging resistance of both the base asphalt and SBS-modified asphalt (Du et al., 2019). However, there is still a significant lack of knowledge regarding the durability and long-term aging resistance of thermochromic asphalt mixtures in current literature.

The evidence suggesting that controlling the temperature of asphalt composites can enhance their mechanical performance and increase resistance to aging (Du et al., 2019), combined with the fact that this also helps to mitigate the effects of UHI by reducing heat absorption (Hu and Yu, 2013), indicates that the thermochromic approach could offer both environmental and economic benefits. These benefits include improving ambient thermal comfort, reducing energy consumption, and promoting the well-being and health of the population, especially in regions with hot climates (Zhang et al., 2022; Xu et al., 2021; Garshasbi and Santamouris, 2019). Therefore, analyzing the impacts of maintenance and rehabilitation frequency, reductions in air temperature, and life cycle assessments are among the understudied areas that should be considered in this field.

The purpose of this paper is to characterize the scientific scenario concerning the research in thermochromic asphalt mixtures. To achieve this, a bibliometric analysis and a systematic review of the available literature will be conducted. The review will discuss the state-of-the-art in terms of thermal, optical, and mechanical performance. Additionally, the main scientific gaps will be inferred, and future perspectives will be considered. Thus, the first approach will address the publication evolution, co-authorship, leading countries, and institutions. The second approach will identify and compare materials, thermochromic incorporation methods, changes in thermochromic asphalt mixture

properties, and the potential to mitigate UHI effects.

2. Methods

Fig. 1 presents the flowchart of the schematic structure of this review, which comprises two main parts: the bibliometric approach and the literature review approach.

The methodology used for this bibliometric analysis can be summarized in three steps. To begin the research, Scopus was selected as the citation database due to its significance, multidisciplinary, and status as one of the largest international peer-reviewed databases. Search criteria were defined to refine the results, including language and document type. Only papers and reviews written in English were included in the search to avoid duplicates and false-positive records (Rocha Segundo et al., 2022; Dos Santos et al., 2021). The search strings were then defined as “thermochrom*” AND “asphalt” OR “bitum*” to find literature on the application of thermochromism in asphalt/bituminous binders and mixtures. The term ‘asphalt binder’ is commonly used by American researchers, while ‘bitumen’ is preferred by Europeans. Therefore, geographic restrictions were not considered in this study. A search was conducted on Scopus using this string combination, with no limitation on the period, on January 25, 2024. The bibliometric analysis included 35 papers and searched titles, abstracts, and keywords.

The data mining process involved manually filtering papers to ensure alignment with the purpose of this work and removing any duplicates. After reviewing all abstracts, the final database consisted of 31 documents.

The third part of this approach involved developing a bibliometric analysis with statistical tool support using the free software VOSviewer. This software can generate network maps from bibliometric data, such as bibliographic coupling, citation, co-authorship, and co-citation. To guide the bibliometric analysis, the following research questions were defined:

- What is the temporal evolution of this theme?
- What are the most frequently used keywords in the papers?
- Who are the most relevant authors?
- What are the most important papers in the scientific community?
- Which are the most relevant research institutions? And where are they located?
- What are the most important sources (scientific journals) for this area?

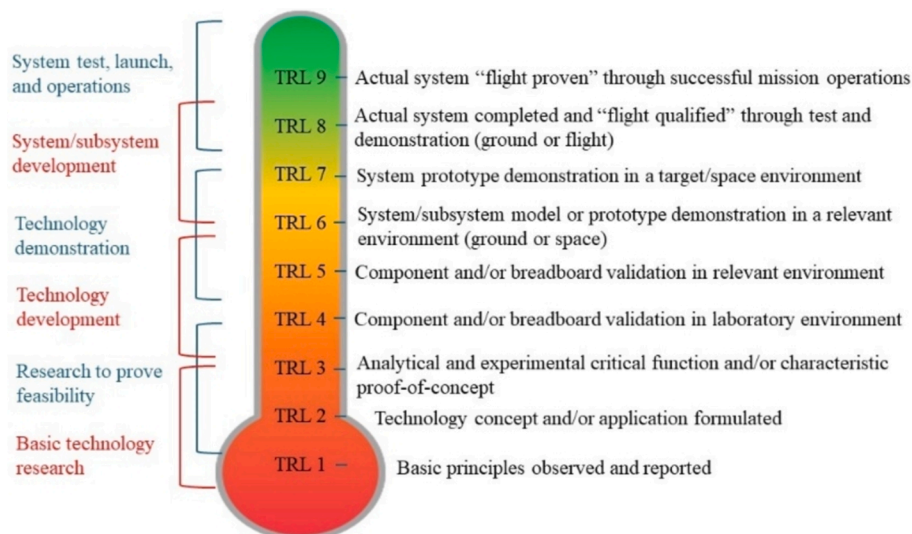


Fig. 2. TRL scales and respective definition (Aeronautics et al., 2017).

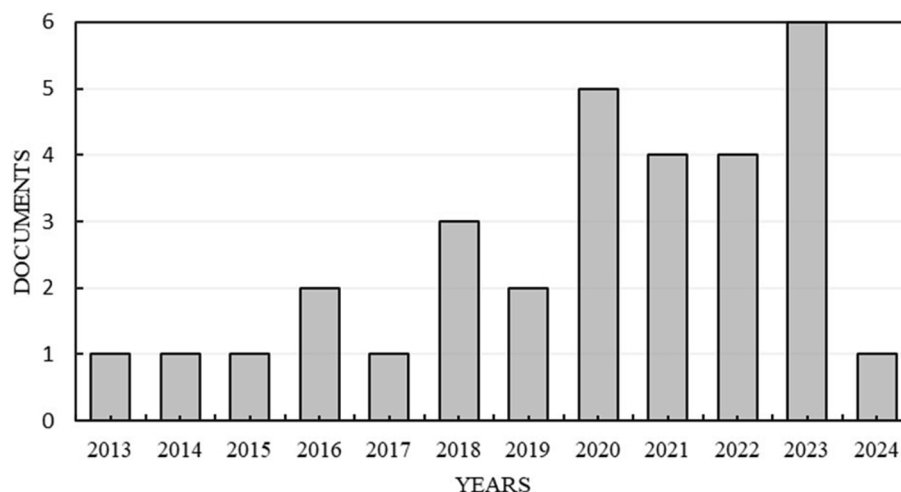


Fig. 3. Evolution of the thermochromic asphalt mixtures publications through the years.

Moreover, the results of visual maps were analyzed to characterize the current panorama of the scientific research of thermochromic asphalt mixture worldwide.

The systematic review focuses on the state-of-the-art in terms of thermal, optical, and mechanical performance of thermochromic asphalt pavements. Therefore, the methodology used for the literature review analysis in the final database involved answering to the following guiding questions:

- Which thermochromic materials were used? And what are their main physical characteristics?
- What were the incorporation methods of the thermochromic powders into the asphalt binder or mixture?
- Regarding thermal, optical, and mechanical performance, what were the main results achieved with the thermochromic asphalt materials?
- What are the main research gaps in the literature? And what are the main future perspectives?

To enhance the comparison and synthesis of key information in papers, three tables were generated. The first and second tables presents the thermal and optical outcomes concerning the incorporation of the

thermochromic materials into asphalt binders and mixtures respectively, while the second table summarizes the mechanical results. The tables provide a summary of the type, composition, and physical characteristics of thermochromic materials, as well as the incorporation methods used, highlighting the outcomes of each study. A critical analysis was conducted on the main outcomes, discussing, comparing, and evaluating them from a research progress perspective associated with cooler pavements results and UHI impacts. The limitations of the studies and research gaps between these mitigation technologies and their effects on the urban environment were also discussed. Finally, this study proposes prospects and conclusions by evaluating the current status of research on mitigating the urban heat island (UHI) impact of thermochromic pavement technologies by the Technology Readiness Level (TRL) method adapted. The TRL is a formalized tool originally developed by NASA to assess and communicate the maturity of technologies during the development process. This method employs a scale ranging from 1 to 9, with each level reflecting a progressive advancement in technological development. Level 1 indicates the initial observation of basic principles, while Level 9 signifies a fully mature technology, proven through successful deployment in its operational environment (Fig. 2). In this work, the scientific gaps and suggestions for

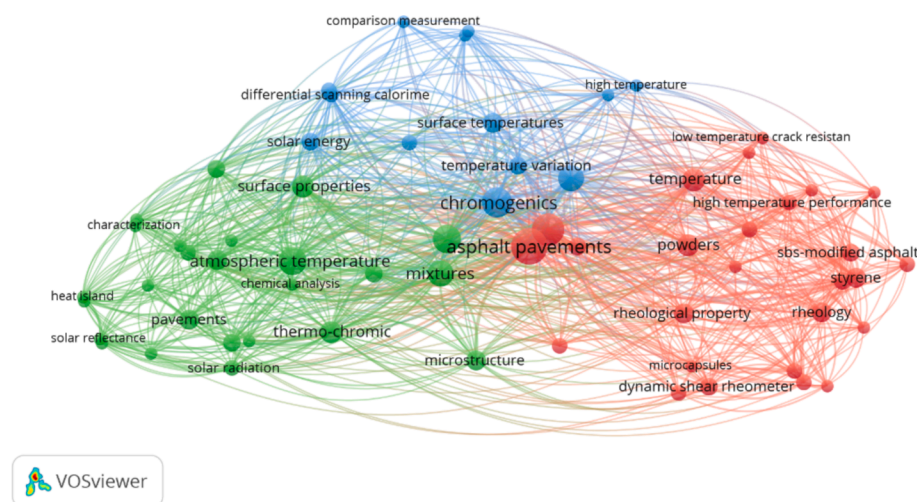


Fig. 4. Co-occurrence network of the most frequent terms present in titles, abstracts and keywords of scientific papers published on thermochromic asphalt mixtures; (i) red cluster related to mechanical properties and aging resistance; (ii) green cluster related to environmental impact of thermochromic solutions; (iii) blue cluster related to laboratory tests and energy efficiency. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

