

Review

Materials to Mitigate the Urban Heat Island Effect for Cool Pavement: A Brief Review

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Abstract: The urban heat island (UHI) effect has a significantly negative impact on the living environment in urban areas. Asphalt pavement is one of the most widely used infrastructures that absorbs solar energy, which leads to the UHI effect and premature failure. As a result, cool pavement technology has been rapidly developed in recent years to mitigate the UHI effect originating from asphalt pavement. Although several outstanding review articles have analyzed previous studies on cool pavement technologies, very few review articles have focused on how to design and expand cool pavement technology from a materials perspective. In this mini-review article, the theoretical and practical factors of the solar reflective coatings and phase-change materials, which are significantly dependent on the design of new materials, have been summarized. The main challenges and potential problem-solving ideas have been presented. In a cool pavement, the solar reflective coatings are composed of epoxy resin or acrylic polymer matrix filled with solar reflective nanoparticles, such as TiO₂, SiO₂, ZnO, Al₂O₃, or Fe₂O₃. The main challenges of the solar reflective coatings are the spalling of the coating polymers from the asphalt pavement surface and the dispersion of the solar reflective nanoparticle in the polymer matrix. Most importantly, it is critical to harmonize the balance between the bonding strength, aging rate, solar reflectance, curing requirements, mechanical properties, and durability of the solar reflective coating. For the nanofillers, the cost of the filler materials, the balance between UV, visible light, and near-infrared reflectance and the dispersion status of the nanofillers in the polymer matrix are the primary factors that must be concerned. For the phase-change materials (PCMs), the interaction between the asphalt and the PCMs, the decomposition of the PCMs, the toxicity of the PCMs, the distribution status of the PCMs in the asphalt matrix, and the cost are the main factors that have to be considered in constructions. This review article can not only provide basic knowledge for the development of new solar reflective pavement materials but also serve as a guide for practical applications of cool pavement in the field.

Keywords: urban heat island effect; polymer; nanofiller; coating; phase-change materials



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1. Introduction

With the rapid development of the urbanization process, the urban heat island (UHI) effect has posed a severe negative impact on the living environment [1–4]. In the past few decades, local governments, academic organizations, and commercial industries have put huge efforts into developing new technologies to mitigate the UHI effect. Thermal reflective coatings, thermal insulation layers, and phase-change materials have been considered as active surface-temperature-reduction approaches to replace the traditional passive cooling methods, including water spray, pipelines under the pavement, and vegetation shade.

However, the knowledge gaps that govern these new active cooling technologies have become a major challenge to the implementation of these UHI mitigation strategies [5].

In urban areas, pavements cover about forty percent of urban land, and over eighty percent of them are asphalt surfaces [6]. In the summer season, solar energy is absorbed and stored in asphalt, resulting in a rising temperature of the pavement surface. Furthermore, with the development of transportation infrastructures, the area of asphalt pavement keeps increasing annually in urban areas, which leads to a severe UHI effect due to the asphalt pavement absorbing and storing more heat than natural surfaces, such as grassland or soil land [7,8]. Therefore, it is compelling to implement new technologies not only to mitigate the UHI effect but also to enhance the service life of asphalt pavement by reducing the rutting problem.

Over the past few decades, various technologies have been used to reduce the surface temperature of asphalt pavements. Typical strategies, including solar reflective coatings, phase-change materials, permeable pavement, urban vegetation, or proper layer design have been applied to mitigate the UHI effect. Previous research works mainly focused on three categories. The first one is investigating the heat transfer mechanisms and their impact on the UHI effect [9,10]. The second one is assessing the UHI mitigation approaches that are determined by the thermophysical properties, including albedo, emissivity, solar reflective index, thermal conductivity, specific heat capacity, and thermal diffusivity [11,12]. The third one is evaluating the absorbance and solar-energy reflection based on the used materials and treatment methods [13,14]. Although the investigation of cool pavement has been developed for decades, and the above-mentioned technologies have been applied, the key challenges of the cool pavement from the materials perspective have not been systematically summarized. For the solar reflective coating, the materials are composed of polymer matrix and nanosized solar reflective fillers [15,16]. In pavement engineering, polymer overlay must satisfy the requirements the same as a traditional pavement, including skid resistance, wear resistance, bonding strength, and mechanical properties. Therefore, only a limited type of polymers can be used as pavement-coating materials. Furthermore, their specific mechanical properties, durability, service requirements, and compatibility with the asphalt and the polymer matrix have barely been summarized in pavement engineering from a materials viewpoint. In addition to the overlay, PCM is also considered a promising approach to realize the cool pavement [17]. Although a few studies have demonstrated that PCMs have great potential to be applied in the asphalt pavement to reduce the surface temperature [18,19], the knowledge gap of the mechanism of the PCMs in asphalt has significantly confined their large-scale applications in the field.

Based on these limitations, this review article provides a thorough summary of the current approaches that can be used as effective strategies to reduce the surface temperature of asphalt pavement and thus mitigate the UHI effect. The main cooling mechanisms are summarized and the cooling effects are assessed and compared from the perspectives of solar energy reflective coatings, thermal resistance layers, and phase-change materials. Most importantly, the main challenges and future research directions are introduced as the guide for the next research steps. This review article not only sheds a light on expanding cooling pavement technologies, but also benefits the engineers who are working on pavement engineering and trying to mitigate the UHI effect.

2. Solar Reflective Coatings

The solar reflective coating may be called the first choice for the cool pavement, due to its cost-effective essence and relative ease of implementation in the field. Generally, solar reflective coatings are composed of two parts: the polymer (or the asphalt) matrix, and the solar reflective (or thermally insulative) filler [20–23]. The main challenges of the solar reflective coatings for cool pavement are the durability of the bonding between the polymer matrix and the asphalt pavement and the dispersion status of the solar reflective nanoparticle fillers in the polymer matrix.

2.1. Polymer Matrix

In the past few decades, various types of polymers have been used as coating materials on bridge decks and pavements. It was reported that over 2724 polymer overlays have been applied on bridges in the United States [24]. Since 2008, the polymer-based overlays have increased by 24.2%, while other types of overlays have reduced every year [25]. To satisfy the mechanical property requirements, the options for road coating are very limited. So far, epoxy resin and acrylic polymers are the two main types of inorganic materials that can be applied as coating materials on the surface of pavements. The molecular structure of epoxy resin has a broad group of reactive compounds of an oxirane or epoxy ring, while the structure of the acrylic polymer is the ester monomers. [26], shown in Figure 1.

