

Passive Radiative Cooling of Structures with Thick Film Nanocomposites

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Abstract

Passive cooling via state-of-the-art “cool roof” coatings significantly drops roof top temperatures by claiming to reflect nearly all electromagnetic radiation incident upon its surface – the result is significant reductions in electricity consumed for air conditioning. However, passive radiative cooling research reports even greater cooling load reductions and potential energy savings. In addition to “reflective cooling”, the surface emits thermal radiation through the earth’s atmospheric window to the cold of outer space. A literature review of many experiments demonstrates radiative cooling through numerous fabrication methods and material combinations, yet many methods rely upon expensive equipment for nanoscale precision in a controlled environment for uniform multi-layer thin films or nanostructures. The less complex methods encase dyes or nanoparticles into very thin polymer films not suitable for exposure to harsh environmental conditions or application to many structures. Hence, passive radiative cooling technology is still not cost-effective, scalable, and robust enough to be extensively commercialized like “cool roof” coatings. This paper presents a thick film nanocomposite coating to improve upon cool roof technology utilizing selectively emitting nanoparticles and thick film nanocomposite application methods. The proposed thick film nanocomposite can be tailored for colder climates to provide the needed cooling power during summer and to avoid costly heating load increases during winter weather. A scalable passive radiative cooling coating applied on buildings and other surfaces could significantly reduce global warming by balancing the global heat flux.

Keywords: radiative cooling, thick film, nanocomposite, cool roof, spectrally selective nanoparticles, global warming

1. Introduction

Approximately 25% of all the energy used in the U.S. is for cooling of buildings. Although that percentage is lower for the rest of the world, it is rapidly increasing. Currently the US EPA refers to modern urban areas with populations of more than 1 million people as “heat islands” because annual mean air temperature can be 1–3°C (1.8–5.4°F) hotter than surrounding rural areas. In the evening, the city can be 12°C (22°F) hotter than the countryside. (U.S. Department of Energy 2021 and US Environmental Protection Agency 2021) Heat islands increase peak cooling loads, air conditioning costs, air pollution, greenhouse gas emissions, heat-related illness/mortality, and even water pollution (Oke, 1997). In developed countries, over 50% of urban surface areas are either roofs or paved surfaces; this does not include vehicles or other surfaces facing the sky (Al-Obaidi et al. (2014) and Kolokotsa et al. (2013)). In some locations, roofing systems constitute 70% of a home’s total heat gain (IPCC, 2021 and Oke, 1987).

Radiative coolers passively cool terrestrial objects by selectively emitting heat through the earth’s atmospheric window to the cold of outer space while reflecting electromagnetic radiation outside the atmospheric window. Surfaces exposed to incoming solar irradiance are prime locations for day and night passive radiative cooling technology. Catalanotti et al. (1975) demonstrated that a 12.5 µm thick TEDLAR (polyvinyl–fluoride plastic) film on top of aluminum substrate can passively cool a surface 12 °C below ambient temperature at night. Yet this technology to reduce the “heat island” effect did not progress to a commercially available product due to application and durability limitations.

At the global level, an Intergovernmental Panel on Climate Change (IPCC) report states with high confidence that “Global surface temperature has increased faster since 1970 than in any other 50-year period over at least the last 2000 years...Global surface temperature will continue to increase until at least the mid-century under all emissions scenarios considered. Global warming of 1.5°C and 2°C will be exceeded during the 21st century unless deep reductions in carbon dioxide (CO₂) and other greenhouse gas emissions occur in the coming decades” (IPCC, 2021).

Outgoing Longwave Radiation (OLR) is a measure of the amount of energy emitted to space by earth's surface, oceans, and atmosphere. For the Earth to remain at a stable temperature, the amount of longwave radiation streaming from the Earth must be equal to the total amount of absorbed radiation from the Sun Mingke et al. (2015). Unfortunately, a report by Stephens et al. (2012) showed that in 2012, the Earth was absorbing $\sim 1 \text{ W m}^{-2}$ more than it was emitting, leading to an overall warming of the climate.