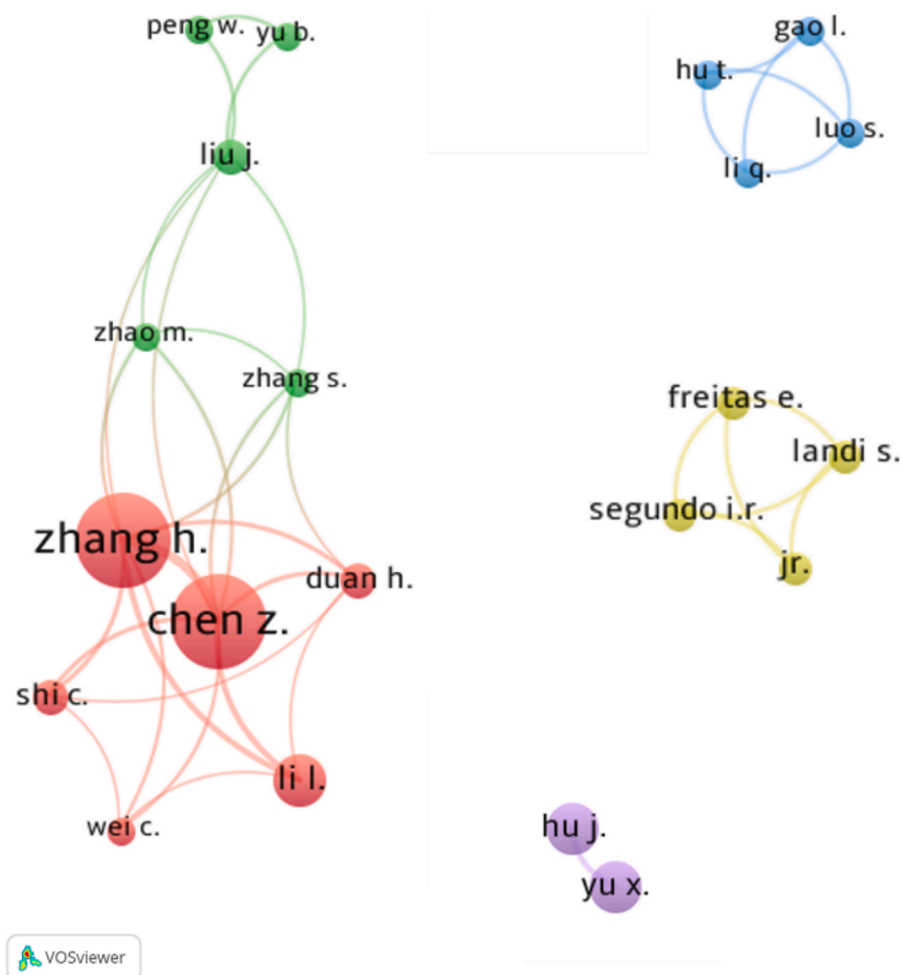


Fig. 5. Network map of co-authorship by author.

future work were grouped into blocks according to TRL: research lab (TRL 1–3); simulated world (TRL 4–7); and real world (TRL 8–9).

3. Results

3.1. Bibliometric results and discussions

Fig. 3 shows the evolution over time of the number of publications on thermochromic asphalt mixtures in the Scopus database. It is evident that there was no published paper before 2013, indicating a growing trend of research on this theme. An increase in the number of papers since 2018 is also observed. The highest number of publications was recorded in 2023, with six papers. It is worth noting that one paper has already been published in 2024, despite the year has just started (the search was conducted on January 25, 2024). This information suggests that thermochromic capability is a recent and innovative subject that has generated interest in scientific investigations of asphalt materials and structures.

3.1.1. Main terms

The network map presented in Fig. 4 plots the main index keywords of each assessed document and their co-occurrence frequency. Only terms that occurred at least twice are included, and there are no duplicated or similar terms on the map. The most frequently used terms in the text are 'asphalt mixtures', 'chromogenic', and 'thermochromic materials', with 15, 13, and 11 occurrences, respectively. These terms are highly relevant to all clusters and are strongly connected to most other terms. Chromogenic materials are materials that can change color

reversibly due to variations in certain properties, such as pH, temperature, pressure, and electrical charge. Thermochromic materials are a type of chromogenic material that changes its optical properties in response to changes in temperature. The terms 'binders' and 'asphalt mixtures' are also related to this topic.

The study identified three clusters based on color and main terms: (i) RED – 'powders', 'rheological property', 'temperatures', 'physical properties', 'SBS-modified asphalt', and 'aging resistance'; (ii) GREEN – 'atmospheric temperature', 'surface properties', 'urban heat island effects', 'microstructure', and 'durability'; and (iii) BLUE – 'surface temperatures', 'Fourier Transform Infrared Spectroscopy', 'solar energy', 'differential scanning calorimeter', and 'specific heat'. Cluster (i) is related to the mechanical properties and aging resistance of asphalt binders and mixtures, as well as thermochromic materials. Cluster (ii) contains terms related to the environmental effects of thermochromic solutions. The keywords in cluster (iii) are mainly related to laboratory tests and measures of energy efficiency of functionalized capabilities.

3.1.2. Co-authorship

Fig. 5 shows the results of the co-authorship network. The network map displays clusters of authors who have co-authored at least two papers, forming five distinct clusters. The nodes show the names of the authors, the lines represent co-authorship relations, and each color corresponds to authors who worked together in the same paper. The red cluster, composed of Chinese authors, is the most strongly connected and accounts for the largest number of publications (big node size). Zhan H. and Chen Z. can be considered the most productive researchers, with thirteen co-authored papers and over 410 citations. The primary focus of

Table 1
Most productive authors on the theme.

Author	Documents	Citations	Country	Affiliation
CHEN, Z. ZHANG, H.	13	410	China	Hunan University
HU, J. YU, X.	6	202	US	Case Western Reserve University
LI, L.	5	93	China	Hunan University

Table 2
Most cited documents in the Scopus database on the theme.

Title	Authors	Journal	Citations	Year
Physical, rheological, and chemical characterization of aging behaviors of thermochromic asphalt binder	Zhang, H., Chen, Z., Xu, G., Shi, C.	Fuel	138	2018
Improvement of thermal and optical responses of short-term aged thermochromic asphalt binder by warm-mix asphalt technology	Chen, Z., Zhang, H., Duan, H., Shi, C.	Journal of Cleaner Production	68	2021
Evaluation of aging behaviors of asphalt with different thermochromic powders	Zhang, H., Chen, Z., Li, L., Zhu, C.	Construction and Building Materials	52	2017
Experimental study of sustainable asphalt binder	Hu, J., Yu, X.	Transportation Research Board	48	2013
Characterization of the optical and mechanical properties of innovative multifunctional thermochromic asphalt binders	Hu, J., Gao, Q., Yu, X.	Jour. of Materials in Civil Engineering	43	2015

Zhang H. and Chen Z.'s work was the investigation of the mechanical and cooling performance of thermochromic binders and asphalt mixtures.

On the other hand, it can be observed that the other clusters have a low number of co-authorship links, which may indicate a high variability of co-authors across publications. When different research teams authored each paper, the authors did not meet the threshold to appear on the network map. This is likely the case for the light blue, light purple, and yellow clusters, which are composed of Chinese, American, and Brazilian-Portuguese researchers, respectively. It is important to highlight the emerging hub for researching the use of thermochromic material in paving applications with Brazilian co-authorship, due to the completely different climate conditions of this country deal comparing to China and the USA, which can strongly contribute to increasing the knowledge of its performance as innovative and sustainable material.

A total of forty-nine authors have written about thermochromic asphalt mixtures, and the top five most productive individual authors have been selected. Table 1 shows the authors with the highest number of publications and citations related to the topic. The authors are from Chinese and American organizations, which indicates that these institutions can be the main centers of scientific knowledge on this topic.

The five most relevant scientific papers according to the number of citations in the Scopus database are listed in Table 2. These papers were primarily published by two research groups from Hunan University (China) and Case Western Reserve University (USA), aligning with the aforementioned analysis of scientific productivity. They had at least 40 citations each, and all the papers were mainly concerned with thermochromic asphalt binders' mechanical and optical performance, aging behavior, and combination with a warm-mix technology. Here, it is possible to see the tendency for research works to use thermochromic asphalt functionalization in the current literature.

Regarding the network map of co-authorship by organizations (Fig. 6), seven clusters of institutions that have developed research on thermochromic asphalt mixtures can be observed. Although there are links between different organizations, they are mostly between groups of researchers from the same university/nationality, which indicates a weak geographical spread of collaboration.

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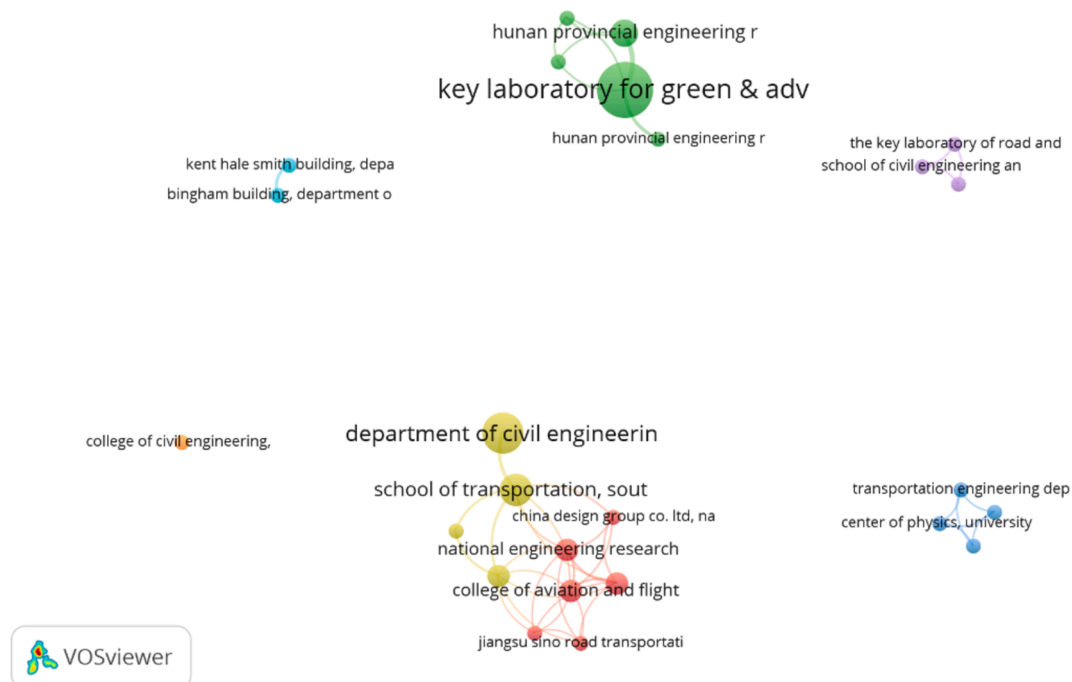


Fig. 6. Network map of co-authorship by institutions.

