

Review

A Review of Near-Infrared Reflective Nanopigments: Aesthetic and Cooling Properties

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Abstract: Excessive use of conventional cooling devices, such as air conditioners, produces an increase in the urban heat island phenomenon, which causes exacerbating climate change and environmental degradation. In response, this review focuses on the potential of near-infrared nanopigments and specifically cool nanopigments as a sustainable alternative for cooling. These innovative materials have been shown to effectively reflect solar near-infrared radiation, reducing the urban heat island effect and mitigating the environmental impacts associated with conventional cooling methods. This comprehensive review explores the aesthetic and cooling aspects of near-infrared nanopigments, highlighting their properties, applications, and benefits as a promising solution for mitigating the urban heat island phenomenon and promoting a more sustainable future. Recent breakthroughs in the use of nanopigment materials are also explored.

Keywords: urban heat island; near-infrared radiation pigments; cool-colored nanopigments; cool coating; cool pavement paint; diffuse reflection; eco-friendly materials



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1. Introduction

The UN's intergovernmental panel on climate change recommended that exceeding the 1.5 °C threshold causes huge risks for climate change such as recurrent and severe droughts, heat waves, and rainfall. Consequently, one of the Paris Agreement goals is to reduce global warming to 1.5 °C. This desired goal was planned to be achieved by reaching the peak of greenhouse gas (GHG) emissions by 2025 and dropping to 43% by 2030. Although, of that, the world is going in a deviated path away from such a target due to the substantial increase in the emission of the GHG [1]. The urban heat island (UHI) phenomenon is considered one of the main factors that increases global warming. UHI is the phenomena of a variance in the temperature inside urban regions and their surrounding rural areas. Figure 1 outlines a variety of factors that led to this uncommon circumstance. This schematic picture depicts the primary causes of the UHI phenomenon. The emission of CO₂ reached its high level in the city centers due to the enormous human gathering. As a result of storing heat by CO₂, the atmospheric temperature is increasing, which contributes to the growing UHI phenomenon [2]. The trapped heat due to air pollution is one of the most significant drawbacks of urban development. In this respect, the exhaust gases from vehicles and industrial pollutants are facilitating the trapping of solar radiation that is leading to the increase in the microclimate effect [2,3]. Furthermore, the urban canopy created by stacked buildings contributes to the trapping of heat. The reflected heat from a building is stuck as a result of multi-layer taller buildings [4]. Also, the heat is trapped due to wind blocking by densely built buildings, which makes air movement

limited, and the cooling effect by convection is reduced [5]. One of the primary causes of UHI is the lack of vegetation due to the excessive replacement of natural vegetation with buildings and paved surfaces [6]. One cannot rule out the adverse impact on the growth of the UHI phenomenon due to the excessive usage of air conditioners as well. The atmospheric temperature increases as a result of the exhaust heat from air conditioning (AC) systems. The features of the materials used in the construction of buildings and roads have a significant effect on the atmospheric temperature. The building with a lot of thermal mass, low albedo materials, and/or dark surfaces has an increase in the amount of stored heat and a slow rate of heat dissipation overnight. The substitution of such materials with other functional ones could play an important role in reducing the UHI phenomenon as well as creating easy solutions for cooling other than conventional air conditioning systems. In this respect, a lot of research works are concerned with developing such materials as eco-friendly cooling tools [7].

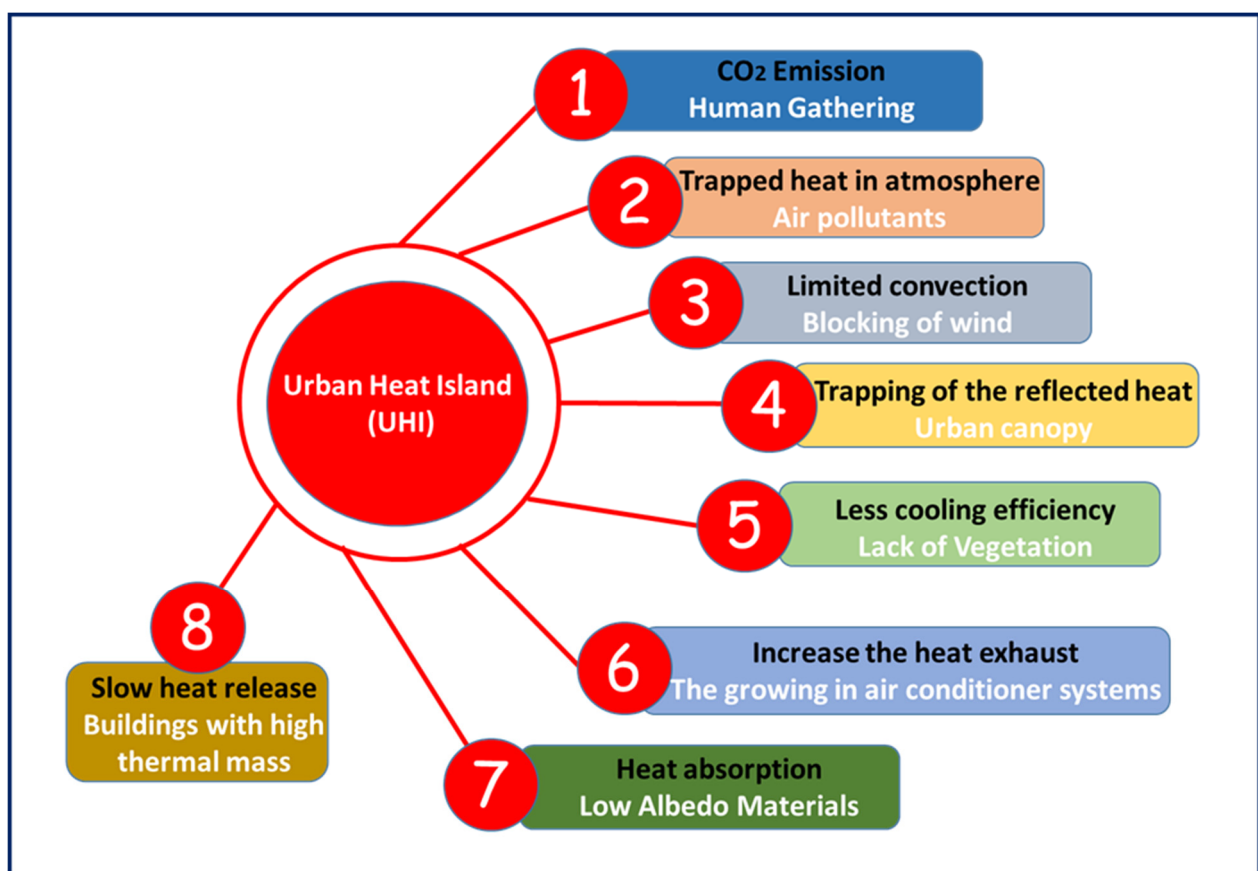


Figure 1. Schematic chart depicts the primary variables that contribute to UHI.