There are many places on earth where the OLR balance is in the negative range. Some of the biggest heat sinks are situated near the equator with an average OLR of -80 W m^{-2} (Lee, 2014). Fortunately, a geoengineering approach to increase radiative heat emission from the Earth to space by covering 1%–2% of the Earth's surface with thermally emissive materials that can radiate $\sim 100 \text{ W m}^{-2}$ through the atmospheric window may counter global warming by bringing the total heat flux back into balance (Munday, 2019). Covering 2 % of the Earth's surface equates to 10,201,440 km², since the total surface area of Earth is 510,072,000 km². It is preferable to place thermally emissive “radiative” cooling coatings on structural surfaces exposed to the sky, like roof tops due to the added benefit of reduced energy consumption and CO₂ emissions, instead of on land in competition with other primal needs such as agriculture, biodiversity, or decentralized production of energy. It may be theoretically possible to balance the global heat exchanges by emitting more infrared radiation from the earth. Presented in this paper is a possible solution to carry out such an ambitious project.

2. State of Art Review

Daytime radiative cooling is the biggest challenge since 95% of the incoming solar heat flux arrives in the 0.3–2.4 μm waveband during the day. To attain diurnal cooling of a surface below ambient temperatures a radiative cooling coating must overcome the strong solar radiation with high reflection in 0.3-2.5 μm wavelength range and high emission in the primary atmospheric window. Achieving daytime radiative cooling is a goal of this research since the greatest benefits, like peak load reduction in buildings, can be realized during this time.

A state-of-the-art review of passive radiative cooling did not find a widely available commercial “radiative cooling” product. However, cool roof coatings are widely available which drop roof top temperatures by high broadband reflectance of incoming solar radiation. The U.S. Department of Energy modeled a 15% reduction in the annual air-conditioning energy use of a single-story building with a cool roof (U.S. Department of Energy, 2021). A modeling study by Lamba et al. (2018) for a passive radiative cooling coating with a cooling power of 100 W m^{-2} calculated significantly better cooling load reduction than cool roof coating. The model predicted a passive radiative cooling coating on 50% of a roof's surface would eliminate a Chicago, Illinois single-story building's peak cooling load in July, while amazingly reducing a Miami, Florida building's cooling load by 95% in July and 90% in August.

According to Grand View Research (2020) the largest market share of passive cooling coatings belongs to cool roof coatings whose global market size was estimated to be worth USD 3.59 billion in 2019 – thick film elastomeric coatings account for over 64% of the revenue.

The cool roof coating selected for a side-by-side comparison in this research is the GacoRoof GR1600 Series White (Firestone, 2021) whose safety datasheet composition information is listed in Table 1.

Tab. 1: Composition information of leading cool roof product (GacoRoof GR1600 Series White)

Material	CAS No.	Weight % *
Dimethyl siloxane, hydroxy-terminated	70131-67-8	30-60%
Limestone	1317-65-3	30-60%
Distillates (petroleum), hydrotreated light	64742-47-8	10-30%
Titanium dioxide (dust)	13463-67-7	7-13%
Butan-2-one O,O',O''-(methylsilylydyne)trioxime	22984-54-9	1-5%
Silicon dioxide	7631-86-9	1-5%
Silica, quartz (dust)	14808-60-7	0.1-1.0%
Aminopropyltriethoxysilane	919-30-2	0.1-1.0%
Octamethylcyclotetrasiloxane	556-67-2	0.1-1.0%

The recommended thickness of cool roof coating examined in this research is 22 mil (558.8 μm). Cool roof coatings contain randomly distributed reflective nano and micro sized particles of varying morphology in an acrylic or silicone binder. This research used similar materials, acrylic paint and siloxane (backbone of silicone), as mediums/binder for thick films. The actual composition of the cool roof coating is proprietary but it is safe to assume the volume fraction of particles is between 38 - 79% which is higher than most randomly distributed particles in passive radiative cooling research. A modest amount of spectral selectivity by cool roof coatings was observed in Fig. 3 which can be attributed to some common reflective and somewhat spectrally selective compounds like TiO₂, SiO₂, and CaCO₃ used in radiative cooling as well. The absorbance spectrum for the previously mentioned common compounds in Fig. 1 below show where the absorbance peaks are in relation to the atmospheric window (depicted by the rectangular box) in frequency range of 769 cm⁻¹ to 1,250 cm⁻¹ and wavelength range of 8-13 μm. (Wiley, 2021).