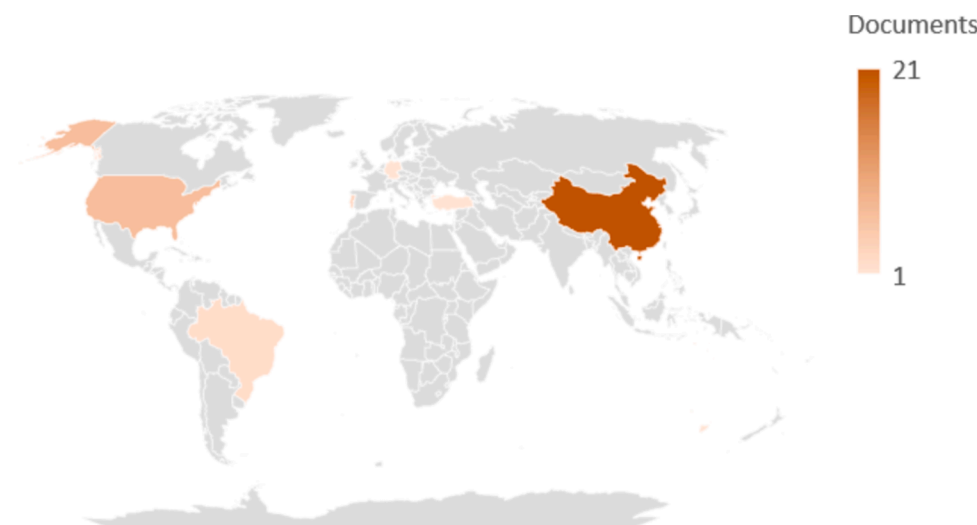


Fig. 7. Number of published papers by country.

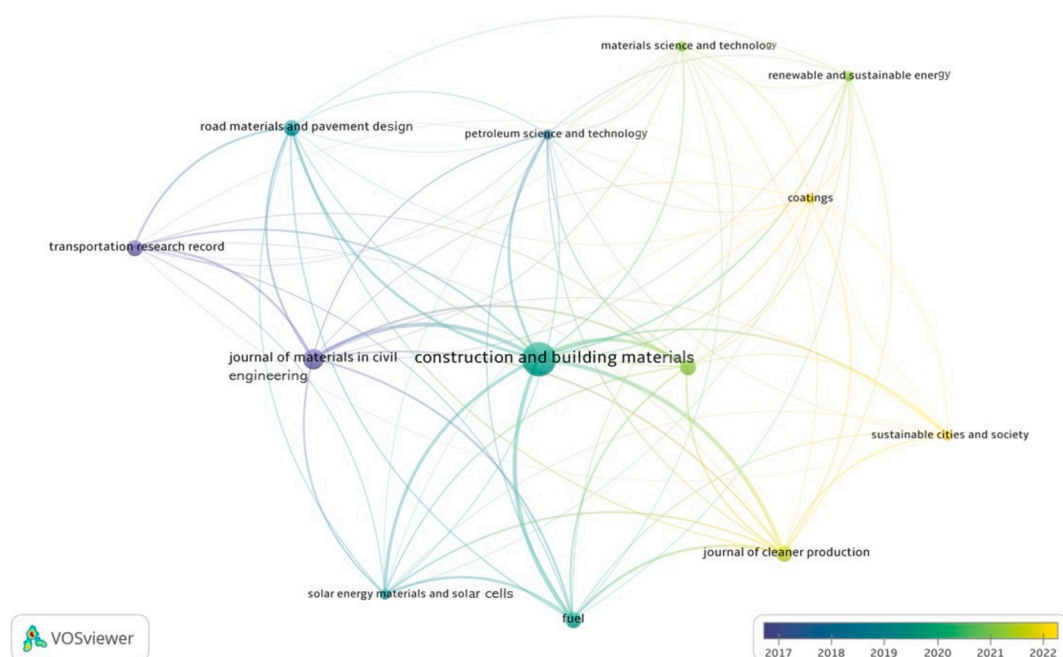


Fig. 8. Bibliographic coupling by sources with temporal metrics.

and Application Technology, from Hunan University – China, is the most relevant organization with the largest number of partnership publications. However, these network links are only with other Chinese groups. Two clusters have links with groups of different nationalities: Portugal – Brazil and China – United States. Therefore, in addition to weak links, there is a low geographical spread of research.

The map of the co-authorship network by country was also examined, and it showed only eight countries, divided into two clusters with no connections between them. The first one is composed of China and the United States, and they represent the most productive countries with at least 70 % and 29 % co-authorship in the papers, respectively. The link between Portugal and Brazil (the second cluster) has only one document for each. This scene reinforces the incipient partnership research and the concentration of published papers in a few countries. The number of published papers by country can be seen in Fig. 7.

3.1.3. Sources

Fig. 8 shows the bibliographic coupling network by source, presented using a temporal metric. The most important journal is Construction and Building Materials with 10 published documents and the highest number of citations (181), followed by the Journal of Materials in Civil Engineering with 3 published documents on the topic. The main fields of knowledge from which the sources come are Civil Engineering, Materials Science and Energy.

More recently, due to the multidisciplinary nature of the topic, other scholarly sources have published papers in a variety of areas, such as sustainable/environmental and smart/technology cities. Examples include the Journal of Cleaner Production and Sustainable Cities and Society.

3.2. Systematic review results

The following sections provide a summary of the information

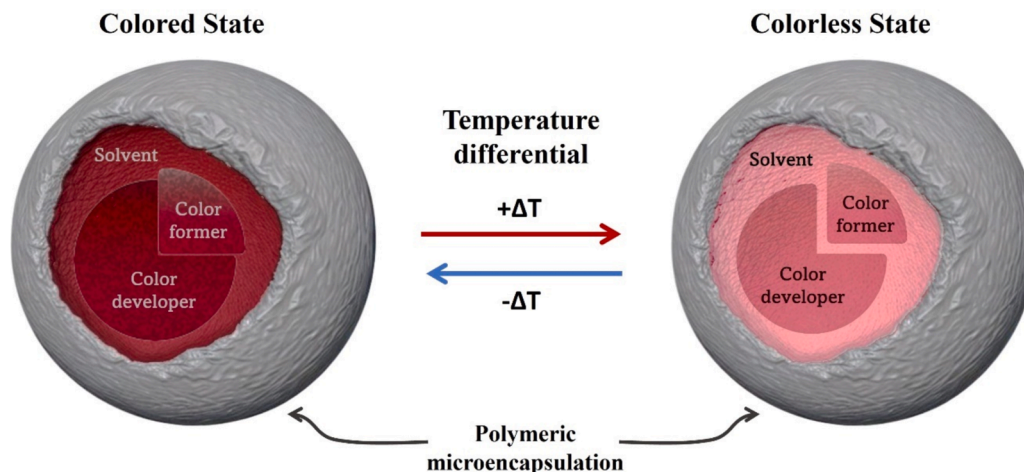


Fig. 9. Schematic thermochromic mechanism in Leuco dyes (Garshasbi and Santamouris, 2019).

gathered through a bibliometric approach from the final database. They present the main results related to the use of thermochromic materials in asphalt binders or asphalt mixtures. The analyses focused on three performance approaches: thermal, optical, and mechanical properties.

3.2.1. Raw materials, obtaining, and incorporation methods

Thermochromic materials (TCM) are based on color changes in their visible optical properties upon temperature changes, due to a chromogenic core. TCM are organic mixtures composed of at least a color former (Leuco dye), color developer, and co-solvent. The color change mechanism is reversible and occurs at a specific transition temperature (TT), determined by the co-solvent melting point. Below TT, the leuco dye reacts with the developer, causing the material to become colored due to the absorption peak in the visible range. When the material is heated, the solvent-developer interaction becomes dominant, and above the TT, the color is lost (Fig. 9) (Hu and Yu, 2013; Hu et al., 2014).

The characterization of the chromogenic core is crucial for understanding the behavior of thermochromic materials. In the articles reviewed, the TCM are primarily of industrial origin, making it challenging to fully understand their composition, quantities, and the proportion of the materials involved. However, the chemical analysis of the chromogenic core has been mostly conducted using Fourier Transform Infrared (FTIR) spectroscopy (Li et al., 2020; Zhang et al., 2017; Chen et al., 2019; Zhang et al., 2018; Chen et al., 2022). The main results of the FTIR spectrum identified key chemical bonds through absorption bands centered around $3,365\text{ cm}^{-1}$ and $1,511\text{ cm}^{-1}$, corresponding to hydroxyl ($-\text{OH}$) and benzene skeleton vibrations, respectively. These are likely derived from bisphenol A (BPA). The characteristics of methyl stearate were identified by the presence of functional groups such as methyl ($-\text{CH}_3$), methylene ($-\text{CH}_2$), and carbonyl ($\text{C}=\text{O}$), indicated by peaks at $2,923\text{ cm}^{-1}$, $2,848\text{ cm}^{-1}$, and $1,740\text{ cm}^{-1}$, respectively. Peaks centered around $1,556\text{ cm}^{-1}$ and 813 cm^{-1} were attributed to the melamine-formaldehyde resin used as the microcapsule shell. More specifically, the main chemical composition of the leuco dyes includes varying proportions different materials: 3-Diethylamino-6-methyl-7-2,4-xyldinofluoran, 3-N-p-tolyl-N-ethylamino-7-methyl-fluoran, and 3-(4-(diethylamino)-2-(hexyloxy)phenyl)-3-(1-ethyl-2-methyl-1H-indol-3-yl)-4-azaphthalide, for black, red, and blue pigments, respectively (Li et al., 2020; Zhang et al., 2017; Chen et al., 2019; Zhang et al., 2018; Chen et al., 2022).

Studies indicate that BPA is commonly used as a color developer in thermochromic pigments due to its ability to produce darker tones and higher color contrast. However, BPA is known to be toxic and carcinogenic, raising concerns about its environmental impact. This is particularly relevant for TCM applications in pavements, where surface damage to microcapsules can release BPA into the environment. In the

textile industry, Tözüm et al. (Tözüm et al., 2021) explored the synthesis of thermochromic microcapsules using phenolphthalein (PP) as an alternative color developer. Their findings demonstrated that these PP-based microcapsules have significant potential for use in textile materials, offering reversible color change and thermal indication without the harmful effects associated with BPA.