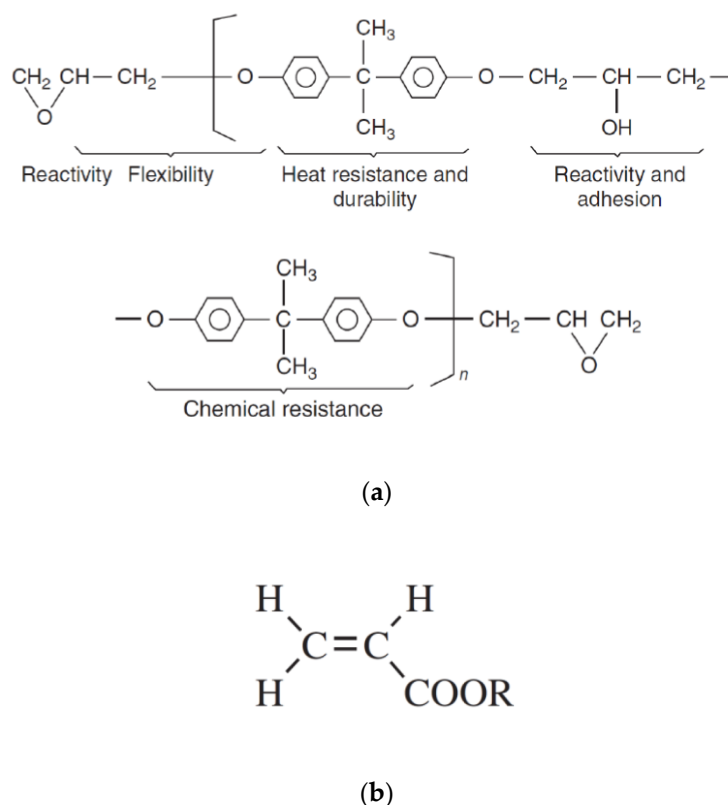


Figure 1. The structure and properties of (a) epoxy resins and (b) acrylic polymers [26].

The epoxy resin has a three-member ring structure that contains an oxygen atom bonded with two carbon atoms along with bonding other elements or groups. The universal formula of an epoxy resin is a linear polyether with epoxy groups and hydroxyl groups along the length of the chain, contributing to the chemical resistance, heat resistance, durability, and adhesion. The molecular terminals and the hydroxyl at the midpoint determine the reactivity of the molecules. The adhesion property is decided by the secondary hydroxyl groups in the molecule chain. Different from the epoxy resin, the R side-chain ester group in acrylic governs the properties of the acrylic acid polymers, and the acrylic ester monomers are formed from the reaction between acrylic acid and the alcohol, which can be expressed as acrylic acid + alcohol \rightarrow alkyl acrylate.

The materials react with the epoxy resin and form a network called the *curing agent* or *hardener*. The proportion of the curing agent varies from several percent to as high as fifty percent depending on the curing conditions. The most widely commercially available curing agents include (1) aliphatic amines and modified aliphatic amines, (2) polyamides, (3) anhydrides, and (4) polysulfides and mercaptans. The most distinctive shortcoming of the epoxy resin is its rigidity. Rigidity is the distinctive shortcoming of epoxy resins when it is used as the coating material for asphalt pavement. As a result, it is compelling to

solve this problem before it can be used in fields. By choosing a proper curing agent, the flexibility of the innate rigid epoxy resins can be changed. For example, by changing the curing agent from hexahydrophthalic anhydride to hexamineethylenediamine, the tensile elongation can be significantly increased and the impact resistance can be two times higher than the other one [26].

Two curing approaches are widely applied in commercial epoxies. The first one is heat curing for the single-component epoxy, and the second one is room-temperature or elevated-temperature curing for the multiple-component epoxy. In general, the heat curing will result in a higher glass-transition temperature and a higher crosslinking density than those cured at room temperature. Therefore, the epoxies cured at high temperatures will provide higher shear strength and chemical stability. Meanwhile, because of the rigidity, the toughness and bonding strength will be lower than those cured at room temperature. In addition to curing agents, curing time is another key factor that determines the properties of the epoxy resins. In most cases, the curing time of epoxy resin requires 18 to 72 h to obtain its full strength, and the curing time can be shortened by increasing curing temperatures. After curing, the epoxy resins are rigid materials with outstanding properties, such as low shrinkage and no volatility during the curing process, compatibility with many types of materials, good mechanical properties, chemical stability, and durability under complex environmental conditions.

Apart from the curing agents and curing time, the viscosity of the epoxy resins is another important factor that needs to be considered before it can be applied in the fields as the pavement coating. It is difficult to implement epoxy resins with a high viscosity as the coating materials on asphalt pavement. In most cases, the viscosity of the epoxy resins is governed by the molecular weight. The higher the molecular weight, the higher the viscosity and the melting point.

For the epoxy resin as coating materials in asphalt pavement, the following problems need to be identified before application. First, the inherent brittleness of cured epoxy resins easily results in cracks on the top of the asphalt pavement because of the mismatch of the deformation capability; second, the cost of the materials is relatively high if they are adjusted to the required properties; and third, the curing condition, such as relatively long curing time and high curing temperature, is critical to pavement engineering.

Unlike the epoxy resin that needs a curing agent to harden, the acrylic ester monomers will randomly incorporate each other and form polymer chains during the copolymerizing process. Furthermore, the acrylic ester monomers can also be copolymerized with methacrylic ester monomers, styrene, acrylonitrile, or vinyl acetate. The final properties of the formed polymers are determined by the molecular weight and the ester side chain of the product [27]. Provided that the side chain is H, the poly(acrylic acid) will be rigid, while the poly(methyl acrylate) is a strong and plastic material at room temperature. The poly(ethyl acrylate) with a long chain length has high plasticity. The poly(butyl acrylate) is tacky at room temperature and can be used as the adhesive. The ductility of acrylic polymers increases with increasing chain length until it reaches the threshold. At this chain length, the side chains will be crystallized and lead to the stiffening of the polymers. Acrylic ester polymers are chemically stable under aggressive conditions, which guarantees their outstanding durability. Although it is oxidation- and hydrolysis-resistant, when exposed to strong acidic or alkaline conditions, they will be hydrolyzed to poly(acrylic acid) and alcohol. The resistance to hydrolysis follows the order of butyl acrylate > ethyl acrylate > methyl acrylate.