Accordingly, the scientists have been concerned to develop various geo-engineering solutions for facing the excessive climate change due to the UHI phenomenon [8]. These solutions are introducing policies and programs to fulfill the reduction in the temperature elevation of UHI. As is familiar, the material with high solar reflectance and high thermal emittance is called cool material [9]. The use of such highly reflective cool materials over building roofs and outdoor pavements is considered one of the most important solutions for mitigating the UHI by decreasing the atmosphere and surface temperature. Indeed, the surfaces of roofs and paved areas are reached to be 40% and 44% of the total area in the urban cities, respectively [10]. So, the usage of cool coating pigments (PGs) on such surfaces is offering comfort levels in indoor temperature as well as a reduction in energy needed for cooling. In this perspective, the increase in the implementation of the roof albedo

from 0.1 to 0.4 led to savings in energy of about 250 kW h/year to over 1000 kW h/year for mild climates and very hot climates, respectively [11]. Likewise, the usage of a cool roof for a public low-rise building in Poitiers (France) has recorded a reduction in surface temperature of up to 10 °C [12]. The widening in the usage of cool coating PGs enables limiting the extensive usage of the conventional AC systems, which are based on non-eco-friendly energy sources, as well as their exhaust heat into the atmosphere [13]. So, the implementation of cool coating PGs is considered an eco-friendly tool with combined impacts, the direct decrease in surface temperature and the reduction in AC systems, on limiting the UHI phenomena.

The working principle of PGs as a cool coating is based on their high reflectivity for near-infrared radiation (NIR). As is known, NIR is considered the direct consequence of heating up in solar radiation and represents about half of the released solar energy [14]. Indeed, the degree of PGs' color is dependent on either the scattered light and/or selective absorption of light. The PG is considered white PG when the absorption is very limited compared with scattering. If absorption is very small compared with scattering, the PG is a white PG [15,16]. Although the high efficiency of the white PGs in cooling performance due to their higher NIR reflectance reached 95.95% [17,18]. The darker-color PGs are demanded in real usages for their aesthetic performance and their comfortable vision. The intensive research works have been concerned with optimizing the cooling and aesthetic performances. The deficiency in NIR reflectance for colored PGs could be compensated by the synthesis of nanopigments (NPGs). Consequently, the nanostructures of PG materials enable the existence of a larger number of reflectance points that develop the scattering of light. In consequence, the present paper is aimed at reviewing the conceptual and developmental aspects of cool-colored NPGs. The figure of merit of cool-colored NPGs on both the cooling and aesthetic sides will be introduced. Moreover, the prospect for the future of cool-colored NPGs for painting applications will be discussed.

2. Basic Concepts

2.1. NIR Reflectance Materials

The composition of the solar spectrum includes 52% NIR radiation (700–2500 nm), 43% visible (VIS) light (400–700 nm), and 5% ultraviolet (UV) radiation (300–400 nm). The infrared radiations account for more than half of the solar radiations [15]. The solar spectral irradiation for direct light for the values at both the upper atmosphere and sea level are represented in Figure 2. These curves are plotted from the data obtained from Terrestrial Reference Spectra from the American Society for Testing and Materials (ASTM) and Solar Energy Monitor In Space (SEMIS) [19,20]. The area under the solar irradiance spectrum curve indicates the total amount of electromagnetic power that impacts the Earth per unit area, expressed in watts per square meter (W/m^2). Materials tend to heat up when they absorb NIR radiation from sunlight through the process of conduction [21,22]. This leads to an increase in their surface temperatures. Efforts are being made to mitigate the rise in indoor temperatures by reflecting near-infrared (NIR) radiation, which aims to reduce energy absorption and create more comfortable indoor environments [23]. Sunlight encompasses a broad spectrum of electromagnetic radiation that reaches the Earth's surface, including UV, visible, and IR radiations. The infrared portion is further divided into three main bands according to their wavelengths: the short wavelength infrared (SWIR) band, the mid wavelength infrared (MWIR) band, and the long wavelength infrared (LWIR) band [24,25].

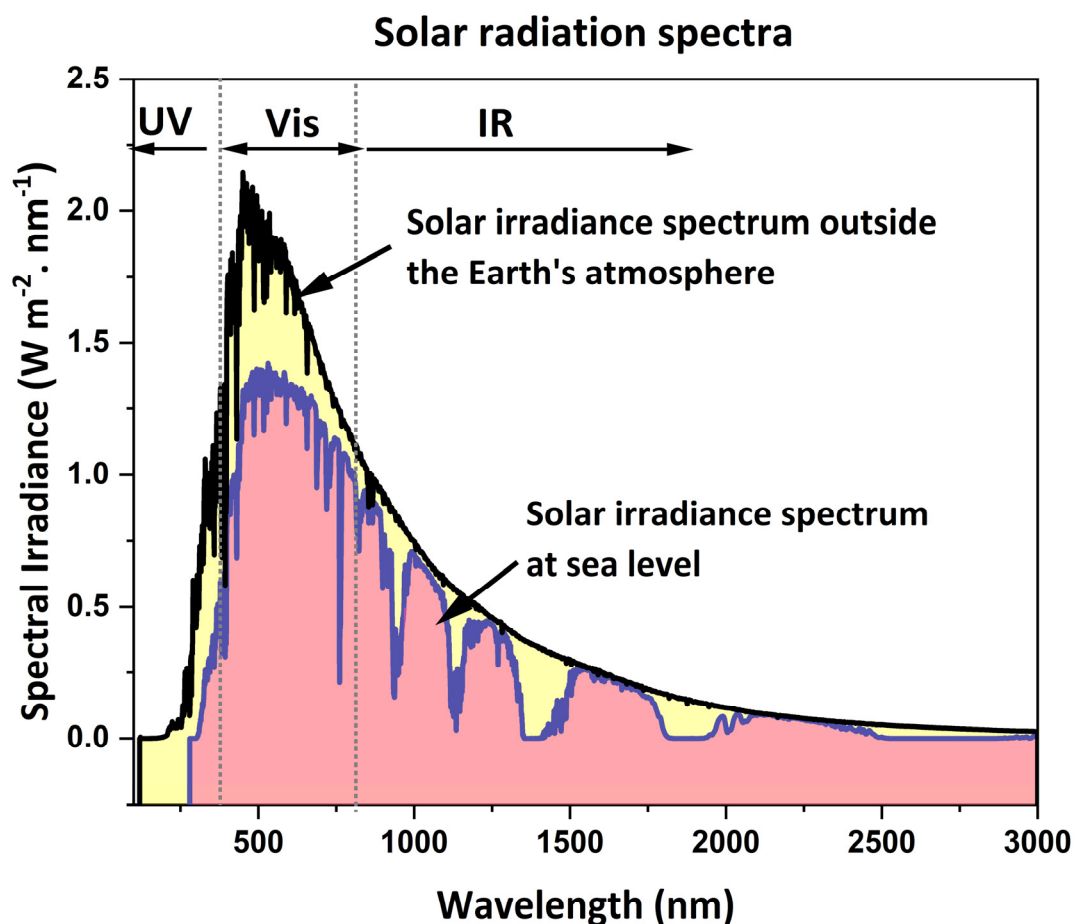


Figure 2. The solar spectral irradiation. This figure was redrawn using raw data in Ref. [19].