The thermochromic core is typically microencapsulated with polymer materials, such as trioctanoinand (Hu and Yu, 2020; Hu and Yu, 2016; Hu et al., 2014; Hu et al., 2015; Li et al., 2020) or Melamine-formaldehyde resin (Zhang et al., 2022; Chen et al., 2019; Du et al., 2019; Li et al., 2020), serving as a shell. The aim of the microencapsulation strategy is to extend the thermochromic capacity, hold the entire thermochromic system together, and protect it from ultraviolet (UV) radiation exposure and physical disruption (Du et al., 2019). The susceptibility of TCM microcapsules poses a significant challenge for road applications. The primary drawback of using these materials in practice is their rapid aging (Garshasbi and Santamouris, 2019), with UV radiation being the main degradation mechanism that reduces thermochromic reversibility (Karlessi et al., 2009). Environmental exposure can cause the capsules to break, resulting in the loss of the core material and, consequently, the thermochromic capability. Zhang et al.'s (Zhang et al., 2022) physical analysis of TCM materials revealed that during transportation, changes in environmental conditions (such as direct solar radiation) caused some microcapsules to break, leading to the leakage of the color-changing content. No studies have been found on the potential effects on mechanical performance of leaking encapsulated material into the binder or asphalt concrete regarding its mechanical performance and durability, highlighting a possible opportunity for future research.

Initial results indicate that microcapsules with shells made of melamine-formaldehyde resin tend to be more susceptible to damage during heating or binder mixing compared to those made with trioctanoin (Li et al., 2020). Furthermore, studies suggest that TCMs have been manually stirred into paint cool coatings to prevent damage to the microcapsules (Berardi et al., 2020; Zhang and Zhai, 2019), which can also increase the heterogeneity of the final product. This caution was not observed in studies involving the modification of asphalt binders, where the use of magnetic stirrers and high rotation speeds prevailed (Zhang et al., 2022; Hu et al., 2015). This underscores the need for improvements in the photoaging resistance and durability of microencapsulated thermochromic materials. Exploring the use of biopolymers in the chemical composition and synthesis of microcapsules could be a valuable research opportunity to enhance their durability and sustainability.

However, raw thermochromic powders demonstrate good thermal stability even when subjected to high temperatures. According to Hu and Yu (Hu and Yu, 2016), thermogravimetric analysis shows that the main pyrolysis of TCM occurs after $200\text{ }^{\circ}\text{C}$, indicating that this

temperature is the material's critical point and minimal thermal weight loss occurs below it. Even after heating for 2 h at 200 °C, the TCM powder maximum weight loss observed was only 7.8 % (Hu and Yu, 2016). This suggests a high likelihood that these capsules will perform well within the typical temperature range experienced by in-service pavements. All the reviewed papers used commercial thermocapsules with a transition temperature (TT) of 31 °C, indicating that this temperature is likely suitable for asphalt material applications. There have been successful reports of modifying binder and asphalt mixtures for different climate regions, including China (Chen et al., 2021) and the US (Hu and Yu, 2020; Hu and Yu, 2013). There is, therefore, room for contributions focused on the synthesis and application of thermochromic microcapsules with higher TT to better accommodate the realities of naturally hotter tropical climates, as well as the increasing frequency of heat waves due to climate change.

Regarding to physical characteristics, TCM powder consists of spherical microcapsule particles, with sizes predominantly ranging from 1 to 10 µm. The particle size can have controversial effects on thermochromic coatings. On one hand, larger particles can negatively impact reflectivity by reducing the surface area available to reflect sunlight. On the other hand, the spherical shape of the particles prevents them from highly interacting particles or agglomeration, leading to better dispersion in the coating. This improved dispersion allows the optical and thermal effects of the thermochromic material to be fully realized (Zhang et al., 2022). Li et al. (Li et al., 2020) analysis of two thermochromic additives with different particle sizes ($TM_1 = 3 \sim 10 \mu m$; $TM_2 = 1 \sim 10 \mu m$) confirms that, following the same mixing process, TM_2 (smaller sizes) particles adhered to the surface of the binder, resulting in better uniformity dispersion and diffusivity compared to TM_1 particles. The differential diffusivity caused by particle size significantly affects the mechanical performance of additives and it is likely the reason why TM_2 particles presented better rutting resistance and cooling effects (Li et al., 2020; Li et al., 2020). Therefore, controlling the particle size of additives used to modify the binder is a fundamental scientific and technical issue that can significantly influence the performance of thermochromic asphalt binder.

In terms of optical characteristics, after the TT, the TCM color changes increases the spectral reflectance in the visible and near-infrared (NIR) spectrum (Hu et al., 2015) and it is dependent of their specific color pigment (Hu and Yu, 2016). The study conducted by Zhang et al. evaluated the effects of six different thermochromic powder colors on the optical and thermal properties of coatings. The results showed that TCM has a higher total solar reflectance compared to traditional cool materials. Red and black TCM powders were found to be more suitable for use in thermoregulation coating pavements, considering both reflectance characteristics and visual effect (glare) (Zhang et al., 2022). Similar results were found by Hu & Yu's to thermochromic asphalt binder, which the reflectance increase was the highest for black powders, followed by blue and red (Hu et al., 2015). Additionally, it was found that the optical properties improved with an increase in powder content (Hu and Yu, 2016).

Some studies combine thermochromic powders and semiconductors to improve the optical characteristics of modified binder or asphalt mixtures. According to Hu & Yu's optical characterization, nano-TiO₂ particles have a solar reflectance of up to 90 % in the NIR solar spectrum, while the thermochromic powder reflectance range is 60–80 % (Hu and Yu, 2020). Zhang et al. investigation of the effect of different rutile TiO₂ contents on the optical properties of thermochromic coatings found that 2 % and 5 % TiO₂ contents are the optimal levels for thermoregulation at high and low temperatures for red and black coatings, respectively (Zhang et al., 2022). TiO₂ is a semiconductor widely used in pavement applications due to its photocatalytic properties (Segundo et al., 2021; Rocha Segundo et al., 2019). It also possesses other characteristics, such as hydrophobicity and the ability to degrade adsorbed organic materials (Segundo et al., 2021). These properties could contribute to the development of thermochromic solutions with multiple capabilities, such as

photocatalytic, super hydrophobicity, and self-cleaning, which have not yet been widely explored in the literature of the field.

Regarding the incorporation of thermochromic powders, most studies have utilized a wet mixing process to modify asphalt binders (Hu and Yu, 2020; Li et al., 2020; Hu and Yu, 2016; Hu and Yu, 2013; Zhang et al., 2022; Zhang et al., 2017; Chen et al., 2019; Li et al., 2018; Zhang et al., 2018; Du et al., 2019; Hu et al., 2014; Chen et al., 2022; Hu et al., 2015; Li et al., 2020; Chen et al., 2021; Hu and Yu, 2016; Hu and Yu, 2020; Yu et al., 2022; Bo et al., 2022; Zhang et al., 2020; Chen et al., 2021; Civan and Kurama, 2021; Li et al., 2018). This process involves mixing the heated binder with the thermochromic additive until a homogeneous mixture is produced, using the weight of the base asphalt binder. Thermochromic asphalt mixtures also can be produced using the bulk incorporation dry mixing process, which involves replacing a portion of the fine aggregate or filler with thermochromic materials. Chen et al. (Chen et al., 2022) investigated both functionalization techniques, and their key findings indicated that the wet process promotes a more uniform distribution of thermochromic microcapsules within the mixtures, while the dry process is more effective in preserving the integrity of the microcapsules. The distinct characteristics of the wet and dry methods, along with different mixing procedures in the dry process, can significantly impact the mechanical performance of thermochromic asphalt mixtures. Overall, the asphalt mixtures prepared using the wet process generally exhibit better mechanical performance and a superior cooling effect compared to those prepared using the dry process (Chen et al., 2022). While both functionalization likely ensures the best TCM immobilization, they can also impact pavement performance. Mastering the mechanical behavior of thermochromic asphalt mixtures and binders is a primary focus of current research (Li et al., 2020; Li et al., 2018; Du et al., 2019; Hu et al., 2015; Zhang et al., 2020; Li et al., 2018).

The technique of spraying coating is another possibility for functionalization, and it involves applying a TCM solution over the surface of the compacted pavement (Zhang et al., 2022; Lima et al., 2023). However, this technique has rarely been evaluated in recent literature on thermochromic asphalt. In the study of photocatalytic solutions, the surface spraying functionalization technique was found to be more efficient. It requires less semiconductor and does not cause mechanical alterations in the asphalt concrete (Rocha Segundo et al., 2019). However, ensuring proper immobilization of the solution to the substrate remains a challenge (Carneiro et al., 2013; da Rocha Segundo et al., 2019). To improve immobilization conditions of thermocapsules to the asphalt substrate, some studies have developed solutions based on epoxy resin. These solutions maintain the thermochromic capacity even after weathering tests (Lima et al., 2023; Li et al., 2019). Yi et al. investigated the friction reduction characteristics of reflective asphalt mixtures and demonstrated adherence to an asymptotic model, with the reduction stabilizing at 48 BPN after 150,000 cycles (Yi et al., 2023). Prior to the abrasion process, an increase in the volume of the sprayed solution resulted in reduced friction, and consequently in a smoother microtexture. However, the efficiency of the solutions after abrasion and the impact of functional characteristics such as friction and texture of the coatings remain a research gap in surface spraying functionalization (Lima et al., 2023; da Rocha Segundo et al., 2019; Yi et al., 2022; Hu et al., 2021).

On one hand, functionalization through spray coating acts as a physical barrier on the pavement surface, which increases the reflective area and potentially enhances thermochromic efficiency. On the other hand, this technique leaves the material more exposed to the elements, making it more susceptible to environmental degradation and traffic wear, which inevitably compromises its durability. Furthermore, investigating how changes in pavement surface characteristics, particularly friction and texture, impact road safety aspects (such as increased stopping distances and the number of traffic incidents) represents a gap in the literature regarding the surface spraying functionalization technique.

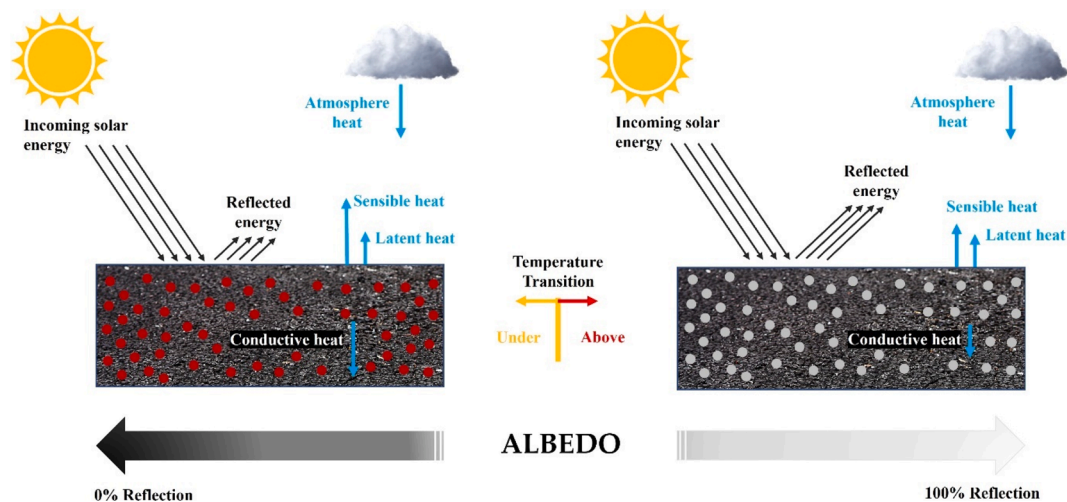


Fig. 10. Schematic optical-thermal balance on the pavement surface (Wang et al., 2016).