UV resistance is another important factor that determines the durability of acrylic polymers as pavement-coating materials. In general, acrylic polymers are good at UV resistance; however, if they are incorporated with UV-absorbing monomers, such as styrene, the UV resistance of the resulting products will be considerably decreased and deteriorate rapidly. On the other hand, if the incorporating UV-absorbing monomers are bonded in a noncovalent way, the UV resistance of the resulting polymers will not be affected.

For example, hydroxy benzophenone will further enhance the UV resistivity of acrylic polymers [28].

In pavement engineering, acrylic polymers have been widely used as paint materials for pavement markings in past decades. Recently, agencies have started to use them as overlay materials for color pavements. However, research on acrylic polymers as coating materials on asphalt pavement is also very limited. Except for the relatively low adhesive or bonding strength when compared with epoxy resins, acrylic polymers have several advantages. First, the flexibility of the cured acrylic polymers is far better than the epoxy resins, which makes them capable of matching the pavement deformation during the service time; second, their chemical and UV stabilities are promising advantages of the acrylic polymers when they are used as overlay materials for pavements; and third, the relatively low cost guarantees their wide application in pavement engineering.

Epoxy resins have been used as pavement coating materials for many years. Most epoxy resin coatings are used as overlays with antiskid or waterproofing functions. Recently, epoxy resin coatings have been used to incorporate the above functions to anti-ice and to create wear-resistant surfaces. The pavement role of the solar reflective coating, including the bonding performance, the abrasion resistance, and the anti-skid resistance, is mainly determined by the polymer matrix. By using diatomaceous earth and rutile TiO_2 powder as the solar reflective fillers, and E-51 epoxy resin and polyamide curing agents as the polymer matrix, the effect of the solar reflective performance has been evaluated in both lab and field. The testing results indicated that both lab and field coatings can distinctively decrease the surface temperature of the asphalt pavement. In addition, the skid resistance and abrasion resistance of the epoxy resin-based coating has been tested. It was found that the E-51 epoxy is a promising material as the coating polymer from the perspectives of anti-skid and abrasion-resistance perspectives with appropriate surface treatment of sand [29].

Another study also claimed that the epoxy resin is an appropriate candidate as the polymer matrix for the solar reflective coating due to its good wear resistance and adhesive force. The mechanical testing results indicated that the epoxy resin, mixed with TiO_2 and SiO_2 , has a suitable viscosity and low glossiness with an outstanding cooling effect. The solar reflective testing results show that the optimized dosage of the TiO_2 , SiO_2 , and extinction powders are 12%, 4%, and 4%, respectively. The highest temperature reduction can reach 12–14 °C in the indoor environment and 7–9 °C in the solar environment by the coating-material usage of 0.8 kg/m² [30].

An experimental study demonstrated that by using epoxy resin as the matrix resin and nanotitanium dioxide (TiO_2) as the functional filler, the solar reflective coating on asphalt pavement can not only deal with high-temperature diseases but also reduce the carbon monoxide (CO) and hydrocarbon (HC) emissions from automobile exhaust. The mass proportion of the epoxy resin/diluent/curing agent/nano- $\text{TiO}_2/\text{Fe}_2\text{O}_3$ was 100/24/36/6.4/0.3. The irradiation reflectance efficiency was evaluated through the tungsten-iodine lamp. It was found that this epoxy-based coating can reduce the surface temperature of the asphalt pavement from 71.3 °C to 57.9 °C. The 3 h rutting testing results revealed that this epoxy coating has excellent wear resistance. Apart from the rutting resistance, the CO/HC decomposition effect of this epoxy coating has also been investigated. It was found that the maximum decomposition rate of the CO/HC was 19.3% at 50 °C and an ultraviolet intensity of 2770 $\mu\text{W}/\text{cm}^2$ [31].

Different from road pavement, for the sidewalk pavement, the requirements for mechanical performance are much less strict than normal highway pavement. The mechanical performances are not the key challenges of the coating but the requirements for human comfort feeling are the priority. In a recent study, a triple-layer solar reflective coating for the sidewalk has been tested. The three layers include the bottom layer, the elastic layer, and the surface layer. The authors mentioned that the resin of the surface layer should satisfy the following requirements: First, it should have high fluidity, which ensures the feasibility of the implementation in the field; second, its euphotic index should be more

than 80%; third, the contents of the C=O and C-O-C groups should be as low as possible to avoid the oxidation from the light and oxygen. Therefore, five resins, including epoxy resin (E51 and E54), methylmethacrylate, polyvinylidene difluoride, and waterborne epoxy resin were chosen as the polymer matrix, and the thickness of the coatings was 0.5–1.5 mm. In addition to the resin matrix, the solar reflective fillers, TiO_2 and SiO_2 nanoparticles, and iron oxides were used as the pigment to change the surface color of the coating. To enhance the comfort level of the pedestrians, the elastic layer was made of methylene diphenyl diisocyanate polymer, and the thickness was 2–4 mm. The bottom layer has a two-fold function, namely enhancing the bonding strength of the coating and the asphalt surface, and preventing moisture damage. The thickness of this layer is very thin and can be neglected. By comparing with the outdoor and indoor tests, the results indicate that the waterborne epoxy resin is the best choice as the surface layer in the five resin candidates from the perspectives of anti-skid performance, abrasion resistance, and environmental friendliness. For the solar reflective fillers, the optimized dosages of the TiO_2 , SiO_2 , and Fe_3O_4 were 10 wt.%, 1 wt.%, and 1 wt.%, respectively. Under this condition, the highest surface temperature reduction can reach 5.9 °C. Moreover, using quartz sand as aggregate is beneficial to improve the skid resistance without sacrificing the cooling performance of the coating [32].

2.2. Bonding Performance and Durability

For the polymer coating technology, the most important challenge is the bonding (adhesion) strength between the polymer and the asphalt surface. It determines the durability and the service life of the coating. As a consequence, it is imperative to find out whether the adhesion strength of the polymer materials can satisfy the requirements as a surface layer of the asphalt pavement.