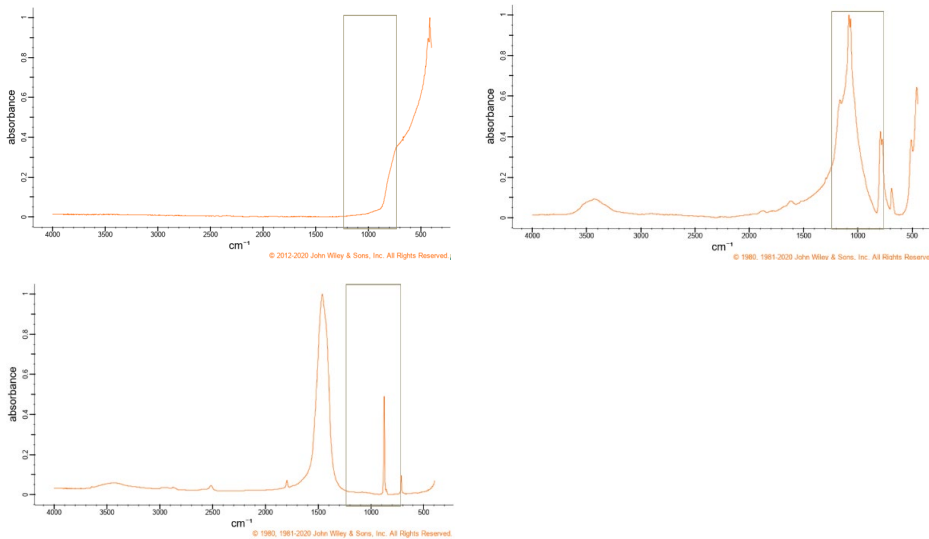


Fig. 1: Infrared absorbance peaks of (a) TiO₂, (b) SiO₂, and (c) CaCO₃ in relation to the atmospheric window (depicted with rectangular box)

In Fig. 1, TiO₂, SiO₂, and CaCO₃ show low absorbance across the infrared spectrum which explains favorable cooling properties, while the most spectrally selective compound is SiO₂. Kirchoff's law states that spectral absorptivity

and spectral emissivity of an object in thermal equilibrium are equal, for every wavelength and direction. Since SiO₂ absorbs more in atmospheric window it stands to reason according to Kirchoff's law that SiO₂ will emit more radiation through the atmospheric window. To improve upon cool roof technology, this research examined nanoparticle-based and thick film "radiative cooling" experimental studies - some are summarized in Tab. 2.

Tab. 2: Experimental studies of nanoparticle-based thick film radiative coolers

Experimental work references	Cooling Power (W m ⁻²); $\Delta T = T - T_{amb}$	Substrate / Binder (matrix)	Type and size materials nano(nm)/micro (μm) in film medium	Total Thickness	Volume Fraction (%)
Bao et al. (2017)	$\Delta T = -5$ °C night	Aluminum/ Double layer acrylic resin	Top layer - rutile TiO ₂ Bottom layer – SiO ₂ + SiC radius = 0.5 μm	>10 μm	4%
Huang and Ruan (2017)	100 W m ⁻² daytime 180 W m ⁻² nighttime	Aluminum/ Double layer acrylic resin	Top layer - TiO ₂ spheres Radius = 0.2 μm Bottom layer - carbon black	500 μm	4%
Zhai, Ma et al. (2017)	93 W m ⁻² daytime	Silver substrate /polymethylpentene (TPX)	SiO ₂ spheres radius = 4 μm	50 μm	5%
Gonome et al. (2014)	$\Delta T = -10$ °C (coated paper - paper)	Black and White Paper/ Acrylic	Copper (II) oxide (CuO) micro-particles 0.05, 0.89 and 1.9 μm	20-60 μm	3-5%
Chae et al. (2021)	$\Delta T = -2.8$ °C	Polyethylene terephthalate (PET)/ dipentaerythritol penta-hexa-acrylate (DPHA)	Al ₂ O ₃ , SiO ₂ , and Si ₃ N ₄ layers 19, 36, and 56 nm, respectively	NP layers were 2.0 - 2.1 μm thick	95%
Li et al. (2020)	37 W m ⁻² $\Delta T = -1.7$ °C	Polyethylene terephthalate (PET) /acrylic paint	Rod-shape CaCO ₃ fillers length ≈ 1.9 μm and diameter ≈ 500 nm	177, 131, and 98 μm	60%

The biggest advantage of a good passive radiative cooling coating is that it can perform both day and night. Huang and Ruan (2017) claimed to achieve a daytime net cooling power of 100 W m⁻² and a nighttime cooling power of 180 W m⁻² with a 500 μm thick double layer acrylic resin coating embedded with 0.2 μm radius titanium dioxide particles in the top layer while the bottom layer contained carbon black over an aluminum substrate.