3.2.2. Thermal and optical performance of thermochromic asphalt mixtures

Thermochromic asphalt pavements should, in principle, exhibit different responses to solar radiation than regular asphalt pavement due to their characteristics. Fig. 10 compares a conventional asphalt pavement to a thermochromic pavement in terms of changes in optical and thermal characteristics. Conventional asphalt pavement absorbs and stores most of the incident solar radiation due to its black color, contributing to the heating of urban centers. In contrast, thermochromic pavement has a colorless surface that reflects large amounts of solar energy after TT. This higher solar reflectivity lowers the surface temperature and reduces energy absorption in the form of heat by the pavement (Zhang et al., 2022). However, issues to prevent glare and other adverse visual effects in order to ensure safety on colorless road surfaces need to be considered, such as colorimetry analysis of thermochromic solutions and lightness control (Zhang et al., 2022).

Thermochromic pavements are a type of passive cool pavement technology that can modify the heat absorption and structural performance of pavements. This is achieved by increasing the solar reflectance and thermal emissivity of the materials (Xu et al., 2021). The pavement harvests solar energy in three ways, through: sensible heat and latent heat released by the pavement, and conductive heat that travels through the pavement structure, as showed in Fig. 10. The release of sensible heat is a significant contributor to the urban heat island (UHI) phenomenon (Qin and Hiller, 2014; Fortuniak, 2008), due to the heat exchange directly influences the temperature of the pavement surface and surrounding air. Prospective technologies aimed at reducing the UHI effect focus on minimizing the release of sensible heat (Xu et al., 2021).

There are several innovative pavement technologies used in urban areas to mitigate the effects of the UHI phenomenon. These technologies can be categorized into four main types based on the pavement solution studied: highly reflective, permeable, thermally modified, and energy-harvesting pavement systems (Xu et al., 2021).

Thermochromic pavements, in particular, offer a significant advantage by acting as smart sealing layers. These pavements are highly reflective in summer and less reflective in winter, giving to them an important characteristic of thermoregulation, which could modulate the pavement surface temperature more efficiently (Karlessi et al., 2009). The reversible color change of thermochromic pavements in response to external temperature variations alters the surface albedo, i.e., the ratio of visible solar radiation reflected by a surface to the total visible radiation incident on it. There are reports of the maximum albedo increase of thermochromic coating was measured to be 43 % when it transferred from colored to colorless phase (Karlessi et al., 2009). Once functionalized, thermochromic pavements are ready for full operation, unlike other types of cool pavements. In comparison, permeable pavements

rely on water for cooling, and in dry conditions, their surface temperature tends to be high (Asaeda and Ca, 2000). Additionally, these pavements are prone to pore clogging under high traffic loads and from vegetation, leading to a quickly reduced efficiency. Permeable paving, however, has other significant advantages such as drainage capacity and reduced surface runoff, increased friction and reduced noise pollution effects (Asaeda and Ca, 2000). Another kind of technology, such as thermally modified pavements (high thermal conductivity) stores heat during the day, causing remission of the heat during nighttime (Dawson et al., 2012), which is the opposite of the desired effect. Finally, energy-harvesting pavement systems come with high initial and maintenance costs, are mostly non-recyclable, and have a high potential for voids between the pipes, that contain circulating liquid to harvest the heat energy, and the asphalt mixture, increasing the risk of debonding (Mallick et al., 2009).

Regarding thermochromic optical performance, the functionalized asphalt materials exhibit a similar trend to thermochromic powder. Specifically, thermochromic asphalt binder and thermochromic asphalt mixtures reflect more near-infrared light than conventional pure asphalt due to temperature-induced color changes (Chen et al., 2022; Hu et al., 2015). In the visible wavelength range of 350 to 550 nm, the reflectance values can increase by over 50 % in the colorless thermochromic phase (Chen et al., 2022). Hu and Yu's main findings indicate that the spectral reflectance properties of thermochromic asphalt binder increase as the powder content increases from 10 to 40 % (Hu and Yu, 2016). To predict the reflectance spectra of thermochromic asphalt binder based on powder content levels, optical mixing formulas such as Maxwell, Mori-Tanaka, and effective medium theory (EMT) were tested. These models provide a reasonable description of the phenomenon and can assist in the multifunctional design and optimization of the thermochromic binder (Hu and Yu, 2016).

Enhanced solar reflectance is preferred because it reflects more solar radiation through the thermochromic effect, resulting in lower asphalt surface temperatures. However, heat exposure, even during production, can affect the thermochromic properties of powders and coatings. Conventional high-temperature asphalt preparation methods may harm the durability of thermochromic microcapsules in the asphalt matrix, undermining the temperature regulation capabilities of thermochromic asphalt materials (Chen et al., 2021). Hu & Yu (Hu et al., 2015), found that subjecting thermochromic binders to 200 °C for 2 h resulted in a decrease in solar reflectance percentages of 4 % for black, 8.5 % for blue, and 15.8 % for red asphalt coatings due to thermal treatment. To minimize excessive reductions in solar reflectance, it is advisable to produce black thermochromic asphalt using the WMA technique, while blue and red asphalt can employ both HMA and WMA technologies (Hu

Table 3

Main thermal and optical results of the use of thermochromic material in asphalt binders and mixtures.

Ref.	Technol. details	Optical properties	Thermal properties
(Hu and Yu, 2013)	TCM asphalt binder with red, blue, and black powder content of 10 % by weight, wet mixing	Notevaluated	TCM binder has higher latent heat (3x), heat capacity (1.2x), and thermal conductivity (1.4x) than pure binder
(Hu et al., 2014)		Notevaluated	From -20°C to 50°C , the specific heat capacity of TCM binder is from 21 % to 33 % higher, while the thermal conductivity is from 5 % to 35 % and the thermal diffusivity is from 3 % to 45 % lower than pure binder
(Hu and Yu, 2016)		TCM binder is more reflective in the near-IR range than pure binder, and it increases with temperature; thermal treatment reduces the solar reflectance of TCM binder by at least 4 % to 15.8 % under 200°C for 2 h.	To keep thermal stability, it is suggested that the blue and red binder should be prepared below 200°C and less than 2 h, to control the weight losses in production conditions
(Hu and Yu, 2016)	TCM binder incorporating powder contents of 10, 20, 30, and 40 % by weight, wet mixing	The spectral reflectance of TCM binder increases as the powder content increases, especially in IR range;	Notevaluated
(Hu and Yu, 2020)	TCM binder incorporating powder 6 %, or 1 % powder + 3 % TiO_2 by weight, wet mixing	6 % powder increases in 43 % the HMA's solar reflectance; After rutting, TCM HMA presents 13–25 % lower solar reflectance;	Notevaluated
(Chen et al., 2021)	TCM binder incorporating powder 6 % by weight and 4 % Sasobit®, wet mixing	6 % TCM + 4 % Sasobit® exhibits the highest optical reflectance (especially within UV and visible light regions) among pure and 6 % TCM short-term aged asphalt binders	Notevaluated
(Zhang et al., 2022)	TCM coating composed by 20 % of red/black and TiO_2 total mass fraction; the water-based silicone acrylic emulsion was used as the binder accounting for 55 wt% of the coating	Increasing TiO_2 content, the reflectance spectra are improved; Considering hiding power, optical reflective characteristics, visual effects, and optical reflective characteristics, the optimal TiO_2 contents in the red and black TCM coating are 2 wt% and 5 wt%, respectively.	Notevaluated
(Chen et al., 2022)	TCM binder incorporating powder content of 6 % by weight, wet process; TCM mixture incorporating powder content of 0.326 % by mass of the aggregates, dry process	To wavelength range from 350 to 550 nm, the reflectance values can be increased by more than 50 % in colorless thermochromic phase	Notevaluated
(Lima et al., 2023)	TCM aqueous solution coating composed by powder 3 % w/v, dye 0.5 % w/v, and epoxy resin 20 mL, superficial sprayed	The addition of resin contributed to an increase in 3 times the colorimeters parameters (ΔE^* , ΔL^*), indicating higher luminosity	Notevaluated
(Xie et al., 2023)	5 % incorporating novel thermochromic pellets, with a TT 45°C , 55°C , and 65°C , to prepare ultra-thin wear layer (UTWC) asphalt mixtures	Notevaluated	The thermochromic pellets have smaller thermal conductivity (9.5 %) and thermal diffusion coefficient (23 %), but higher specific heat capacity (17.5 %) compared to the reference mixtures.
(Chen et al., 2024)	TCM binder incorporating powder content of 6 % by weight, wet mixing	At a temperature larger than 31°C , the solar albedo of TCM HMA is 0.30, which is six times larger than that of the conventional asphalt pavement material	Notevaluated

and Yu, 2016). Chen et al. found that WMA technology can reduce the mixing and compaction temperatures of thermochromic asphalt mixture by 16°C (Chen et al., 2021). Additionally, the use of Sasobit® additive was shown to be effective in preserving the structural integrity of thermochromic microcapsules, reducing the leakage of core materials, and enhancing the optical reflectance in short-term aged thermochromic asphalt (Chen et al., 2021). This highlights that the optical properties of thermochromic microcapsules can vary based on the powder's chemical characteristics and ambient temperature.

While increasing reflectance is desirable for reducing pavement surface temperatures, it is also a huge concern associated with the use of thermochromic pavements. Large areas of highly reflective pavement in urban settings could cause surrounding buildings to absorb the reflected light, potentially exacerbating the UHI effect (Xu et al., 2021; Jia and Wang, 2021). Additionally, during the daytime, the reflected light may lower thermal comfort for people and animals near these pavements (Jia and Wang, 2021). It is essential that studies on thermochromic pavements take these issues into account to determine suitable locations for their application, enhancing to improve local microclimatic conditions and enhance the well-being of the population. More specifically, microclimate simulation studies using software to investigate the effects of replacing conventional pavement surfaces with thermochromic pavement may offer a valuable tool for informing the decision-making process when applying resilient strategies in urban areas.