The use of such NIR reflective coatings is on the rise due to their crucial advances in decreasing solar heat absorption and lessening the UHI [26]. The painting of roofs and surfaces with cool coatings is among the most efficient options to provide enhanced thermal performance better than other cooling materials such as white marble and mosaic. The application of selective cool coatings enhances the thermal and optical properties of surfaces by changing the way they interact with solar radiation [26]. Moreover, NIR reflective coatings are capable of keeping surface temperatures lower than those of conventional coatings. NIR reflective materials can be classified into four main categories: transition metals, inorganic compounds, organic compounds, and natural materials [2,27].

2.1.1. Transition Metals

Transition metal materials for infrared reflection are known for their high infrared reflectivity and varied visible light absorption. Metals are known to be extremely reflective due to the large amount of free electrons they contain [25]. Metallic coatings ensure broad reflectivity and stable thickness. For example, the silver is being chosen as a top coating for its exceptional commercial reflectivity applications [28]. Metallic materials intended for reflective coating applications must exhibit high reflectance across a broad spectrum while minimizing absorption. Analysis of reflectance data for some of the transition metals over a wavelength range of 200 to 2000 nm reveals that metals such as aluminum, nickel, and platinum exhibit insufficient reflectance to qualify as effective reflective materials (see Refs. [27,29]). On the other hand, other metals such as silver with a high reflectance, make it frequently used in mirror optical applications [27]. Gold and copper exhibit comparable reflectance properties in the infrared spectrum, making them

effective choices for infrared reflective coatings at wavelengths above 700 nm. Metallic coatings like silver, aluminum, and gold films are highly beneficial for windows due to their exceptional infrared reflectivity [30]. Incorporating chromium into gold films and employing an energetic deposition technique, their strength and adhesion are enhanced. Also metal oxides such as $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ and $\text{ZnS}/\text{Ag}/\text{ZnS}$ coatings were reported as transparent thermal regulators and thermal camouflage [31], while other yellow pigments, such as Mica/(Ni, Sb)-doped rutile compounds, have a high near-infrared reflectance [32].

2.1.2. Inorganic Materials

Inorganic materials with infrared reflectivity can reflect wavelengths within the infrared spectrum while also selectively reflecting specific wavelengths of visible light. Choosing materials for infrared reflective applications necessitates a thorough assessment of their compatibility and durability to guarantee optimal performance and extended lifespan [33,34]. NIR inorganic substances are primarily used as PGs in coatings. The use of these inorganic materials in paint formulations has been found to enhance the optical stability of the coatings, as well as their solar reflectivity and emissivity, relative to white organic coatings [25,33]. Among the most common types of inorganic materials are metal oxides, nitrides, and sulfides, such substances are known for their NIR reflective properties [15]. Examples of other sets of inorganic materials that reflect infrared radiation are borides and carbides of transition metals like titanium, gold, copper, chromium, molybdenum, niobium, and tungsten [25]. Most metal oxides are used as NIR reflective coatings in fine powder form. The coating of the surface is a high-reflectivity metal, making them susceptible to wear and corrosion. Metal oxides such as TiO_2 could be used as a protective layer for the reflective metal and enhance their durability [35]. In addition, annealing improves the crystal quality and increasing the infrared reflectance of TiO_2 films [36]. Metal nitrides, such as (Ti, Al) N, are being studied as infrared reflective materials for solar applications. They offer exceptional heat, abrasion, and oxidation resistance, along with broad spectral selectivity [37]. High band gap metal oxides, like indium tin oxide and antimony or fluorine-doped tin oxide, have a band gap of about 3 eV, which permits transparency to visible light. These materials are effective in reflecting wavelengths greater than 1300 nm, making them appropriate for use as transparent heat mirrors [31].

2.1.3. Organic Materials

The development of organic materials that respond at NIR wavelengths is a significant research area [38]. The limited availability of NIR reflective organic materials is a consequence of the need to develop multilayer systems from such organic materials to obtain a significant infrared reflection. This difficulty is primarily due to the low variation in their refractive indices [25]. Since the reflectivity of a material is reliant upon the chemical composition and its refractive index, for a material to be considered ideal for reflection, it should not only demonstrate high reflectance but also sustain a low emissivity. On the other hand, the organic materials are widely used as a base material or in multilayered structures to regulate the infrared radiation [25]. Several inorganic materials have been studied for their suitability as infrared reflective materials. Examples of PGs derived from organic sources include halogenated copper phthalocyanine [39], azo compounds, perylene [40], polycyclic, and aromatic CH compounds [15].

2.1.4. Natural Materials

Chlorophyll is at present the sole acknowledged natural substance that reflects NIR light. This characteristic enables green leaves to reflect both green light and NIR light from sunlight. However, its limited chemical stability restricts its use in commercial applications. A noteworthy characteristic of chlorophyll is that green leaves reflect both green light and

infrared light. This reflection is among the spectral signatures that aid in distinguishing between different types of plants [27,41]. Examples of the reflection spectra that show the differences among the spectral signatures of some plants can be seen in [42]. A greater concentration of chlorophyll within a plant correlates with the enhanced reflection of NIR energy, which is a marker of the plant's health and productivity [27].

2.1.5. Applications of NIR Materials

Organic materials that regulate NIR light are becoming essential components or coatings. Notable examples in textile printing, inks, and paints include multilayered surfaces. Researchers created an electrically switchable polymer suitable for broadband infrared reflectors as smart windows. The idea is to create a dye-infused polymer that can be electrically toggled, enabling molecules to return to their original alignment once the voltage is removed [42,43]. Recent advancements in applications on NIR materials predominantly emphasize energy conservation and the reduction in UHI effects [2]. For instance, to minimize the cooling load on air conditioning units, coatings made from NIR reflective materials are applied on roofs as well as external walls of the building [4,22]. The energy required for cooling exceeds that for heating by more than threefold, making it a priority area for attention [27]. Another application is by coating the glass of the windows with transparent NIR reflective PGs that allow light to enter and prevent the greenhouses effect. Furthermore, the reflectance spectrum of the green PG that occurs naturally in plants is quite close to that of a green camouflage PG. Green NIR materials reflecting are utilized to create a camouflage effect in camouflage applications as well [21,44].