Bao et al. (2017) conducted an experimental study of coating combinations with commonly used radiative cooling compounds. The theoretical predictions of cooling 5 °C below ambient under direct solar radiation and 17 °C below ambient at night were not observed; their best daytime surface temperatures were a few degrees above ambient and at nighttime about 5 °C lower than ambient.

Gonome et al. (2014) placed a CuO coating (black in color) with particle volume fraction of 5% and a thickness of 10 μm on black paper; instead of comparing to ambient temperature they compared its temperature to non-coated black paper and found the coating was 10 °C cooler.

Another experimental study by Chae et al. (2021) reported 2.8 °C below ambient temperature for a 1:1:1 ratio mixture of Al₂O₃, SiO₂, and Si₃N₄ nanoparticles. This research also used compounds which exhibit strong absorption peaks in or near the atmospheric window and low absorption outside the window.

Recently, researchers Li et al. (2020) applied a 60% concentration of CaCO₃ in acrylic paint, without the use of spectrally selective nanoparticles, at a thickness of 177, 131, and 98 μm for solar reflectance of 95.1%, 93.4%, and 88.9%, respectively. The thicker film provided the highest reflectance. Their spectral analysis reveals good reflection in UV-VIS spectrum but lacking selectivity in infrared spectrum; nevertheless, a surface temperature 1.7°C below ambient during the day was reported.

The research review revealed the need to develop a practical, cost effective, scalable, and robust passive "radiative" cooling coating which reduces heat transfer to surfaces by reflecting a maximum amount of solar radiation (0.3-2.5 μm wavelength) while strongly emitting within primary atmospheric window (8-13μm wavelength) to the deep space.

3. Fabrication of Thick Film Nanocomposite

The objective of our radiative cooling research and development is the design and fabrication of a thick film nanocomposite coating containing spectrally selective nanoparticles with highest absorption in atmospheric window and very low absorption outside the window.

The fabrication concept in this research is akin to microelectronic thick film fabrication only in the sense that it's also an additive process, which entails the application of thicker paste layers and thinner ink layers upon substrate as needed. The layers are added sequentially to the substrate to create a thick film with the desired properties. Another departure is much lower energy for fabrication since high curing temperatures are not required. The resultant performance is dependent upon many factors like substrate properties, nanoparticle properties, medium properties, and climatic conditions.

For this research the thick film's thickness scale can be 1 μm to $\sim 600 \mu\text{m}$ consisting of ink and/or paste layers. A thin film's thickness is approximately 1000 times thinner than a 100-micrometer thick film. A thick film will be more robust and have an engineering tolerance to still provide radiative cooling despite variations in film thickness and surface properties.

The simple, cost-effective thick film coating application methods in this research are chemical solution deposition (CSD) and/or spray coating of a paste and ink layers upon a substrate illustrated (not to scale) in Fig. 2.

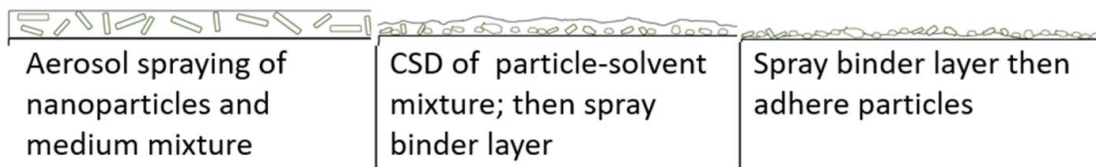


Fig. 2: Scalable, cost-effective thick film nanocomposite application methods with side-view drawing of particles and binder on surface
Note: not to scale

Combinations of these methods with or without paste layers (not shown in Fig. 2) can provide a novel direction for tailoring spectrally selective radiative cooling thick film nanocomposite coatings for colder climate locations to avoid increased heating loads during winter months.