The addition of thermochromic material to asphalt binder also

modifies its thermal properties and performance. The latent heat of thermochromic asphalt can also be up to 3 times higher than that of conventional asphalt (Hu and Yu, 2013). According to Hu & Yu, the specific heat capacity of thermochromic binder is 21–33 % higher than pure binder in the air temperature range from 20°C to 50°C . A higher heat capacity can help regulate temperature fluctuations by reducing the maximum surface temperature and increasing the minimum surface temperature (Hu et al., 2015). Additionally, the thermal conductivity and thermal diffusivity are lower by 5–35 % and 3–45 %, respectively. (Hu et al., 2014), indicating that it takes longer to heat or cool these materials compared to conventional asphalt pavements.

The specific heat capacity and thermal conductivity properties were modeled as a linear function with different angular coefficients for low temperatures (-20°C to 20°C) and intermediate temperatures (20°C to 50°C) (Hu et al., 2014). These empirical models can be used to forecast the specific heat capacity and thermal conductivity of asphalt binders under conditions that include low and intermediate pavement service temperatures, as well as scenarios with low powder concentrations. However, further research is needed to develop models for high temperatures and higher concentrations of thermochromic pavements. This is a relevant issue from the perspective of climate change, as ambient temperatures and pavement surface temperatures are increasing.

As a great significance to the improvement of urban microclimate and the sustainable development of city, Table 3 summarize the main outcomes of using thermochromic materials in asphalt binders and

Table 4

Main cooling effect results of the use of thermochromic material in asphalt binders and mixtures.

Ref.	Cooling effect	Study method	Gaps
(Hu and Yu, 2013)	Surface temperature reduction of as much as 6.6 °C during summer	Experimental outdoor (surface temperatures were recorded for 24 h during a typical hot summer day in Cleveland, Ohio)	Possible reductions in high temperature-related performance degradation
(Hu and Yu, 2016)	Surface temperature reduction of as much as 6.6 °C during summer	Outdoor temperature was measured experimentally at 60-second intervals.	Improvements in the temperature meter instrument in the range of temperature control
(Hu and Yu, 2020)	TCM HMA keeps 5–15 °C cooler during daytime and 1 °C warmer during nighttime in summer	Outdoor temperature test (24 h on a typical hot day in Cleveland, OH.)	Design optimization of TCM asphalt mixture by conducting field evaluation during both summer and winter seasons
(Li et al., 2020)	The average temperature reductions in the whole top asphalt layer (0 ~ 4 cm) are approximately 2, 4, and 5 °C for TCM additive contents of 4 %, 6 %, and 8 %, respectively	Laboratory solar radiation simulation (typical summer day in Nanjing – China, from 5:30 to 18:30)	Comparison with outdoor experiments
(Chen et al., 2021)	TCM asphalt mixture prepared by WMA technology can achieve 2 °C decrease in pavement temperature compared with pure asphalt mixture.	Outdoor solar radiation in hot summer (from 9:00 a.m. to 5:00p.m. on a summer day in the city of Changsha-China)	The durability of TCM asphalt materials prepared by WMA technology by indoor accelerated long-term aging or long-term outdoor weathering aging test / the effects of mechanical blending and aggregates movement, during mixing and paving procedures, on the integrity of microcapsules
(Zhang et al., 2022)	Under experimental indoor high temperatures, the cooling effect of TCM coating on road surfaces could reach 10.57 °C; The CFD simulation results lead to a maximum 1.5 m air temperature drop of 1.5 °C.	Indoor solar radiation simulation experiment / CFD simulation method applied to a typical extremely hot summer day in Shanghai	The combination of TCM and suitable phase change materials for improvement in the heating effect of TCM coatings / to explore the effect of TCM coating on the improvement of urban microclimate considering using it in more areas and a wider variety of urban surfaces (roofs and walls) as well as in combination with greening.
(Chen et al., 2022)	Maximum decrease of 4–7 °C in pavement surface temperature during the daytime	Outdoor experiments, 72 h during summertime in the city of Changsha, China	Influence of wet/dry process on the cooling function sustainability of thermochromic microcapsule modified road materials
(Zhao et al., 2023)	During the day, the internal temperature was greater than the outside temperature, and in the early afternoon, the discrepancy between the two temperatures reached about 30 °C to 7 %TCM + 25 % Low freezing point materials.	The temperature data inside different asphalt mixtures exposed to outdoor solar radiation from 8:00–19:00 in hot summer	Investigation of different asphalt mixture types and long-term performance evaluation
(Xie et al., 2023)	The UTWC with thermochromic pellets exhibits a maximum temperature reduction of 5.5 °C	Indoor radiation solar simulation with infrared light from a Tungsten iodide lamp (500 W).	Investigation of road safety performance (skid resistance)
(Chen et al., 2024)	The average temperature reduction of TCM compared with the pure asphalt pavement material in different regions is 6.26 °C	Outdoor solar radiation test for successive three days in hot summer and numerical simulation for outdoor solar radiation test (heat transfer theory of asphalt pavement)	The influence of the cooling function of TCM on the temperature and modulus distribution inside pavement structure, as well as the pavement mechanical response and service life

mixtures, regarding optical and thermal properties. This article focuses on the results of applying thermochromic asphalt in hot weather conditions.

Regarding to promotion a cooling effect, previous studies have exposed thermochromic binder to open spaces during hot summer days in Cleveland, OH, US. At temperature ranges between 20–70 °C, the thermochromic binders consistently exhibited lower surface temperatures than the conventional asphalt binder. The maximum reduction in surface temperature was 6.6 °C, 2.7 °C, and 4.9 °C for the black, blue, and red asphalt coatings, respectively (Hu and Yu, 2013; Hu et al., 2015). The laboratory tests have shown that the addition of thermochromic additives to HMA mixtures with binder modification results in a linear reduction of the maximum temperature. The average temperature reductions in the top asphalt layer (0 ~ 4 cm) are approximately 2, 4, and 5 °C for 4 %, 6 %, and 8 % of thermochromic additive content, respectively (Li et al., 2020). During experimental outdoor tests, the maximum temperature reduction in HMA depths showed absolute errors of less than 3 °C between laboratory and field results, satisfying the accuracy requirements of engineering practice (Li et al., 2020). Zhang et al. achieved better results for the cooling effect in indoor experiments by covering the HMA with thermochromic coating, resulting in a surface temperature 10.5 °C lower than the conventional HMA (Zhang et al., 2022). Furthermore, the study found that the cooling effect of TCM coating was 2–5 degrees Celsius lower than that of cool coating with the same color (Zhang et al., 2022), suggesting that thermochromic coating is more effective among reflective pavements.

Although the initial results are promising, the studies remain in the early stages. One major limitation is the duration of field trials, which

typically range from just days to a few months, and are often conducted in summer. Long-term environmental exposure is crucial for understanding the behavior of thermochromic asphalt pavements over the course of annual weather variations. Moreover, no studies have yet examined the application of thermochromic asphalt pavements on a full-scale, presenting a valuable opportunity for further scientific investigation with practical implications. In laboratory tests, incandescent lamps are used to simulate the effects of solar irradiation in controlled settings. However, this approach neglects the influence of other environmental factors, such as wind, cloud cover, humidity, and air pollution, which may lead to discrepancies between field and laboratory results.

On the other hand, computer simulations have been employed to estimate and to evaluate the cooling effects of thermochromic asphalt applications. Chen et al. found promising results using an interpolation methodology to match numerical simulation with experimentally monitored specimen temperature data. In summer conditions, a cooling effect of 6.26 °C was obtained when the thermochromic surface of the asphalt mixture had an albedo six times higher than the base asphalt (Chen et al., 2024). Zhang et al. (Zhang et al., 2022) utilized Computational Fluid Dynamics (CFD) simulations to evaluate the impact of replacing all traditional asphalt pavements in a typical residential area of Shanghai, China, with thermochromic pavement. The results, modeled for an extremely hot summer day, indicated a maximum reduction of 1.5 °C in air temperature measured at a height of 1.5 m. The use of simulations to study thermochromic pavements remains under-explored in the current literature, particularly in terms of quantifying their potential to reduce UHI effects on human health, well-being, air

pollution, and smog formation. Additionally, the relationship between the cooling effects of thermochromic pavements and energy consumption in urban centers represents a significant knowledge gap. It's important to note that the relationship between environmental impact and cost is highly complex due to the presence of several indirect benefits, and because environmental costs are not easily quantifiable. Addressing these areas could offer valuable insights for urban planning and environmental enhancement.

The use of additives has been also explored to assess their combined effects with thermochromic materials. Zhao et al. (Zhao et al., 2023) found that incorporating 7 % by weight of thermochromic powder and 25 % of low freezing point materials in HMA specimens reduced surface temperature by 4–9 °C. This was due to the internal differential temperature caused by the additives, which lowered the internal temperature of the modified asphalt mixture compared to the unmodified specimen. This delay in high-temperature distress of asphalt pavement has important implications for pavement durability and performance (Zhao et al., 2023). However, given the early stage of thermochromic asphalt research, several promising areas warrant further investigation. These include exploring the integration of thermochromic capabilities with environmentally friendly materials, such as phase change materials (PCM) and reclaimed asphalt pavement (RAP), among others. PCM is already well-established in asphalt materials applications due to its broad phase change temperature range and substantial enthalpy during transitions (Pinheiro et al., 2023), which can enhance cooling effects and help mitigate UHI impacts. Additionally, combining asphalt pavements recycling techniques with cooling strategies could represent an innovative approach to improving sustainability.

The potential of thermochromic cooling performance is also affected by the production process. This is relevant when choosing strategies to efficiently mitigate UHI. For instance, the use of WMA technology to prepare thermochromic asphalt mixture can reduce pavement temperature by 2 °C compared to traditional thermochromic HMA mixture (Chen et al., 2021). The asphalt mixture modified with thermochromic microcapsules, prepared using the wet process, exhibits a superior cooling effect compared to those prepared using three dry processes, with a difference of at least 2 °C (Chen et al., 2022).

To advance in this area, the underlying theory of pavement design for thermochromic pavements must be thoroughly understood and requires further investigation. Table 4 summarizes the key findings on the cooling effects from the thermochromic studies discussed here, detailing the methods used and highlighting the main scientific gaps.