2.2. Pigments

PGs are classified as either inorganic or organic substances that are insoluble, meaning they are dispersed in the medium, typically referred to as the binder, in which they are incorporated. The creation of paint or coating involves the dispersion of these PGs in binders, with the addition of solvents and certain additives. The resultant properties of the paint are closely linked to the characteristics of the PGs, binders, and other additives utilized in the formulation [45]. The branch of PGs has attracted a significant amount of interest from researchers and industry professionals as well. As illustrated in Figure 3, column charts show the total number of publications per year for the last decade that contain the keywords "Cool Pigments", "NIR Pigments", and "Cool Nano Pigments". The results reveal the importance of this area of study. Noticeably, the total number of publications that are related to the keyword "Cool Pigments" is exceptionally high. This can be attributed to the broad and versatile nature of the keywords, which encompass a wide range of applications and research topics. In contrast, the number of publications that specifically address "Cool Nano Pigments" is the focus of nano PGs, particularly those that are utilized in the particular cool area of research.

There are several ways to classify PGs. The classification of PGs can be approached from several perspectives, such as their chemical composition, source (origin), color, crystallographic structure, and, most importantly, their functional properties [21]. The classification of PGs based on their chemical composition is divided into two major categories: inorganic and organic PGs [25]. Inorganic PGs can be further classified into different subclasses according to their chemical content, such as oxides [25], chromates [28,46], sulfides, silicates, and borates [45]. Organic PGs include a variety of compounds like azo compounds and indigoids [21]. Additionally, PGs can be categorized by their origin into natural and synthetic PGs. Natural PGs can be subdivided into mineral PGs, which include cinnabar, malachite green, talc, mica, and carbon black, as well as biological PGs that are extracted from animals or plants, such as garcinia, alizarin red, and indigo. For a collection of ref-

erences concerning these PGs, check with reference [21]. Under the category of synthetic organic PGs, one finds various artificial substances, including titanium dioxide [28] and phthalocyanine blue [33]. These PGs are classified according to their color into three primary categories: white PGs, colored PGs [18,28,47], and black PGs [48], regardless of their chemical composition and/or their origin [49]. Additionally, PGs are categorized by their functional characteristics, such as anti-rusting resist, magnetic, luminescent, electrically conductive, and near-infrared reflecting (NIR) PGs. NIR PGs are mainly classified into two groups: inorganic compounds and organic compounds. A preference for organic PGs is reported due to their non-toxicity, biocompatibility, and their cost-effectiveness [50]. In addition, organic reflective materials are soluble in standard solvents, thereby increasing compatibility with polymers [51].

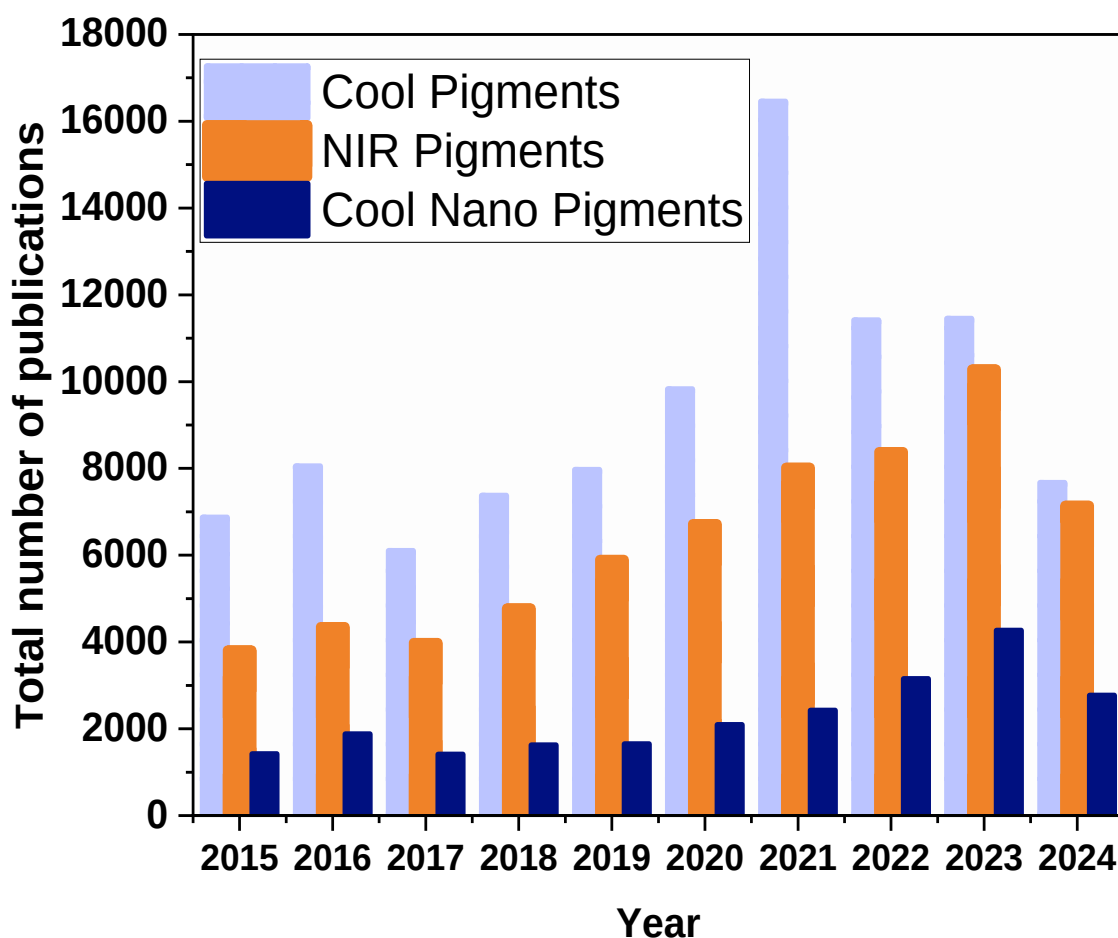


Figure 3. Column chart on total number of publications that contain the keywords “Cool Pigments”, “NIR Pigments”, and “Cool Nano Pigments” for period (2015–2024). This work is sourced from Dimensions®, at www.dimensions.ai, accessed on 15 October 2024.

The addition of NIR PGs to coatings offers a wide range of benefits. One of the stand-out advantages is the enhanced durability of the coatings, the reduced thermal degradation. These PGs are allowing the coatings to withstand harsh environmental conditions effectively. NIR PGs also provide a diverse selection of attractive color options. It is particularly noteworthy to mention that their ability to maintain cooler surface temperatures not only promotes safer handling practices but also significantly reduces the amount of heat that is transferred into buildings. This reduction in heat transfer leads to lower energy demands for air conditioning systems, which is especially advantageous in regions characterized by high temperatures. Such above benefits contribute to energy efficiency and sustain-

ability by using NIR PGs [21,45,52]. There is a growing trend in the use of NIR PGs for cooling applications related to roofs and buildings [22]. NIR reflective screens composed of polymers containing NIR PGs can be utilized to create a greenhouse effect, facilitating the transmission of visible light while reflecting NIR light. Additionally, NIR PGs of some metal oxides may be used in camouflage for both infrared applications [27].