A medium is the matter electromagnetic radiation travels through to reach nanoparticles. A coating medium not only encapsulates the spectrally selective nanoparticles and fillers, but also functions as a binder to facilitate adhesion of film to a multitude of surfaces. Some mediums/binders add favorable spectral selectivity properties, like acrylic's absorption bands within atmospheric window shown in Figs. 4 & 5, which enhance passive radiative cooling. Spectrally selective nanoparticles are chosen with varying levels of absorption within the atmospheric window and high levels of reflectivity outside the window. The nanoparticles chosen enable tuning of the cooling power of the thick film.

4. Characterization

The thick film nanocomposite was characterized using Ultraviolet – Visible (UV-Vis) spectroscopy, infrared spectroscopy by Fourier Transform Infrared (FTIR), Scanning Electron Microscope (SEM), and performance testing techniques.

The UV-Vis spectroscopy was performed with an Ocean Optics USB-2000 UV-Vis-NIR spectrometer equipped with an enclosed chamber. Tuning film for high reflectance within 0.3-2.4 μm waveband is essential for daytime radiative cooling due to the strong solar heat flux during the day (Naghshine and Saboonchi, 2018).

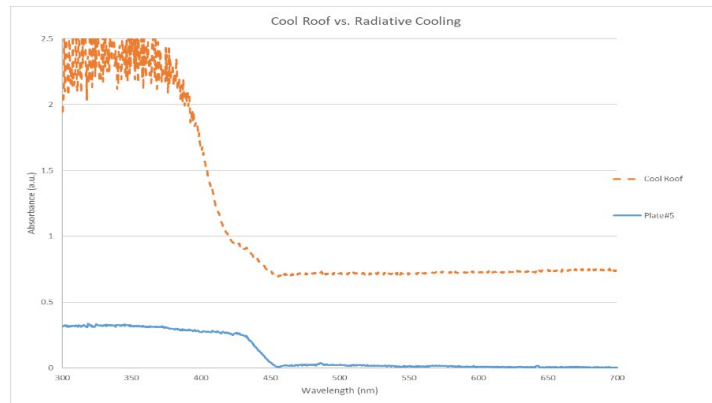


Fig. 3: UV-Vis spectrum comparison of cool roof coating (top) and thick film nanocomposite (bottom)

The UV-Vis absorbance spectrum graph in Fig. 3 analyzed two samples on aluminum substrates -- the cool roof coating (top orange line) the radiative cooling thick film nanocomposite (bottom blue line). The strong ultraviolet light absorbance of cool roof coatings is significantly more than thick film nanocomposite and ironically due to white pigments like titanium dioxide (Kolokotsa et al. 2013). Tuning the thick film for lower UV absorbance not only increases cooling power, but also extends the life of the coating and surface beneath, since the higher energy UV radiation can readily degrade many materials.

Since the primary atmospheric window is within the infrared spectrum this research concentrated on infrared spectroscopy to obtain desired spectral selectivity. Infrared spectroscopy was accomplished with a Jasco FTIR-6300 spectrometer and a Pike 30spec specular reflectance attachment with a variable aperture designed for the measurement of thick films held the samples. Maximization of absorption in the atmospheric window (illustrated with rectangular boxes in spectra graphs) in Figs. 1, 4, 5, 6, 8, and 9 is an objective in this research to obtain high radiative cooling levels.

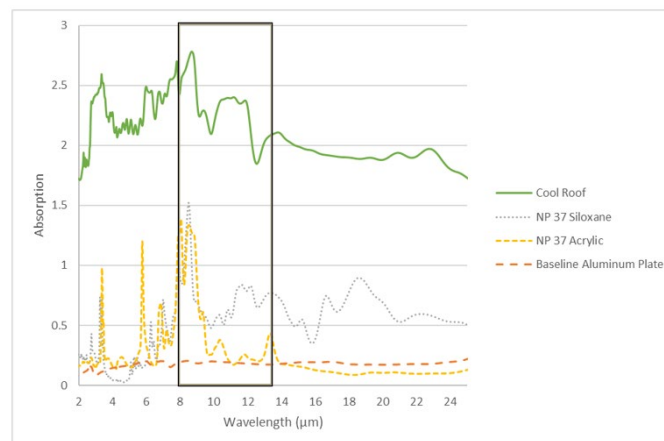


Fig. 4: FTIR absorption spectra of a baseline aluminum plate, two nanoparticle samples (Batch 37) one coated with acrylic binder and another with siloxane binder, and cool roof coating all on aluminum plate substrate

In Fig. 4 the same batch of spectrally selective nanoparticles on aluminum foil produced different infrared spectrums with different mediums. The siloxane acrylic binder increased absorption – a change in nanoparticles with Acrylic binder on aluminum foil improves spectrally selective absorption.