Typical road properties of a series of thermosetting polymers, including polyester powder, modified polyester powder, polyester/epoxy powder, and modified polyester/epoxy powder, were investigated. The results demonstrated that the temperature of the pavement surface can be reduced by about 12 °C after implementing the coatings. However, the skid resistance, abrasion resistance, and adhesion strength are determined by multiple factors. First, these road properties are influenced by the application rates. It was found that when the implementation rate was in a range of 50 to 150 g/m², the adhesion strength was higher than the control one, while when the implementation rate was in a range of 150 to 200 g/m², the adhesion strength started to remain a constant. Second, the resin type is critical to determining these properties. The adhesion strength of the polyester powder coating is slightly higher than the polyester/epoxy powder coating. Other factors, such as the texturing agent, are also important points that need to be considered [33].

Both epoxy resin and acrylic polymers have their pros and cons when they are used as coating materials for pavements. *Epoxy resins* have outstanding fatigue strengths, high-temperature properties, and chemical stability. However, during the implementation process, the bonding surface must be completely cleaned. In addition, the curing time is relatively long and the curing temperature is high. Most importantly, the rigidity of the cured epoxy is the biggest challenge as the coating material for asphalt pavement. *Acrylic ester polymers* have a distinguished plasticity advantage for asphalt pavement. Meanwhile, it is chemically stable in aggressive environments. Table 1 gives the comparison of mechanical properties and durability between the epoxy resins and acrylic acid polymers.

The challenge of choosing the most appropriate polymer matrix is to figure out the balance between the bonding strength, aging rate, solar reflectance, curing requirements, mechanical properties, and durability. First, the bonding strength between the polymer and the asphalt and aggregates is the decisive factor in the service lives of the coating. The bonding strength of the polymer matrix and the asphalt pavement can be divided into two parts, namely the asphalt binder and the aggregates. Although a few studies have investigated the bonding strength between the polymer and the asphalt pavement, the difference between the asphalt binder and the aggregates has barely been discussed. Due

to the inorganic aggregates being chemically inert to the polymers, it is not hard to find out the regularity of the bonding strength between the various types of polymers and the different aggregates. Unlike the aggregates, the chemical composition of the asphalt binder is much more complicated. Many types of contents in asphalt can react with the applied polymer, and thus lead to the degradation of the bonding. It is compelling to investigate the chemical reactions between various types of polymer and the asphalt binder. The reaction mechanisms need to be elucidated on which parts in the asphalt binder will react with polymers and how to avoid premature degradation. Second, the implementation of the solar reflective coating can significantly reduce the aging of the asphalt; however, the aging problem of the polymer matrix has replaced the asphalt and become an inevitable challenge of the solar reflective coating. With the extension of the service time, the aging of the polymer will lead to cracks and peeling off, which fails the coatings. Although using solar reflective fillers can significantly retard the degradation process, and the aging process of various polymers has been investigated independently, the aging process and aging mechanism of the coating on the top of the asphalt surface have barely been discussed. Third, most previous studies have focused on exploring how to enhance the reflectance by designing the nanofillers; the reflectance of the polymers is barely compared. Fourth, the curing requirements of the polymers are one of the biggest challenges when applying them in the field. In most cases, to achieve an outstanding road performance coating, the polymer needs to be fully cured in a required condition and a curing time. However, when the polymer coatings were applied in the field, they are not allowed to satisfy the optimized curing condition due to traffic concerns. As a result, it is imperative to develop revised polymers and curing agents that can satisfy the curing conditions of the polymers as road coatings. Fifth, the mechanical properties, such as the elastic modulus, wear resistance, flexural strength, and rheological properties are imperative parameters that have to be considered. If the mechanical properties of the polymer mismatch the ones of the asphalt mixture, it will also lead to failure and can not guarantee the longevity of the coatings. Sixth, the durability of the polymer materials with exposure to extreme service conditions needs to be further investigated. For example, in a tropical area, the surface temperature of the road might be as high as 70–80 °C. At this temperature range, the service assessment will be considerably different from the room-temperature environment. In cold regions, however, the polymer might experience freeze/thaw cycles, which will also accelerate the degradation process. If the application field were located near a marine environment, it would face an attack from the salt. These factors are also challenges for the durability assessment in future studies. Last but not least, these above-mentioned requirements are not independent factors; they must be considered at the same time, which significantly increases the difficulty of the materials design.

Table 1. Comparison of the epoxy resin and the acrylic ester polymers.

	Epoxy Resin	Acrylic Ester Polymers
Curing requirements	Relatively long curing time and high curing temperature with curing agents.	Heat curing, radiation curing, and natural curing.
Mechanical properties	High bonding strength, high fatigue strengths, high-temperature properties, but mostly poor rigidity.	Might be brittle or plastic depending on the side chain groups of the ester.
Durability	Chemically stable, not UV-resistant.	Chemically stable, good UV resistance without UV-absorbing monomers such as styrene.

2.3. Fillers

Solar radiation consists of about 5% ultraviolet radiation, 46% visible radiation, and 49% near-infrared radiation. In this proportion of radiation, the percentage of UV light

mainly results in the aging of the polymer matrix, while the other two are the primary factors that contribute to the temperature rising of the pavement. Currently, the biggest challenge is that most of the nanoparticles can not satisfy the reflectance from both the near-infrared region and the visible region at the same time. For the near-infrared reflectance fillers, the most widely investigated fillers include (1) coloring agents or white pigments (such as nanosized TiO_2 , SiO_2 , ZnO , Al_2O_3 , and Fe_2O_3) coated with another non-NIR absorbing pigment [34]; (2) rare-earth manganese oxides $(\text{RE}_x\text{Mn})\text{O}_y$ (RE represents the Y, Ce, Pr, Nd, and Sm) with an IR reflectivity above 50% [35]; (3) polyolefin compositions containing a black-colored pigment such as chromium-iron oxide [36]; (4) zirconium dioxide modified by silicon dioxide [37]. For visible-light-reflectance materials, the research is burgeoning in recent years. Although the application of a cost-effective visible-light-reflecting filler is still on a lab scale, several types of nanoparticles with high visible-light reflectance have been explored in the past decade, such as doped TiO_2 or bismuth oxybromides [38].

To enhance the solar reflective performance of the coating, typical white oxides were mixed with the polymer matrix to prepare the solar reflective coatings. The widely used filler materials at the present stage are TiO_2 , SiO_2 , ZnO , Al_2O_3 , and Fe_2O_3 , and are mostly nanosized. Although the white-color filler is beneficial to enhance the solar reflective performance, it brings an inevitable glaring side effect for the pavement. Therefore, non-white fillers were also developed to test whether the solar reflective coating can be obtained without sacrificing the dark color of the pavement. In addition, with the development of materials science and technology, new types of nanomaterials with distinctive solar reflective potential have also been tested as fillers, such as thermochromic materials.