2.3. Factors Affecting Infrared Reflectivity of Pigments

There are some variables that impact the reflective performance of NIR PGs when they are added in the coating. Several factors affect the reflectance behavior in the near-infrared (NIR) spectrum, with the selection of PGs being particularly critical. Other influential elements include particle size, crystallinity, and the aggregation state. The NIR reflectivity of coatings is contingent upon multiple factors, such as the choice of individual PGs, the techniques of milling and dispersion, particle size, the blending of infrared reflective PGs, opacity, molecular structures, and any potential contamination. Of course, the method used from sample preparation would certainly affect the NIR reflectance of the PGs, which may contribute to the contradicting conclusion in the studies of affecting factors [51]. A discussion of some of these factors will be presented below [30].

2.3.1. Pigment Selection

It is crucial to select PGs that possess optimal reflectivity in the NIR range when developing NIR reflective PGs and coatings. The selection of PGs should be guided by the values of color scale parameters (L^* , a^* , and b^*), which are helpful in determining the intended color shade. The L^* value, which can range from 0 to 100, is particularly significant, with 100 indicating a perfect white and 0 representing black. Although values of a^* and b^* do not need to conform to exact numerical values, they are indicative of the color characteristics; a positive a^* value reflects red, while a negative a^* value indicates green. In terms of the b^* value, a negative value corresponds to blue, and a positive value signifies yellow color [27].

2.3.2. Dispersion

Achieving thorough the dispersion of PG particles within the emulsion is essential for ensuring high-quality PGs. It is crucial that these PGs exhibit compatibility with various types of solvents and water-based coatings. One significant factor that contributes to the complete dispersion of PGs in a small media mill is the attainment of a fine grind. However, it is important to avoid excessive grinding as this can lead to the fragmentation of the particles, adversely impacting both the color and the infrared reflectivity of the PGs. The optical behavior of a pigmented layer of paint is contingent upon the optical properties of the PG particles, especially the optical properties of the generally colorless dispersion medium [21].

2.3.3. Blending Pigments

The final properties of the paint or coating are influenced by the properties of the binder PGs, and/or any other additives [45]. It is essential to follow the correct process when preparing multiple coatings that utilize various PGs. The standard practice involves combining the PGs prior to their incorporation into the paint, necessitating the mixing of PGs with water to create a paste. Nevertheless, there are cases where the absorption characteristics of different PGs in various regions lead to a total reflectance that is inferior to that of the separate PGs [45].

When the PGs absorb light in distinct spectral regions, the overall reflectance may fall shorter than that of the separate PGs. In such cases, the effect of absorption can overshadow that of scattering. You et al. [53] in their study explore the optical and cooling characteristics

of blended coatings with equal mass proportions of CuO and TiO₂, using different CuO particle types. The results highlight the importance of considering interactions between functional reflective particles at micrometer and nanometer scales in coating formulation. The preliminary analyses of coatings show that raising the TiO₂ nanoparticle concentration slightly improves spectral reflectance for both nano and micro particles [53].

2.3.4. Opacity

Infrared reflective PGs, such as titanium dioxide (TiO₂) powder, exhibit significant visible opacity [21]. This means such PGs primarily scatter or transmit the infrared radiation only. On the other hand, thin films are not proficient in scattering or reflecting all infrared radiation, which allows a portion of this radiation to penetrate to the substrate. Therefore, despite the visible opacity of these coatings, they do not fully block infrared radiation. Increasing the thickness of the coating is one approach to improve infrared opacity. Additionally, the volume concentration of PGs plays a significant role in determining the infrared opacity of the coatings.

2.3.5. Contamination

The design of an infrared reflective coating requires a conduct to maximize the solar reflectance and minimize contamination results from infrared-absorbing materials. To ensure optimal reflectivity, it is vital to eliminate any cause of contamination that could present in infrared reflective PGs [30]. One must be cautious to prevent the contamination of infrared reflecting products with materials that exhibit high absorption. Contamination can occur when two PGs with differing infrared absorption properties are combined, leading to a situation where the infrared absorption of the latter PG predominates, thereby degrading the performance of the infrared reflective PG. Such contaminations can significantly impair the reflectance of the coatings [45].

2.3.6. Particle Size

The size of PG particles is a critical parameter that significantly affects both optical properties and PG dispersions. The optical characteristics of a product play a vital role in determining the final appearance of the coated surface. For example, the finishing of a paint can vary from gloss to matte or satin, contingent upon the particle size. However, predicting the impact of PG particle size on the optical properties of coatings presents considerable challenges. This complexity arises from the fact that PGs are composed of solid particles suspended within a liquid or solid medium. The properties of these PGs are closely linked to their particle size and distribution [25]. Generally, larger particle sizes provide enhanced opacity, although there exists a limit beyond which opacity may decrease. In contrast, smaller particle sizes tend to improve tint or color strength, but excessive reduction can lead to changes in the PG's color shade [21]. Additionally, particle size affects the compacted properties of the PG, with viscosity being influenced as well; smaller average particle sizes usually result in higher viscosity due to more efficient packing and increased surface area interactions among the PG particles [45]. For highest reflectivity, the size of particles within a compound significantly influences its color, which in turn affects its reflective properties. A reduction in particle size results in an increased number of reflections occurring at the grain boundaries [21,54]. Consequently, this leads to a decrease in the penetration depth of incoming light, resulting in lower absorption rates and higher reflectance. Furthermore, for light to be effectively reflected, the particle size must exceed half the wavelength of the light. For infrared light, which has a wavelength ranging from 700 to 1100 nanometers, the particle size should be no less than 0.35 to 0.55 microns [38,55].

3. Developmental Status and Limitation of Cool Pigments

The application of materials with high solar reflectivity and spectral emissivity, known as cool materials, has been acknowledged as a highly promising and effective strategy for addressing the heat island issue [8]. This technique plays a crucial role in enhancing urban albedo. The incorporation of cool materials in architectural structures and urban environments has garnered considerable attention and is increasingly pervasive. The aforementioned studies [56–59] have provided evidence of the significant potential of these materials in mitigating surface and ambient temperatures. PGs possessing favorable thermal characteristics exhibit the capacity to reflect the infrared segment of the solar spectrum, so diminishing the energy consumption linked to air conditioning systems through the mitigation of heat transfer into the building [8]. Furthermore, the recent development of synthetic materials that possess reflective and infrared-reflecting properties for pavements has significantly enhanced the albedo of urban areas and reduced the highest recorded ambient temperature.