3.2.3. Mechanical performance of thermochromic asphalt mixtures

The use of thermochromic materials in pavements has been shown to have an impact on their mechanical performance mainly due to the potential effects of these materials on temperature control (Hu and Yu, 2020; Li et al., 2020; Zhang et al., 2017; Chen et al., 2019; Li et al., 2018; Du et al., 2019; Chen et al., 2022; Hu et al., 2015; Li et al., 2020; Chen et al., 2021; Zhang et al., 2020; Chen et al., 2021; Li et al., 2018; Li et al., 2020). Therefore, it is relevant to consider these factors when incorporating thermochromic materials into pavement design.

Experimental results on the physical and rheological properties of asphalt binders with the addition of thermochromic materials indicate that the softening point, viscosity, complex modulus, rutting resistance, fatigue resistance, and stiffness of the asphalt binder increase with the addition of the thermochromic additive. Conversely, the penetration, phase angle, and creep rate decrease (Hu and Yu, 2016; Hu et al., 2015). The addition of 3–10 % black powder can improve the high temperature of performance grade (PG) by 8.6–15.8 % (Hu and Yu, 2016; Hu et al., 2015). Furthermore, increasing the thermochromic powder content can enhance the high-temperature stability of asphalt (Hu and Yu, 2016; Hu et al., 2015) and its resistance to weathering and aging (Hu and Yu, 2016; Chen et al., 2019). Thermochromic asphalt binders are greatly less influenced by sunlight than base asphalt binders, which is attributed to the best solar radiation reflection that protects the asphalt matrix from

sunlight influence, leading to higher weathering aging resistance of thermochromic asphalt binder (Chen et al., 2019). The study also suggests that the weathering aging behavior of thermochromic asphalt binders is influenced by both the characteristics of the asphalt matrix and the thermochromic powder.

This suggests that thermochromic asphalt binders may be a resilient alternative for use in cities with hot climates. Given the increasing frequency of climate change-induced extreme events, such as heat waves, the importance of pavements with enhanced thermal stability is growing. This increased stability can directly impact the durability of asphalt materials, reducing the frequency of maintenance and repairs needed for pavements. This represents a significant knowledge gap in applying thermochromic materials in paving. More broadly, conducting a life cycle analysis of thermochromic asphalt materials remains an open challenge in the current literature.

Controversially, studies have shown that the use of thermochromic materials in SBS-modified asphalt binders can slightly weaken their high-temperature performance compared to pure asphalt binders (Li et al., 2018; Li et al., 2018; Li et al., 2020). Specifically, the addition of 4 % TCM to SBS-modified binders resulted in a change in PG from 82.6 to 24.7 (pure SBS-modified binder) to 81.6–38, reflecting a decrease in high-temperature performance and an improvement in low-temperature performance. Additionally, adding 2 % TCM to SBS-modified asphalt binders enhances their low-temperature crack resistance by approximately 24 %, their resistance to natural aging by over 5 % (Du et al., 2019; Li et al., 2020), and their deformability by up to 22 % (Li et al., 2018).

Despite research into the combined effects of thermochromic materials and asphalt binders, there has been limited analysis of the impact of TCM nanomaterials on long-term pavement aging performance. Long-term aging, caused by prolonged exposure to high temperatures and continuous oxidation, can increase the susceptibility of HMA to intermediate-temperature cracking (Wu et al., 2017). Thus, performance-based approaches for the design and selection of materials that resist cracking are crucial. Furthermore, the incorporation of additives that reduce cracking potential remains an area of significant interest.

The semicircular bending (SCB) test on notched samples has attracted significant attention from pavement researchers for assessing the fracture resistance and cracking parameters of thermochromic HMA. Li et al. (Li et al., 2020) observed that TCM-modified asphalt mixtures exhibited an increase in failure strain and a decrease in tensile strength as the additive content increased during SCB tests conducted at –10 °C to evaluate low-temperature cracking resistance. These TCM-modified mixtures absorbed more solar heat by changing color at low temperatures, which softened the binders and improved low-temperature performance in conditions exposed to sunlight. However, while the study found statistically significant effects of TCM additives, with smaller particle sizes having a greater stiffening effect on asphalt binders, the overall impact of TCM additives was minimal when the same base binder was used (Li et al., 2020).

Li et al. also investigated the effects of two different thermochromic particle sizes on SBS-asphalt binders. The study found adding powder with small particle size can improve the rutting resistance as it hardens the pure binders and ensures a better cooling effect (Li et al., 2020). However, the complex modulus of the asphalt decreases with the addition of powder, likely due to the thermochromic powder negatively affecting the rutting resistance (Zhang et al., 2020). The mechanical performance of asphalt mixture is influenced by the modification process used. Both dry and wet mixing method showed similar moisture susceptibility (Chen et al., 2022) and an increase of 30 % in the dynamic modulus when compared to the base samples. In this context, the complexity of the impacts of adding nanomaterials to the mechanical performance of asphalt materials becomes evident, highlighting the importance of optimizing the percentage of thermochromic additives to balance both mechanical behavior and cooling effects.

Table 5

Main mechanical performance results of use of thermochromic material in asphalt binders and mixtures.

Ref.	Technological Details	High-Temperature Performance / Rutting Resistance	Low-Temperature Performance / Cracking Resistance	Water Stability	Aging Analysis
(Chen et al., 2021)	Asphalt binder incorporating 6 %TCM by weight and 4 % Sasobit®, wet mixing	The larger complex modulus of modified binder indicates Sasobit can improve its high-temperature stability	–	–	The WMA technology can mitigate the adverse effects of short-term aging on functionality of modified asphalt binder
(Chen et al., 2021)	Asphalt binder incorporating 6 %TCM by weight, wet mixing	Modified asphalt binder after 20 Day of UV exposition decreased rutting resistance due to leakage of methyl stearate	DSRFn of the control asphalt binder is 3.25 higher than modified asphalt binder and, thus, more susceptible of cracking	–	Modified asphalt binder can slow down the oxidation of the bitumen matrix during long-term aging condition
(Li et al., 2018)	SBS-modified asphalt binder incorporating 2, 4, 6 and 8 %TCM by weight, wet mixing	TCM slightly affected the high-temperature stability (from PG 82.6–24.7 to PG 81.6–38 with 4 % TCM)	Analysis of BBR test indicates adding 4 % TCM of modified asphalt binder represented the best low-temperature performance	–	Considering short- and long-term aging by TFOT test, 2 % and 4 % red TCM asphalt binders showed superior aging resistance
(Li et al., 2020)	SBS-modified asphalt binder incorporating 4, 6, and 8 % TCM by weight, wet mixing	The small TCM particles (1–10 µm) cause hardening and further improve rutting resistance by URLPD test	The addition of TM additive has little effect on the low-temperature cracking resistance of asphalt mixtures using SCB test	Higher particle sizes TCM-modified asphalt mixtures show slightly better moisture resistance than small TCM by T-IDT test.	–
(Li et al., 2020)	SBS-modified asphalt binder incorporating 2 %TCM by weight, wet mixing. Thermochromic HMA	Modified HMA presents a slightly (~5%) reduction in high-temperature performance according to results of Rutting test	Low-temperature splitting test showed the addition of TCM can improve the low-temperature performance of SBS-modified HMA	TSR results of the TCM with SBS-modified HMA is 6.8 % better than that of the control HMA	TCM can improve the antinatural aging ability of SBS-modified HMA, and 2 % red powder has the best antinatural aging effect
(Li et al., 2018)	SBS-modified asphalt binder incorporating 2 and 4 %TCM by weight, wet mixing	Ductility test showed TCM improved the deformability by up to 22 % of modified asphalt binder compared to the control asphalt binder	–	–	the anti-aging properties of modified asphalt binders are better than control binder. 2 % of black TCM showed the best aging resistance (TFOT test)
(Chen et al., 2022)	Asphalt binder incorporating TCM content of 6 % by weight, wet process; and HMA incorporating TCM content of 0.326 % by mass of the aggregates, dry process	Considering tensile strength, tensile deformation and rutting resistance, modified HMA prepared by wet process can display a better performance than dry process. Both are better than control HMA	–	Freeze-thaw ITS test showed the addition of TCM improves the adhesion between asphalt binder and aggregates for both processes, which reduces moisture susceptibility	–
(Hu et al., 2015)	Asphalt binder incorporating 3, 6 and 10 %TCM by weight, wet mixing	TCM is found to increase the Superpave high-temperature performance grades of modified asphalt binder; TCM helps to improve rutting resistance at the corresponding service temperatures in DSR test	The increase of TCM content reduces the resistance of modified asphalt binder to low temperature cracking	–	The stiffness and rutting resistance of modified asphalt binder increased after short-term aging (RTFO test).
(Zhang et al., 2017)	Asphalt binder incorporating 4 and 6 % TCM by weight, wet mixing	TCM can improve high-temperature stability of asphalt binder by increasing its softening point, viscosity, complex modulus while decreasing penetration and phase angle.	The stiffness of modified asphalt binder was lower than control asphalt binder, which could improve capacity of deformability of modified asphalt binder in low temperatures.	–	6 % red TCM represented good anti-aging performance after long-term thermal oxidation aging (PAV aging test).
(Chen et al., 2019)	Asphalt binder incorporating 6 % TCM by weight, wet mixing	Modified asphalt binder exhibits lower Jnr and higher ZSV than control asphalt binder, manifesting TCM can improve the high-temperature stability; the rutting resistance is gradually decreased until 6 months of weathering aging, but after which the rutting resistance begins to be improved	The stronger solar reflection of red TCM and heat absorption of methyl stearate mitigate thermal aging effect on cracking resistance	–	Rheological indexes of modified asphalt binder are not always increasing or decreasing with growth of weathering aging time but there is a turning point that is at the sixth month
(Hu and Yu, 2020)	Asphalt binder incorporating 6 % TCM, or 1 % TCM + 3 % TiO ₂ by weight, wet mixing; Thermochromic HMA	The addition of nano-TiO ₂ improves rutting resistance of thermochromic HMA according to APA rutting test	–	TSR results and stripping observed show the addition of nano-TiO ₂ improves the moisture resistance of thermochromic HMA	–
(Li et al., 2020)	Asphalt binder incorporating 4, 6, and 8 % TCM by weight, wet mixing	According to TFS test and MSCR test, the small TCM particles (1–10 µm) cause hardening and further improve rutting resistance	–	–	–

(continued on next page)

Table 5 (continued)