A recent study was conducted to assess the cooling effect of the solar reflective coating. In this work, the polymer matrix was epoxy resin emulsion, and the TiO_2 , SiO_2 , diatomite, and hollow glass bead were used as the solar reflective fillers. Compared with previous studies, in which the fillers were dispersed by high-speed mixing, in this case, the particles were surface-modified with the silane coupling agent (KH-550) to enhance the dispersion efficiency. The testing results indicate that this surface-modification method is quite effective to prevent the agglomeration of the solar reflective fillers. The optimized coating can reduce the surface temperature of the pavement by about 11 °C, and the outdoor testing results show that the temperature of the surface had been reduced 5 °C from 51 °C to 46 °C without sacrificing the anti-skid resistance and wear-resistance performance. Furthermore, the addition of 2% iron oxide black can be beneficial to leverage the cooling effect and the glaring problem [39]. The distinctive shortage of this study is that the 30% dosage may have exceeded the acceptable cost limit for the field application.

Recently, a series of thermochromic materials were used as fillers in hot mixing asphalt to manufacture the solar reflective pavement material. In this work, the thermochromic powders, which were leuco dye-based organic mixtures with colors of black, blue, and red, were encapsulated by trioctanoic acid. The color-change point was about 31 °C, which is the critical value for the material to change its color from dark to light when the temperature is above this value. According to the microstructure analysis and the rutting and the moisture susceptibility tests, it was found that both the thermochromic materials and the nano- TiO_2 can be used as solar reflective fillers. The results indicate that 1% red thermochromic powder and 3% nano TiO_2 show the highest rutting resistance performance, and that the 6% black thermochromic filler can not satisfy the HMA specification. The solar reflectance testing result reveals that the solar reflectance of the HMA can be enhanced by about 7% to 10% by incorporating the thermochromic materials with nano TiO_2 . The outdoor-temperature testing results demonstrate that this type of solar reflective pavement can reduce the surface temperature up to 15 °C during the daytime [40]. Similar work was implemented by using two types of thermochromic fillers in an asphalt mixture to realize the solar reflective pavement (shown in Figure 2). The color-changing temperature of the thermochromic fillers is also about 31 °C, and the thermochromic material is also the core-shell structure with the core materials composed of 5–15% bisphenol (developer), 2–10%

3-diethylamino-6-methyl-7-2, 4-xylinofluoran (leuco dye), and 50–80% of methyl stearate. The shell materials were melamine–formaldehyde resin and trioctanoinand, respectively. The main contribution of this study is the authors found that different thermochromic materials will lead to discrepant rutting resistance performance due to the chemical reaction between the asphalt binder and the thermochromic fillers [41].

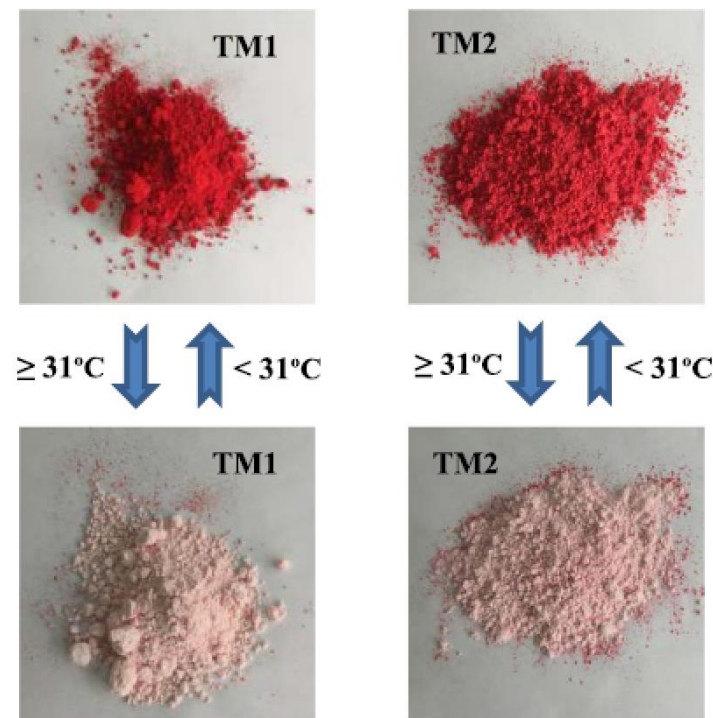


Figure 2. Thermochromic material used as additive to mitigate the heat island effect for pavement [41].

No matter whether nanosized TiO_2 , SiO_2 , or thermochromic core–shell-structured powder fillers, the conflict between the bright color and the solar reflectance seems to be the most difficult challenge that constrains the application of these fillers. As a consequence, it is imperative to explore a dark-color filler that can be used to enhance the solar reflectance but without sacrificing the dark color of the pavement surface. To find out whether black-color fillers can be used to realize this purpose, CuO nanoparticles, which have a black near-infrared reflective property, were used to enhance the solar reflective performance. In this work, CuO nanoparticles with various morphologies were used as fillers to prepare the solar reflective coatings. In this study, spherical, rod, and flake CuO nanoparticles (shown in Figure 3) with dosages of 6.8%, 12.8%, 18.0%, and 22.6% were used to find out the optimized shape of the CuO nanoparticles. The results indicate that the flake-shape CuO nanoparticles have the highest spectral reflectance for a fixed dosage. The highest temperature reduction can reach 12.6°C during the summertime, and 10.0 in the fall season [42]. Two years later, the authors combined CuO and TiO_2 nanoparticles and produced a more effective solar reflective coating than only using CuO nanoparticles. They found that combining CuO nanoparticles and TiO_2 nanoparticles is an effective way to enhance the near-infrared reflectance of the coating [43]. Although the authors claimed that the mechanism of this phenomenon is contingent upon the particle-size distribution and the microstructure of these nanoparticles, this work did not bring solid proof to stand for this statement.