The series of inorganic white PGs with infrared reflectivity comprises PGs such as TiO_2 , Al_2O_3 , and ZnO [2]. Table 1 illustrates the NIR reflectance of some white pigments prepared with various synthesization techniques. These PGs have garnered significant attention due to their notable reflectivity in VIS and NIR spectra. However, the high reflectance in the VIS range results in a bright and visually displeasing white color to the human eye. In addition, it should be noted that the aforementioned white surfaces tend to accumulate dirt due to the presence of air pollution or stains caused by rain. Consequently, a considerable number of individuals tend to opt for the use of white PGs in non-exposed locations [4]. In addition, it should be noted that the NIR reflectance of white cool PGs diminishes over time. Furthermore, mutually facing structures, which form an urban canyon, may cause radiation to bounce back and forth without reemerging. The utilization of color elements in doping offers a promising strategy for addressing the limitations associated with white cool PGs. The introduction of color elements such as chromium (Cr), nickel (Ni), cobalt (Co), and others into a material leads to a decrease in NIR reflectivity. This decrease can be attributed to an increase in absorbance, which arises from the creation of trap levels associated with the doped elements [13,60,61]. In order to improve the reflectance of cool-colored materials, the utilization of nanostructured PGs, NPGs, is employed. The synthesis technique enables the generation of a greater quantity of reflectance points, hence enhancing the scattering of light [62]. Figure 4 depicts the relationship between particle size reduction and the augmentation of light's diffuse reflectance by providing a greater number of sites. The roughness of a material's surface is a crucial factor that considerably influences the intensity of diffuse reflection. The greater surface area possessed by nanoparticles, as opposed to microparticles, may potentially result in increased reflectivity for various materials. The high degree of reflectance observed allows for the mitigation of the elevated light absorbance caused by trap levels associated with the doped color element in color NPGs. Recently, nanosized amorphous TiO_2 has demonstrated potential for high NIR reflectance, which can be attributed to its substantial surface area resulting from its prominent surface characteristics [61,62]. Figure 4 demonstrates that the roughness of a surface enhances the reflectance, even with small particles like amorphous and mesoporous particles (Figure 4c), as compared to crystalline nano- and micro-sized particles (Figure 4a,b). According to Mansour et al. [28], nanosized amorphous TiO_2 doped with Cr showed effective performance as cool-colored NPS. This approach increases the likelihood of utilizing amorphous and mesoporous particles in applications involving cold painting. Using such materials offers an additional advantage, such as the low temperature required for synthesis and their low density, which will improve their dispersion in epoxy paint. However, the thermal stability

of amorphous materials is relatively lower than that of crystalline materials, which might have an impact on their reflectance properties and color indices.

Table 1. Examples of white pigments and their NIR reflectance values.

White Pigment	Structure	Synthesis Technique	NIR Reflectance %	References
ZnO	Hexagonal, Wurtzite	Thermal decomposition	80.47	[50]
Oleic-acid-treated ZnO	Hexagonal, Wurtzite	Thermal decomposition	84.75	[50]
ZnO	Hexagonal, Wurtzite	Thermal decomposition	64.8	[46]
ZnO	Hexagonal, Wurtzite	Modified polymer pyrolysis with γ -irradiation	88	[13]
ZnO	Hexagonal, Wurtzite	Arc discharge	14 to 54	[15]
TiO ₂	Tetragonal, Anatase	Hydrolysis process	79.53	[28]
TiO ₂	Tetragonal, Rutile + Anatase	Hydrolysis process	94.72	[32]
TiO ₂	Amorphous	Hydrolysis process	88.14	[61]
TiO ₂	Rutile	Polymer pyrolysis	87	[16]
La ₂ Ce ₂ O ₇	Fluorite type	Sol–gel	95.95	[17]
LaYO ₃	Fluorite type	Sol–gel	92	[63]
ZnTiO ₃	Perovskite	Solid state	95	[64]
ZnAl ₂ O ₄	Spinel	Solid state	85	[65]
BiPO ₄	Monoclinic	Hydrothermal	80	[66]

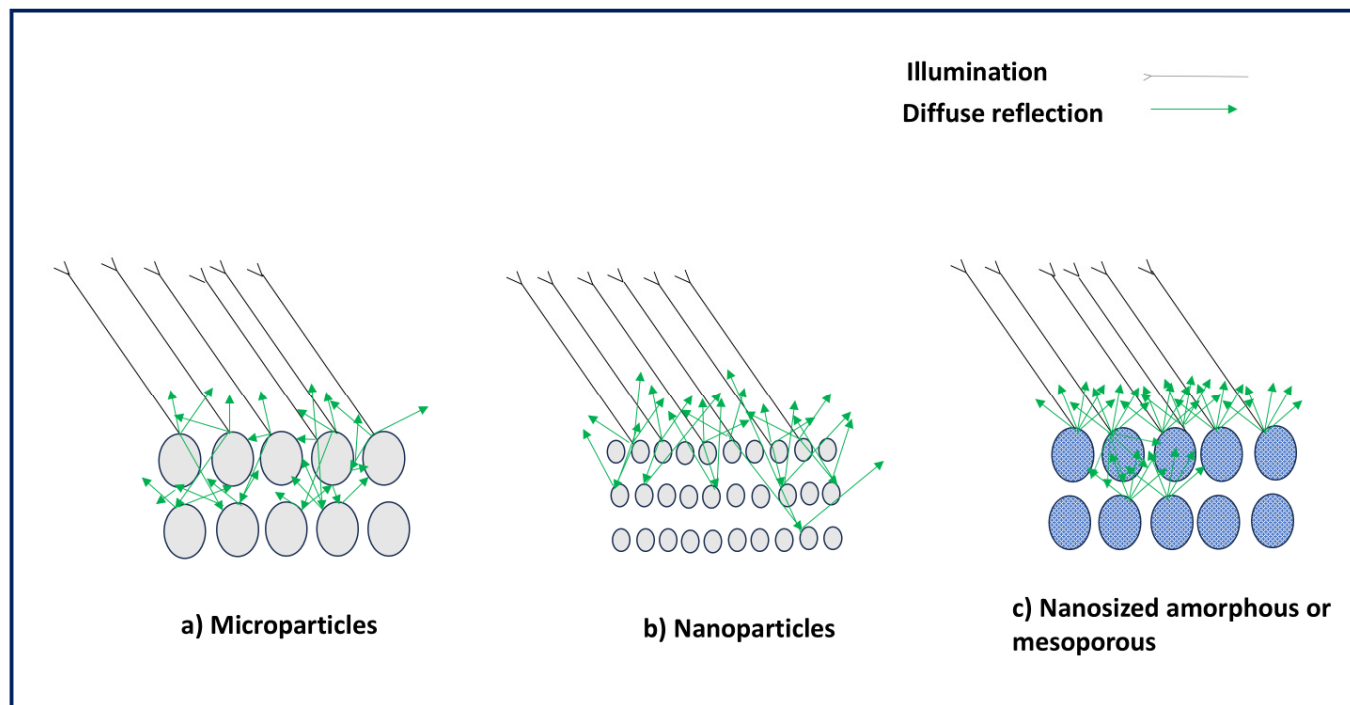


Figure 4. The influence of particle size and surface variation on diffuse reflectivity.