Ref.	Technological Details	High-Temperature Performance / Rutting Resistance	Low-Temperature Performance / Cracking Resistance	Water Stability	Aging Analysis
(Zhang et al., 2018)	Asphalt binder incorporating 2, 4, 6, and 8 % TCM by weight, wet mixing	—	Creep stiffness modulus indicates low-temperature properties are improved along with an increment of TCM content first and then decline, and optimum content is 6 %.	—	4 % is the optimal content in which modified asphalt binder exhibits the best aging resistance measured by FTIR analysis,
(Du et al., 2019)	SBS-modified asphalt binder incorporating 6 %TCM by weight, wet mixing	According to MSQR test results, TCM microcapsule weakens rutting resistance of base asphalt binder, and has few effects on rutting of SBS modified asphalt binder	Thermochromic microcapsule improves fatigue of base and SBS asphalt modified binder	—	Thermochromic microcapsule can also improve thermal and photo oxidation aging resistance of SBS modified asphalt binder
(Chen et al., 2024)	Asphalt binder incorporating TCM content of 6 % by weight, wet mixing; Thermochromic HMA	Considering the cooling effect, the dynamic modulus at 1.59 Hz of the modified HMA is 12.4 % higher than control HMA	The Failure Tensile Strain (FTS) of modified HMA is 30 % over the control HMA after aging.	—	Relaxation strength spectra of modified HMA do not significantly change after aging, indicates the cooling effect can mitigate the aging degree of HMA
(Zhao et al., 2023)	Asphalt binder incorporating TCM content of 3, 5 and 7 % by weight, wet mixing; + low freezing point materials replaced 25, 50, and 75 % of the mineral filler volume in HMA specimens	The content of 5 %TCM + 25 % low freezing point additive achieved the best highest dynamic stability results to modified HMA	ITS test of modified HMA increased with a 25 % of low freezing point, and the inclusion of TCM has little impact on that. The bending test of modified HMA is 2–8 % lower than control HMA.	Modified HMA freeze–thaw splitting strength fell to a maximum of 10.19 % compared to control HMA, with the increase of low freezing point materials	—

Concerning the encapsulation strategy, the same effects were achieved using microcapsules composed of trioctanoinand compared with shells made by melamine–formaldehyde resin to thermochromic asphalt mixtures (Li et al., 2020). However, the potential impact of leakage from thermochromic core components on the mechanical behavior of asphalt mixtures remains unknown. For example, methyl stearate, similar to oil released from TCM microcapsules, compensates for flexibility loss but reduces the ability of asphalt binders to resist thermal stress at low temperatures (Li et al., 2020).

To sum up, by reducing the inner asphalt temperature, the mechanical properties of certain asphalt materials can be improved. The mechanical behavior of thermochromic binders and mixtures remains a relevant research subject with many opportunities for improvement and development. There is a lack of studies on the mechanical performance of thermochromic alternatives under long-term use and weathering conditions, field application studies, as well as further numerical studies are necessary to quantify the impacts of thermochromic additives. Cost assessments including the impact on maintenance and restoration interventions and a life cycle assessment are also essential. It could be inferred that the thermochromic capability has a relevant potential to improve the mechanical conditions of asphalt pavements while mitigating UHI effects. Table 5 presents the primary findings regarding the mechanical performance of thermochromic asphalt materials.

4. Technology readiness level of thermochromic pavements' future perspectives

From the bibliometric and systematic analyses conducted throughout this work, it has been possible to ascertain that the thermochromic asphalt pavements' functionalization represents an innovative theme with significant potential for relevant scientific investigation. Certain scientific gaps have been identified as potential contributions to the field of transportation infrastructure, particularly within the paving sector.

In this context, the Technology Readiness Level (TRL) method was adapted to estimate the technological maturity level best suited for each proposed future work perspective. The nine TRL levels were grouped into three categories: laboratory research; simulation world; and real world, as illustrated in Fig. 11. It is evident that there is potential for scientific contribution at all TRL levels, which aligns with the state of the art of the thermochromic asphalt technology in question.

5. Conclusions

This document presents a literature review dedicated to thermochromic asphalt binders, mixtures, and pavements used to mitigate UHI effects. To achieve this goal, two approaches were taken: first, a bibliometric analysis was conducted to produce network maps and analyze the current literature. Second, a systematic review was performed and a state of the art was elaborated basing a discussion on the thermal, optical, and mechanical performance of thermochromic asphalt mixtures, identifying the present scientific gaps, and outlining future perspectives. The results indicate that the theme has been a popular subject of scientific research, with a growing number of publications since 2013. The analysis revealed the following key findings:

- China is the most productive country, with a relevant and well-established group of researchers. There is an initial connection among organizations, highlighted by a few numbers of partnerships between authors and scientific groups. Research papers were mostly published in the Journal of Construction and Building Materials.
- Most studies available that investigated organic powders as TCM reported that the color, content, particle size, chemical composition, and method incorporation are crucial aspects to define the behavior of asphalt functionalization. Black thermochromic powders typically exhibit the best spectral reflectance in the visible and near-infrared

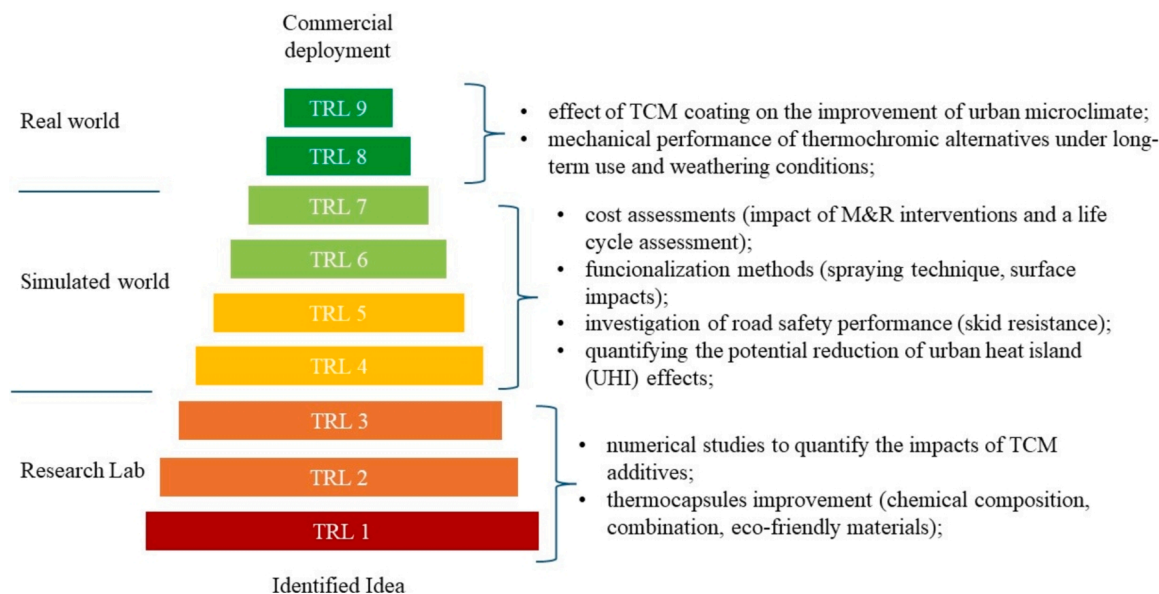


Fig. 11. Technology Readiness Levels for thermochromic pavements' future perspectives.

spectra. The optimal content of TCM for achieving optimal optical and thermal performance is 4–6 %. Smaller particle sizes improve the dispersion and intactness of thermochromic microcapsules in base binders, resulting in better physical properties and rutting resistance. The shell materials composed of trioctanoin showed less damage during heating or mixing, preventing leakage and loss of thermochromic efficiency. The incorporation of TCM using the dry process results in higher reflectance compared to the wet process, indicating that the dry process better preserves the integrity of thermochromic microcapsules. Additionally, the addition of TiO_2 to thermochromic coatings has been shown to improve optical characteristics due to its high reflectance in the NIR solar spectrum.

- In laboratory tests, temperatures dropped by up to 10 °C in terms of thermal performance. Experimental outdoor tests on a typical sunny day showed a cooling effect between 3–7 °C for the thermochromic binder and 5–15 °C for HMA. These results demonstrate that thermochromic asphalt is particularly effective in hot weather for improving thermal comfort and mitigating the UHI effect. The technology known as WMA can lower the mixing and compaction temperatures of thermochromic asphalt mixture by 16 °C. This suggests that combining sustainable strategies can be an effective method for enhancing cooling efficiency and decreasing pollutant emissions.
- The mechanical properties of thermochromic binders are improved compared to the base samples. This is especially true for the high-temperature performance grade, which reinforces the recommendation to use it in cities with hot climates. The reversibility of TCM can slow down the oxidation of the bitumen matrix, ensuring greater stability to UV aging for thermochromic asphalt binder, including SBS-modified binder. WMA can also provide benefits by reducing the short-term aging temperature of thermochromic asphalt binders. Therefore, thermochromic asphalt pavements have the potential to serve as a thermoregulation alternative, helping to mitigate temperature-related distresses, improve durability, and reduce the frequency of maintenance needs.

In summary, thermochromic technology shows to be promising in mitigating the urban heat island effect, particularly in hot weather locations, while also enhancing the mechanical performance of asphalt pavements by changing color in high temperatures. However, there are still relatively few studies on this topic, and further scientific research is needed. The methods of incorporation, particularly the spraying coating

technique, should be further investigated due to their versatility and functionalization of new and old pavements ease. Particularly, the immobilization of thermocapsules in the asphalt substrate under weathering conditions, the impact of the solutions on road safety, and the superficial characteristics of thermochromic coatings must be further studied to increase the level of readiness of this technology and give confidence on its use in field.

CRediT authorship contribution statement

Larissa Virgínia da Silva Ribas: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Iran Gomes Rocha Segundo:** Writing – review & editing, Validation. **Joaquim Carneiro:** Writing – review & editing. **Elisabete Fraga de Freitas:** Writing – review & editing, Validation, Methodology. **Verónica Teixeira Franco Castelo Branco:** Writing – review & editing, Validation, Supervision, Conceptualization.

Declaration of competing interest

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

Acknowledgments

The authors gratefully acknowledge financial support from the Brazilian agencies CNPq, CAPES, and FUNCAP, including the grant “Mulheres na Ciência: Apoio a Projetos de Pesquisa Coordenados por Mulheres” (FUNCAP N°06277098/2022). This research was also funded by the Portuguese Foundation for Science and Technology (FCT/MCTES) through national funds (PIDDAC), under the projects NanoAir PTDC/FISMAC/6606/2020 (doi.org/10.54499/PTDC/FIS-MAC/6606/2020) and UIDB/04650/2020, as well as through the R&D Unit Institute for Sustainability and Innovation in Structural Engineering (ISISE), under reference UIDB/04029/2020 (doi.org/10.54499/UIDB/04029/2020). Additional support was provided by the Associate Laboratory Advanced Production and Intelligent Systems (ARISE), under reference LA/P/0112/2020.

Data availability

No data was used for the research described in the article.

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