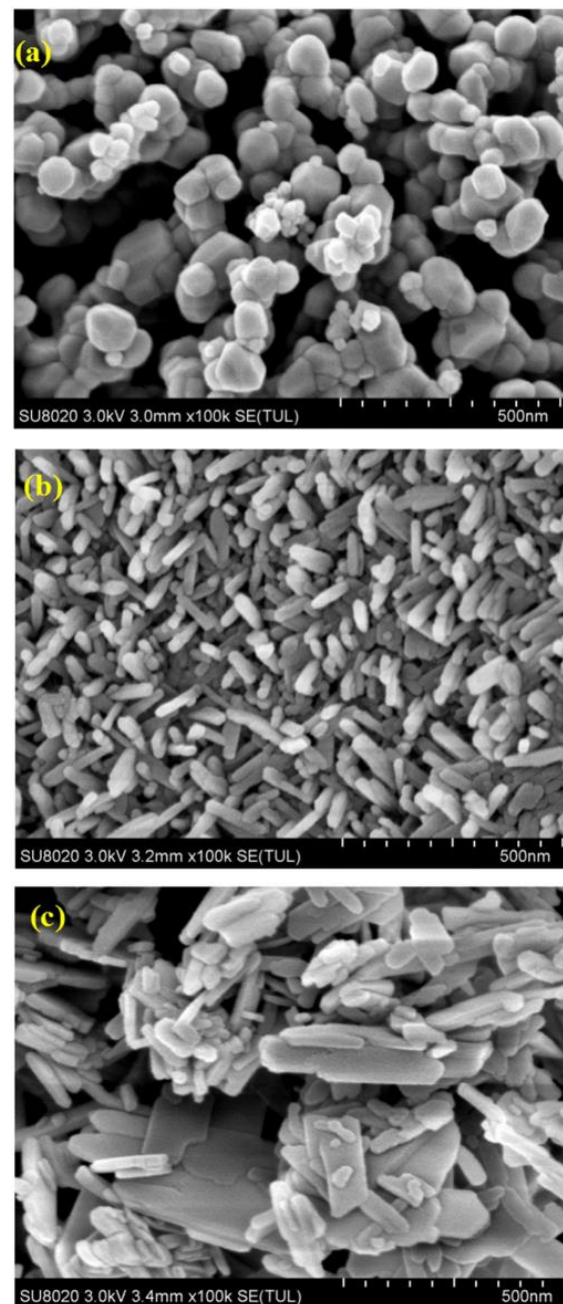


Figure 3. SEM morphologies of CuO nanoparticles with shapes of (a) spherical, (b) rod, and (c) flake [43].

Another possible solution to mitigate this conflict of bright color and reflectance is using chemical doping to change the color of the nanoparticles without sacrificing or even enhancing the reflectance of the nanoparticles. It was found that after doping with N, the solar reflectance of the TiNO_2 nanoparticles is higher than the TiO_2 nanoparticles and the color of the TiNO_2 has changed from pure white to light yellow [44]. This study has shown a pathway to solve the problem of the bright color of the coatings; however, further deep cost analysis must be followed up, as the increasing cost of the doped nanoparticles will significantly hinder their practical applications in the fields.

Apart from using the optical properties of nanosized oxides, using the thermal insulation properties of some particles as fillers is another branch to realize a cool pavement. These groups of particles mostly have high thermal-resistance properties and are thus able to achieve the goal of thermal isolation. The advantage of this type of filler is that

they are cost-effective and easy to disperse in the asphalt binder. It was reported that using cenospheres and graphite can prepare a thermal insulator material. The thermal conductivity testing results demonstrated that the optimized ratio of the binder:normal filler:cenosphere equal to 2:1:1 by weight can reach the best thermal insulation of the cool pavement, and the ratio of binder to graphite of 1:1 can improve the thermal transmission and rutting resistance. The underlying mechanism of the cenosphere's thermal insulation was elucidated by SEM images. It was found that the cenospheres have a hollow structure with an average size of about 10 μm . The cenospheres had a thermal conductivity of 0.065 W/mK which is much lower than the base asphalt with thermal conductivity of 0.175 W/K. This guarantees the thermal insulation performance of the cool pavement [45].

Similar work was carried out by replacing basalt and limestone powder with shale ceramistite and fly ash cenospheres as the aggregates and fillers in the asphalt matrix to realize the cool pavement. It was reported that the low-density shale ceramicist and fly ash cenosphere are beneficial to produce a lightweight asphalt mixture that has a lower thermal conductivity than the control asphalt mixture. Although the addition of the shale ceramistite harmed the shear strength and dynamic stability of the asphalt mixture, the temperature of the pavement can be distinctively reduced at the depth of 4 cm under the pavement surface. To mitigate the problem of the reduction in mechanical properties, the authors used fly ash cenospheres to reinforce the asphalt binder. It was found that the addition of the fly ash cenospheres is beneficial to increasing the shear strength and the dynamic stability of the asphalt binder and decreasing the surface temperature of the pavement [46].

Unlike the organic polymer matrix, which needs to face the challenges of bonding strength, aging, and durability problems directly, the filler materials are mostly inorganic oxides. As a result, the main challenges of the inorganic fillers are distinctively different from their polymer matrix. To develop new inorganic solar reflective fillers, there are several factors that need to be considered. First, the cost of the filler materials must be acceptable for practical application. Although there are many new types of nanomaterials with outstanding solar reflective performance, the complicated manufacturing process or using expensive rare-earth elements or noble metals significantly increases the cost of these materials. As a consequence, these nanofillers are not suitable to be used as fillers on cool pavement. Based on our understanding, first, the main research directions of the nanofillers should be focused on doping cheap elements (C, N, or S) in the current widely used oxides (TiO_2 , ZnO , Fe_2O_3 , or CuO) to enhance the reflected efficiency. Furthermore, the doping technology needs to be facile, environmentally friendly, and cost-effective, such as calcination, or sol-gel methods. Second, the balance between UV, visible light, and near-infrared reflectance must be leveraged. So far, most of the nanofillers can not reflect the radiation from all these three ranges. Third, the crystalline structures and morphologies of the nanofillers are also important parameters that govern the solar reflective performance of the cool pavement. Although the crystalline structure of oxides is limited, their morphologies of them can be infinite. It will be beneficial to find out the regularity of the morphologies on the reflective performance, especially after being dispersed in the polymer matrix. Last, the dispersion of these nanofillers in the polymer matrix is always the most important challenge that limits the reflectance efficiency. Due to the viscosity of the polymer matrix, it is hard to realize a good dispersion condition of the nanofillers in the polymer matrix by using simply mixing them. It is necessary to find out effective methods or materials that can be used to help the dispersion of nanoparticles in the polymer matrix, such as polarized solutions.