4. Cool-Colored Nanopigments

The presence of colored NPGs within the nanostructure aids in offsetting the decrease in NIR reflectance. Undoubtedly, the advancement of techniques for creating nanoparticles allows for the investigation of unique material characteristics. The method and conditions

utilized to create nanoparticles have a significant impact on several parameters. Among these parameters are the particles' size, shape, microstructure, and surface morphology. Hence, the utilization of cool-colored PGs in nanostructures enables the manipulation and enhancement of both cool and aesthetic characteristics. Both sides' performances are based on the reflectance behavior in the NIR range and the VIS range, respectively. The next section elucidates the methodology for assessing the capacity of these PGs to reflect solar radiation in both the near-NIR and VIS spectrums. Consequently, the optimization parameter, encompassing both ranges, was established to assess the efficacy of NPGs in terms of both coolness and aesthetics.

4.1. Figure of Merit for Cool-Colored Pigments

To meet the thermal and aesthetic needs at the same time, pigmented coatings that highly reflect the NIR portion of sunlight while minimizing the reflectance of visible light are applied to dark surfaces. In order to meet these specifications, the optimization parameter is suggested as [67]:

$$R_{R/V}^* = \frac{R_{NIR}^*}{R_{VIS}^*} \quad (1)$$

The NIR solar reflectance (R_{NIR}^*) refers to the reflectance ability over the NIR region from 700 to 2500 nm. It may be computed based on ASTM standard number G173-03 using the following equation [67]:

$$R_{NIR}^* = \frac{\int_{700}^{2500} r(\lambda) i(\lambda) d(\lambda)}{\int_{700}^{2500} i(\lambda) d(\lambda)} \quad (2)$$

where $i(\lambda)$ is the solar spectral irradiance ($\text{Wm}^{-2}\text{nm}^{-1}$), and $r(\lambda)$ is the spectral reflection (Wm^{-2}) that was measured in the lab.

While R_{VIS}^* represents the aesthetic quality of a pigmented coating, it considers the sensitivity of the human eye to different wavelengths of light. It can be computed using the subsequent formula:

$$R_{VIS}^* = \frac{\int_{390}^{700} r(\lambda) \eta(\lambda) i(\lambda) d\lambda}{\int_{390}^{700} i(\lambda) d\lambda} \quad (3)$$

The variable $\eta(\lambda)$ represents the standardized luminous efficiency, which is based on the photopic conditions of the CIE #1931 model [67]. The term “photopic condition” relates to conditions that are well lit, such as ordinary daylight, and how they are perceived by the eye.

The optimization parameter $R_{R/V}^*$ is influenced not only by the doping element, doping concentration, and microstructure but also by particle size, as described by M. Baneshi et al. [68]. The influence of the particle size on $R_{R/V}^*$ has been investigated at different film thicknesses and volume fractions for TiO_2 /acrylic resin. The fluctuations in $R_{R/V}^*$ exhibited a nonmonotonic pattern in relation to particle size across all tested values of film thickness and volume fraction. The nonmonotonic fluctuations have been ascribed to disparate rates of change in reflectance in both the NIR and VIS ranges. The highest value of $R_{R/V}^*$ was 4.278, seen for TiO_2 pigmented coating with a particle size of 778 nm. Conversely, the lowest value of $R_{R/V}^*$ was 0.5, observed at a particle size of around 100 nm. Furthermore, it is noteworthy that particles with sizes ranging from 10 to 30 nm and particles larger than 40 μm have a limiting value of R that is approximately 3.0 [68].

It should be emphasized that the reflectance of PG particles is largely reliant on their ability to absorb and/or deflect solar light [69]. The importance of PG particle absorption and scattering is evident in the UV range, particularly for highly absorbing UV radiation materials such as TiO_2 and ZnO . To improve the benefits of such materials in coating

applications, their scattering effectiveness for UV rays, which account for 5% of solar radiation, should be increased in order to prevent the destruction of the organic host coating materials [70]. As a result, particle size plays a vital role in determining scattering efficiency, which influences PG cooling performance. Many studies have confirmed that when particle size decreases, scattering effectiveness increases in the UV area, according to the calculations based on Mie theory [67,71]. Such findings support the great efficiency of cool PGs in nanoscale structures.

The correct color identification of PGs is critical for coating applications. Furthermore, the research of the color stability of cool-color PGs provides crucial insights into their performance and durability. A colorimeter is utilized to examine the color of PGs based on the L^* , a^* , and b^* characteristics. The color lightness coordinate, L^* , has a value of 0 for black and 100 for white. The coordinates a^* and b^* range from negative values for green and blue to positive values for red and yellow, respectively. The polar representation of color space is employed to differentiate colors based on two parameters, h^o and C^* , which are referred to as hue and chroma or saturation, respectively. The transformation from CIE (L^* , a^* , and b^*) representation to h^o and C^* representation was achieved using the following equations [72]:

$$h^o = \tan^{-1} \left(\frac{b^*}{a^*} \right) \quad (4)$$

$$C^* = \sqrt{(a^*)^2 + (b^*)^2} \quad (5)$$

Another way to characterize colors in the CIE system is to use a brightness parameter Y and two-color coordinates x and y to identify a point on the chromaticity diagram. These characterized coordinates provide more precise color measurement than other systems because the parameters are based on the spectral power distribution (SPD) of light emitted from a colored object and are influenced by sensitivity curves measured for the human eye [73]. Use the easy red–green–blue (RGB) color calculator to convert CIE $L^*a^*b^*$ to xyy coordinates [28]. Also, the XYZ coordinate can be calculated directly from the measured spectral distribution of reflectance $r(\lambda)$ using the following formulae [74]:

$$\begin{aligned} X &= \frac{1}{k} \int_{380}^{780} i(\lambda) r(\lambda) \bar{x}(\lambda) d\lambda \\ Y &= \frac{1}{k} \int_{380}^{780} i(\lambda) r(\lambda) \bar{y}(\lambda) d\lambda \\ Z &= \frac{1}{k} \int_{380}^{780} i(\lambda) r(\lambda) \bar{z}(\lambda) d\lambda \end{aligned} \quad (6)$$

where k is a normalization factor established in such a way that an object with uniform reflectance $r(\lambda) = 1$ produces a brightness Y equal to 1. Using the values of X , Y , and Z obtained from equations, the normalized color coordinates are determined as follows:

$$\begin{aligned} x &= \frac{X}{X+Y+Z} \\ y &= \frac{Y}{X+Y+Z} \\ z &= \frac{Z}{X+Y+Z} \end{aligned} \quad (7)$$

As $z = 1 - x - y$, it does not add any new knowledge. In this manner, the chromaticity coordinate (x, y) and brightness Y are used to describe the color of a refractive object.