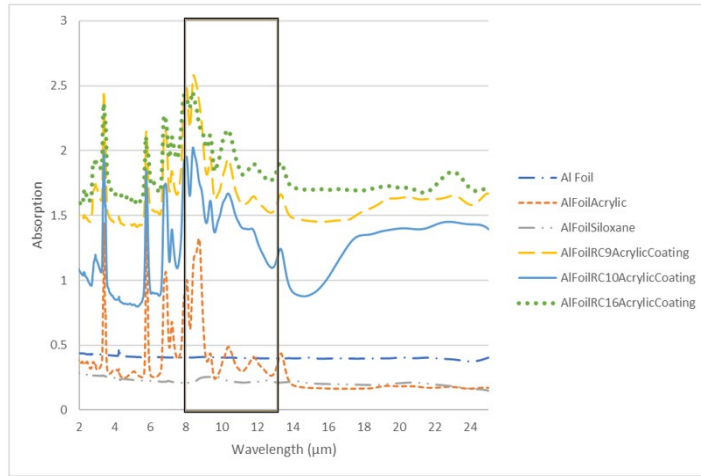


Fig. 5: FTIR absorption spectra of baseline aluminum foil and with selective nanoparticle batches RC 9, RC 10, and RC 16 coated by acrylic binder on the baseline aluminum foil.

Spectrally selective nanoparticles will increase absorption; however, the spectrum shape remains similar to the medium. The varying material properties of the nanoparticle, with the known variable of differing absorption levels in the atmospheric window, produced a range of absorption levels. Experimental results in Figs. 4 and 5 show that the absorption peaks closely follow the spectrum of acrylic medium, however, when the nanoparticles are integrated, there is an increased absorption within the whole spectrum and the more spectrally selective the nanoparticle the more influence it will have in making the nanocomposite film more selective. For example, in Fig. 5 since the RC 10 nanoparticle's spectrum is more selective than the RC 9 and RC 16 nanoparticles that thick film is more selective.

A cool roof coating, with recommended thickness of 22mil (558.8 μm), is regarded as a paste layer thick film with high reflectivity fillers. The fillers in a paste layer can prevent transmission of solar radiation to the substrate and limit heat transfer. If the substrate is not reflective, a reflective bottom layer and/or a reflective and/or selective cover or ink layer is added to obtain radiative cooling. In Fig. 4 the degree of absorption and selectivity vary with the medium used for nanoparticles – here acrylic medium shows selectivity and higher absorption within the atmospheric window.

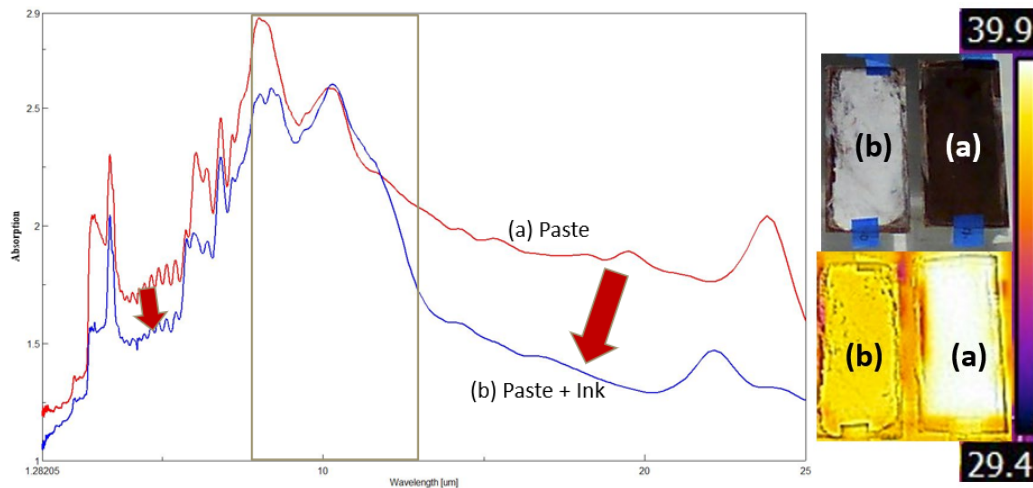


Fig. 6: FTIR absorption spectra (left) and photograph (right top) thermal images (right bottom) for (a) Paste over aluminum plate substrate (b) Paste with TiO₂ ink layer on top over aluminum plate substrate; red arrows identify increased reflectivity. The thermal images (right) with temperature scale (white is hottest).