3. Aggregates

There are several types of solid wastes with high porosity, including furnace slag or various types of tiles that can be served as the aggregates to produce the cool pavement. It was pointed out that using furnace slag to replace normal aggregates can significantly enhance the emissivity of the pavement. As the replacement of the furnace slag is higher than 55%, the samples showed a better cooling effect compared with the normal aggregates.

It was found that the furnace slag can not only mitigate the UHI effect but also increase the mechanical properties of the cool pavement [47].

A typical solid waste material, which can be used as the solar reflective aggregate in a cool pavement, is the waste tile. The application of waste tiles can achieve the goal of near-infrared reflectance. By comparing the full-body porcelain, monoporosa, and porcelain glaze, it was found that the NIR reflectance value of 0.51 can be achieved by using 50% of full-body porcelain and 50% of monoporosa tiles. If using 100% of monoporosa tile, the NIR reflectance can reach 0.53, and the surface temperature of the asphalt pavement can be reduced to a range of 4.1 °C to 9.6 °C, whereas using 100% porcelain glaze has less of a temperature-reduction effect (only 4.2 °C) compared with the other two materials [48].

Waste ceramics have great potential to be considered as aggregates to manufacture cool pavement. The rutting resistance, moisture susceptibility, bending beam, fatigue beam, and indoor thermal insulation performance were investigated to assess the feasibility of being the cooling aggregates. The results indicated that although the rutting resistance, moisture susceptibility, and anticracking resistance were all slightly reduced, the general performance of the pavement still satisfies the requirements when the content of the waste ceramics is less than 40%. More importantly, the waste ceramics show their palpable enhancement of the cooling effect on the temperature reduction above 11 °C at the bottom of the pavement. This leads to the alleviation of the permanent rutting deformation of the asphalt pavement by at least 45% compared with the traditional asphalt pavement [49].

4. Phase-Change Materials (PCM)

Phase-change material is a relatively novel branch in building cool pavement to mitigate the UHI effect. The underlying mechanism of this system is based on the intrinsic property of these materials that they can absorb and release energy through the phase-change process. Typical PCMs that were used to manufacture cool pavement can be either organic or inorganic. The inorganic PCMs are mostly salt hydrates and low-fusion metals and alloys. The most distinctive advantage of the inorganic PCMs is the low cost and ease to obtain them. In general, the inorganic PCMs have a high latent heat capacity and high thermal conductivity, which are the most important parameters of PCMs. However, the shortcoming of the inorganic PCMs is that they are not able to realize supercooling. Compared with the inorganic PCMs, organic PCMs are mainly divided into two groups, namely paraffin and non-paraffin. The paraffin compounds are mainly hydrocarbons and the phase-change temperature is significantly dependent on the length of the hydrocarbon chain. The non-paraffin compounds are mostly fatty acids, alcohols, esters, and glycols. The benefits of the organic PCMs are the relatively high latent heat capacity and the capacity of freezing without supercooling. These are considered the primary advantages of a stable system. Of course, the inorganic PCMs have their downsides, including high cost, use of nonrenewable resources, and flammability, which severely constrains their applications in construction due to safety concerns. Previous studies have already demonstrated that the phase-change materials can be used to effectively decrease the surface temperature of the asphalt pavement [50]; however, there are still a few challenging barriers that need to be overcome. For example, the high-temperature leakage phenomenon is one of the inevitable side effects of road performance when the PCMs were applied to an asphalt mixture. In addition, the decomposition of the paraffin-based materials is still a big challenge that confines their durability in the cool pavement [51].

Paraffin is generally a hydrocarbon compound that has a straight chain with the general formula of C_nH_{2n+2} and is named *n*-alkanes or alkane. When the carbon number of *n* is higher than 17, the paraffin will be solid (also called paraffin wax), which means that the melting point of the paraffin wax is also contingent upon the carbon numbers and varies from 45 °C to 75 °C. The application of paraffin-based PCMs in the asphalt mixture has been investigated for a decade. To the best of our knowledge, the paraffin-based PCMs were either mixed with an asphalt binder through the microencapsulation method or saturated in porous aggregates. By soaking lightweight porous aggregates in a paraffin wax with a

phase-change temperature of 6 °C for 6 h, the PCM lightweight aggregates were prepared and partially replaced normal aggregates to manufacture the cool pavement. In this case, the lightweight aggregates had up to 10 weight percent of aggregates and paraffin wax concentration of 1.25% and 2.5% by the mass of aggregates. After calculation and testing, it was estimated that the absorbed PCM in the aggregates was about 13.3 wt.%. The results indicated that the addition of the PCM is effective to control the surface temperature of the asphalt mixture [52].

Polyethylene glycol-polyacrylamide graft copolymer is a typical non-paraffin organic PCM that can be used on cool pavement. In Cheng's study, polyethylene glycol (PEG with a molecular weight of 800) was used to absorb and release energy, and the polyacrylamide (PAM), which was used to maintain the shape stability of the PCM, was produced by polymerization of acrylamide under the catalysis of potassium persulfate. The manufacturing process is shown in Figure 4. In this manufacturing process, the acrylamide and potassium persulfate were firstly mixed with a ratio of 10:1 in deionized water at 40 °C, followed by adding polyethylene glycol 800 into the solution with a ratio of 3:2 and increasing the temperature to 80 °C and holding for 5 h. Finally, the PCM was extracted from the aqueous solution to obtain the solid-state PCM. The testing results indicated that adding the PCM to the asphalt mixture can not only improve the insulation capacity, but also enhance the Marshall stability, flexural-tensile strain, and moisture resistance of the asphalt mixture. It was found that the optimized dosage of the PCMs in the asphalt mixture should be 7.5% to 10.0% to guarantee road performance [53].