4.2. Application and Impact of Cool-Colored Nanopigments

The use of colored PGs in reflective coatings for claddings and roofing components has sparked interest in addressing the environmental and economic challenges caused by the UHI effect. Cool materials include cool tiles, cool facade coatings, and cool roof paintings. A cool roofing coating is made of paint (or glaze) containing colorful PGs with a high reflection efficiency in the near-infrared region of the sun spectrum. Another approach

is to coat the pavements with such cool PGs, which reflect a greater proportion of solar radiation than standard darker asphalt coatings, minimizing heat storage in the pavement. In 2020, the city of Phoenix launched the “Cool Pavement Pilot Program” in partnership with Arizona State University, in which the city deployed GuardTop®’s CoolSeal product to 36 miles of residential neighborhood roads and one public parking lot. According to the program, an overview report is published on the city of Phoenix’s website, stating that using cool pavement reduces the surface, subsurface, and air temperatures when compared to non-treated asphalt concrete [75]. In June, subsurface temperatures dropped by 6.0 °F but declined to around 4.0 °F over the winter. In summer, the surface temperatures are reduced by up to 12 °F compared to the standard aged pavement during the daytime. However, the reduction in air temperature caused by cool pavement was minor, reaching an average of 0.13 °F lower than non-treated asphalt at all heights and periods that were examined. The largest significant difference came after sunset, with a 0.6 °F lower temperature over cool pavement at all recorded heights. Although employing cool pavement results in a slight drop in air temperature, it has a huge impact in terms of potential electrical power savings. A simulation study for buildings across the entire city of Phoenix suggested that extending cool pavement on residential roads could save USD 10 million to USD 20 million in avoided residential air conditioning annually, according to the published summary of the program in October 2024 on the city’s website [75]. Figure 5 depicts images of the pilot scale for the “Cool Pavement Pilot Program” in the city of Phoenix. In addition to the benefits of cool pavement in lowering surface, subsurface, and air temperatures, another beneficial effect is the reduction in UV remission. The overall UV radiation was 5.9% when cool pavement was used on such a pilot scale, which is somewhat less than the reflection from asphalt (8.8%) and concrete (6.0%). This is referred to as the positive impact of cool pavement because it reduces the risk of sunburn by decreasing the reflected UV rays. Another pilot scale for cool pavement was implemented on “Abdullah bin Jassim Street by Souq Waqif, Doha, Qatar” by the public works authority “Ashghal” to reduce asphalt temperature on roads. According to Ashghal’s assessment, the employed cool pavement had a considerable impact on overall temperature reduction, but there were no specifics on the efficiency of surface, subsurface, and air temperature reductions [76]. The implementation of a pilot cool pavement project featuring an asphalt-cooled material in Doha city is represented in Figure 6.



Figure 5. The city of Phoenix is implementing a pilot project for the “Cool Pavement Pilot Program” [75].



Figure 6. The implementation of a pilot scale for cool pavement paint is underway on Abdullah Bin Jassim Street [76].

Other applications for colorful NPGs besides cool coatings include vehicle coatings and NPG TV screens [77]. NPGs are utilized in automotive coatings because they improve tribological and mechanical qualities [78]. The use of NPGs in paints allows for increased scratch and abrasion resistance, hardness, and strain to failure while maintaining toughness. However, the phosphor screen utilized on cathodic ray tube TVs based on NPGs improves PG stability in the dispersion medium, contrast, color strength, contrast, and transmittance [79]. Despite the fact that near-infrared nanopigments show promise and have useful features in a number of possible applications, they are not currently used extensively. This restriction is imposed by the considerable initial expense of the nanostructured pigments. Finding efficient and low-cost synthesis methods with high yield should be a major focus of future studies in order to overcome this obstacle. In addition, by investigating the cooling and color performance of natural materials, sustainable and environmentally friendly alternatives to synthetic-colored pigments may be provided. For large-scale applications requiring cool-colored nanopigments with unique and functional properties, research into blending or treating certified natural materials with nanopigments may provide an appropriate solution.

5. Summary

In this review article, an overview of the basic concepts and progress in NIR-reflecting cool-colored NPGs is provided. In purpose of that review, it provides an overview of the different types of NIR reflective materials, specifically focusing on PGs, and discusses the factors that affect their performance. Classifications of the pigments by chemical composition, origin, color, and their functional properties are discussed. NIR-reflecting

PGs are mainly classified into two groups: inorganic and organic compounds. The NIR reflectance property allowed us to determine the cooling performance of PGs. One of the reasons for the limited use of cool PGs in cool painting applications is their high reflection of solar radiation in the visible spectrum, which is uncomfortable for the human eye. Because of this constraint, colored PGs are best suited for realistic cool painting applications. However, adding color to the PGs reduces their NIR reflectivity, making them less comfortable to see. One of the most important ways to mitigate the detrimental impacts of PG coloring is to use PGs in nanostructures, either by reducing the particle size of the PG materials or by changing their surface characteristics. In this regard, we have discussed the effect of particle size and surface variation on the diffuse reflectivity of PGs. Cool-colored nanostructured PGs found in cooling coatings, also known as NPGs, are considered promising functional materials for fighting the UHI problem. This review study focused on cool-colored NPGs and investigated their cooling and aesthetic merits. In this respect, the optimization parameter, $R_{R/V}^*$, measure for pigmented coatings was defined as checking how well the pigment can reflect the NIR part of sunlight while reducing the reflectance of visible light. The use of NPGs in cool pavement is considered an environmentally benign method that effectively solves the UHI phenomena by lowering surface, subsurface, and air temperatures while reducing dependency on air conditioning equipment. To maximize the benefits of these colors while minimizing any negative features, eco-friendly, toxic-free pigments must be developed. Recent uses and successes using cool-colored NPGs were also investigated. Future research should focus on efficient, low-cost, high-yield synthesis approaches to widen the usage of nanopigments in large-scale applications. Moreover, alternatives to synthetic-colored pigments can be produced by examining natural materials' cooling and color properties.

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Nomenclature

$R_{R/V}^*$	Optimization parameter of pigmented coatings.
R_{NIR}^*	Reflectance ability over the NIR region from 700 to 2500 nm.
R_{VIS}^*	Reflectance ability over the visible region.
$\eta(\lambda)$	Standardized luminous efficiency based on the photopic conditions of the CIE #1931 model
$r(\lambda)$	Spectral reflection (Wm^{-2}) that was measured in the lab.
$i(\lambda)$	Solar spectral irradiance ($\text{Wm}^{-2}\text{nm}^{-1}$) that was measured in the lab.
L^*	Color lightness coordinate.
a^* and b^*	CIE color coordinates for green and blue ranges for red and yellow colors, respectively.

h^o	Hue of color space.
C^*	Chroma or saturation of color space.
X and Z	CIE color coordinates of the chromaticity.
Y	Brightness parameter.
x and y	Normalized color coordinates of the chromaticity.
k	Normalization factor.
$\bar{x}(\lambda)$, $\bar{y}(\lambda)$, and $\bar{z}(\lambda)$	Color coordinates of the chromaticity.

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