A nanocomposite paste in Fig. 6, comprised of SiO particles and spectrally selective nanoparticles was formulated to maximize absorption in the atmospheric window while achieving some degree of selective emittance. The paste layer had an absorption peak of 99.9% within the atmospheric window; while the overall infrared emissivity between 6 to 10 μm was greater than 99.875%, where less than 0.125% of solar radiation is transmitted to substrate. Despite an absorption level over 99% and an absorption peak in the atmospheric window the paste layer didn't exhibit cooling properties because the absorption outside the window was still too high. Then a TiO₂ ink layer placed over the paste, in Fig. 6b, increased selectivity by reducing absorption outside the atmospheric window (red arrows point out change) on both sides of the atmospheric window. The TiO₂ layer slightly reduced the absorption peak in the window increased cooling was observed in the thermal imagery for an estimated 4°C surface temperature reduction. According to Wiley Spectrabase (2021) TiO₂ has its infrared absorption peak outside the atmospheric window at a wavelength of 20 μm or frequency of 500 cm^{-1} , suggests a more spectrally selective nanoparticle ink layer on top of the paste might increase selectivity and cooling to a greater degree.

The Scanning Electron Microscope (SEM) analysis of thick film samples were performed by a Hitachi S800 or a Hitachi SU70 SEM. The sample analysis provides insight into the nanoparticle size, morphology, and spacing in the nanocomposite.

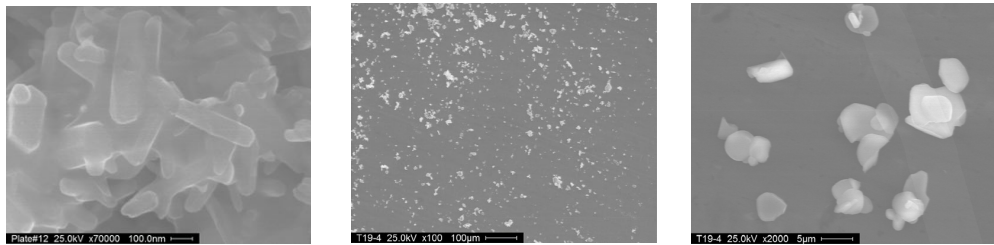


Fig. 7: SEM micrographs (a) Surface morphology of CSD of solvent and nanoparticle mixture with spray coated binding layer (b) Surface of 47 μm thick film nanocomposite ink layer shows random dispersion of spectrally selective nanoparticles in siloxane binder (c) Magnification of thick film nanocomposite ink layer's spectrally selective nanoparticles, size range 4-6 μm

The surface of nanocomposite films in Fig. 7 reveal randomly distributed spectrally selective nanoparticles and particles spaced 5-10 μm apart with slight agglomeration. Agglomeration will occur in coatings with randomly dispersed particles in a medium and applied by cost effective means. Because of this larger range of particle sizes and agglomeration absorption and reflectance is enhanced throughout the spectrum also in part because of the spectral selectivity of the materials. Nevertheless, both films exhibited spectrally selective properties, because of the spectral selectivity of the binder and nanoparticles

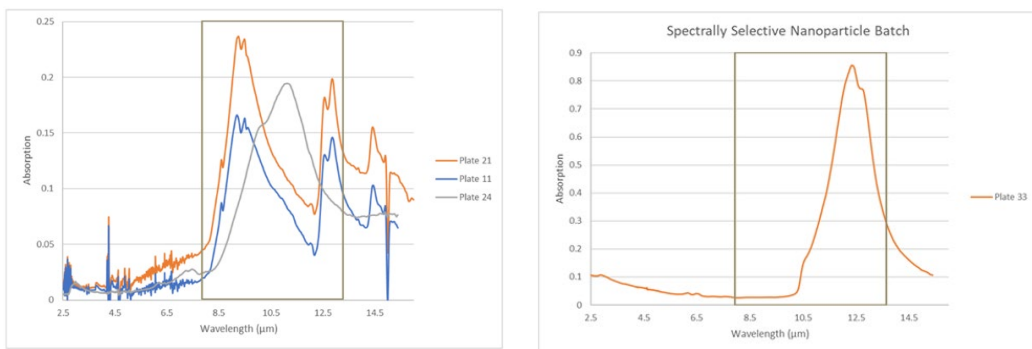


Fig. 8: Infrared spectrum of spectrally selective nanoparticles: (a) Plates 11, 21, and 24 and (b) Plate 33 in Fig. 9 whose cooling levels exceeded that of cool roof coating.