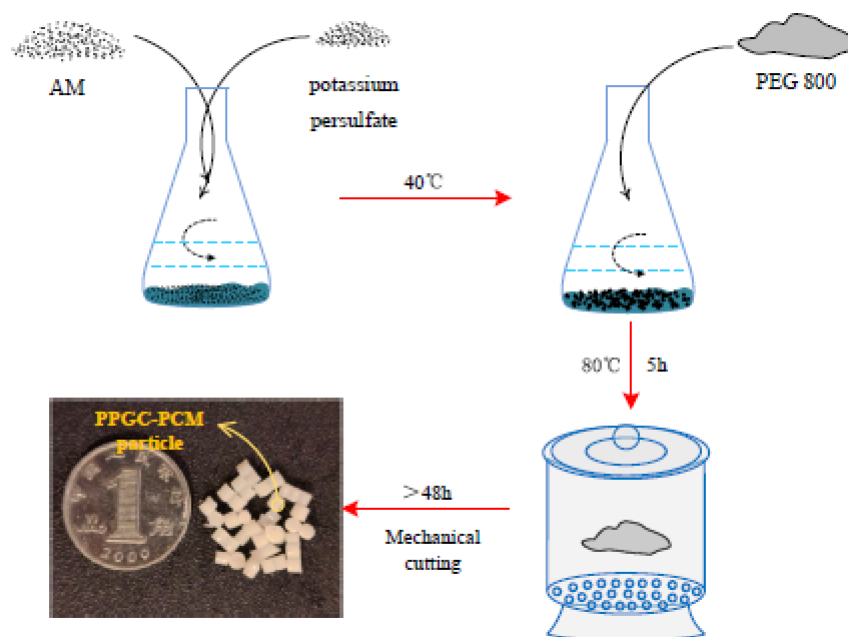


Figure 4. The preparation process of polyethylene glycol-polyacrylamide graft copolymer for cool pavement [53].

Another type of non-paraffin PCM that can be used to reduce the surface temperature of the asphalt pavement is organic acids. In Chen's study, two types of organic acids were supported by porous graphite to form the shape-stabilized PCMs. In this study, the phase-change temperature and the latent heat of these two acids are 45 °C, 50 °C, and 110 J/g and 100 J/g, respectively, and the mass ratio of the PCMs is 5 wt.% of the aggregates. The temperature increasing rate of the samples with the addition of PCMs shows a lower increase rate compared with the samples without the addition of PCMs [54].

In addition to the materials' perspective, the structure of PCM is another key factor that determines road performance. Previous studies have found that one of the most widely used branches of the PCMs is micro- or macrocapsules, which were prepared via wrapping the phase-change materials as the cores by a chemically inert shell to form a core–

shell microencapsulate structure. By using n-tetradecane as the PCM core, ethylcellulose functioned as the membrane, and ethyl alcohol as the solvent was used to manufacture the shell material. In this manufacturing process, silica and n-tetradecane were mixed to ensure the absorption of the n-tetradecane by the silica. Meanwhile, the ethylcellulose was dissolved by anhydrous ethanol to obtain the gel solution. After that, the prepared composed silica and n-tetradecane were mixed with the gel and followed by a drying process to obtain the PCM microcapsules. The results indicated that the micro-PCMs have a distinctive thermoregulation effect with a dosage of 0.5% weight percent of the asphalt binder. The highest temperature difference could reach above 10 °C [55].

Another typical structure is using porous material as a carrier to hold the PCM, which is called shape-stabilized PCM. It was found that using polyethylene glycol (PEG) as the PCM and the diatomite, expanded perlite, and expanded vermiculite as the porous holders is a promising method to manufacture the shape-stabilized PCMs. In this case, the highest PCM absorption rates for diatomite, expanded perlite, and expanded vermiculite could reach 72%, 67%, and 73.6%, respectively, by using the vacuum impregnation method. The thermal and chemical stability of the PEG/EP is higher than the other two after 100 times of heating-cooling cycles. Meanwhile, the latent heat of the PEG/EP compound is slightly higher than the PEG/DI and PEG/EVM compounds. By replacing 50% fine aggregates with these PCMs, the surface temperature of the asphalt mixture can be reduced by up to 7.0 °C in the laboratory and 4.3 °C in the field [56]. Several years later, the authors developed a new series of compound PCMs which are composed of diatomite, expanded perlite, and stearic acid (SA) as the raw materials to prepare the compound PCMs. Similarly, by using the vacuum impregnation method, the mixture of SA and minerals was infiltrated in the porous diatomite and expanded perlite, forming the compound PCMs as the replacement of fine aggregates with the diatomite size of 0.3–0.6 mm, and the expanded perlite size of 0.15–0.3 mm, respectively. The results reveal that the maximum temperature difference can reach about 11 °C on the upper surface and 5 °C on the lower surface of the asphalt mixture. It was found that the thermal storage and release rate of SA/diatomite was considerably lower than that of the SA/expanded perlite [57].

In the past decade, new types of PCM have been extensively explored. This is beneficial to expanding the application of PCM in pavement engineering. Before the practical applications, several important factors must be systematically investigated from both academic and practical perspectives. First, the interaction between the asphalt and the PCMs is the key factor that determines the durability of the PCMs in the asphalt. Although using microencapsulation or porous aggregates can significantly enhance the service lives of the PCMs in the asphalt, the long-term interactions between the PCM and the capsulation materials, the capsulation materials, and the asphalt, and the diffusion phenomenon of the PCMs in the asphalt matrix have barely been discussed. Second, the decomposition of the PCMs is still the biggest challenge that confines the application of the PCMs in asphalt pavement. Third, the toxicity of the PCMs needs to be carefully investigated, including the PCMs and the microencapsulation materials. Fourth, the distribution status of the PCMs in the asphalt matrix is another critical factor that has to be considered. From the microstructure characterization perspective, it is a hard job to identify whether the PCM materials have been well-dispersed in the asphalt matrix, and thus new devices, new technologies, or new assessment methods need to be further systematically investigated. Finally, the cost is the biggest challenge that limits the applications of PCM in constructions, even by using the cheapest paraffin-based PCMs, it is hard for the constructions to use them on a large scale.

5. Concluding Remarks

Although there are still several methods, including permeable pavement, physical approaches (water spray or underground pipelines), energy-harvesting roads, and urban vegetation, that can be used to mitigate the UHI effect, the main challenges of them are determined by the device design or the field implementation rather than the materials

perspective. This mini-review article focuses on the material design and development perspectives and tries to give a guide on how to develop new materials for the cool pavement and thus mitigate the UHI effect. The main challenges of the polymers (as the overlay coating matrix), the nano oxides (as the solar reflective fillers), the aggregates, and the phase-change materials are summarized. The guide the future design and research directions are generally provided. This review article can be utilized as a roadmap to explore environmentally friendly, cost-effective, and high-solar-reflective cool pavement materials from both academic research and practical application viewpoints.

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