The spectrally selective nanoparticles in Fig. 8 had varying levels of absorption in the atmospheric window with Fig. 8b having the highest levels. In Fig. 9, thermal imagery using FLIR “forward looking infrared radiometer” infrared

camera and a photograph of plates are shown.

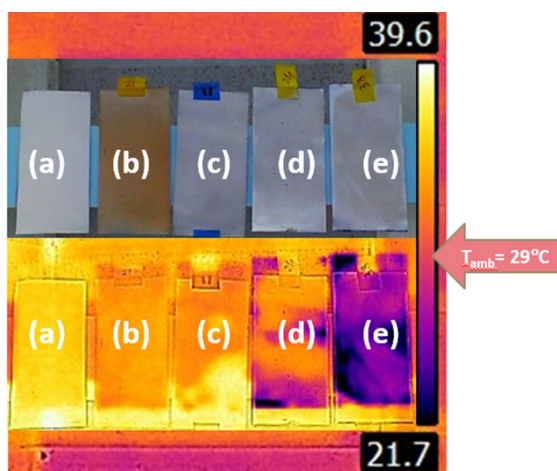


Fig. 9: Photograph of plates (top) and thermal imagery (bottom) with color temperature scale to the right; Coated plates from left to right: (a) Cool roof, (b) Plate 21 (c) Plate 11 (d) Plate 24 (e) Plate 33

Ambient conditions for testing were 29°C, 78% humidity, and wind of 8 mph. The side-by-side comparison of thick film coatings and a cool roof coating on aluminum substrates in Figs. 4, 5, and 9 provide a positive indication that the thick film's cooling power can be significantly higher than cool roof coating. The samples on aluminum plates are cool roof coating (Fig. 9a) – a 440 μm thick paste layer. Nanoparticle and siloxane mixture sprayed on aluminum substrate for Plate 11 (Fig. 9c) - 47 μm thick w/5% particle #37 and Plate 21 (Fig. 9b) - 40 μm thick w/5% particle #39. A chemical solution deposition of siloxane mixture for Plate 24 (Fig. 9d) - 58 μm thick w/5% particle #10 and application of nanoparticles adhered to acrylic binder surface for Plate 33 (Fig. 9e) ~30 μm thick w/5% particle #44. The lighter color on the bottom on Fig. 9c is feedback from the thermal camera. Siloxane binder on aluminum foil had lower absorption than non-coated aluminum and acrylic without nanoparticles in Fig. 5; and when particles were added to siloxane the resulting spectrum did not have the same amount of selectivity as the acrylic binder in Fig. 4. Nevertheless, the cool roof coating was above ambient temperature while the thick film nanocomposite ink layers whether in siloxane or acrylic paint were still below ambient. Also, this experiment demonstrated the ability to tune the emissivity of the thick film nanocomposite by varying medium and spectrally selective nanoparticles.

5. Conclusion

This research experimentally demonstrated a thick film nanocomposite coating fabricated from spectrally selective nanoparticles in complementary mediums over an aluminum substrate can lower surface temperatures more than a leading cool roof coating over an aluminum substrate. Also, a thick film nanocomposite placed on a building in cold climates can avoid heating load penalties by tuning cooling power with nanoparticles offering less spectral selectivity and absorption in the atmospheric window. The passive cooling by cool roof coatings can be attributed to some common compounds found in radiative cooling as well, but the cooling power is limited by its lack of spectral selectivity. The thick film nanocomposite coating combines cost-effective, scalable, and robust properties of a cool roof coating with a higher spectral selectivity and higher thermal emittance through the atmospheric window to offer an alternative with greater cooling power. Theoretically, if a thick film nanocomposite coating with a cooling power of 100 W m^{-2} covered 2% of the earth's surface, approximately 10 million square kilometers, it could emit enough thermal radiation to balance the global heat flux.

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