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The Effect of Interspersed Nanoparticles on Long Wavelength Heat Radiation through Opaque and Transparent Passive Skylight Glass

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Abstract **-** The materials of passive radiant cooling (PRC) skylights play a critical role in achieving the PRC performance required for building cooling. However, finding window materials that provide a cooling effect at an affordable price remains a challenge, although it is a prerequisite for widespread market acceptance. Research conducted at Åbo Akademi University led to the development of a passive cooling skylight technology that enables higher heat transfer rates by combining thermal convection and thermal radiation (Qconv > > Qcond) instead of thermal conduction and thermal radiation. This prototype enabled significant PRC during the night, but extending its use to PRC during the day presents a greater challenge. A coating of dispersed TiO_2-SiO_2 or ZnS-SiO_2 nanoparticles (NPs) was used on simple window glass (WG) and on long-wavelength (LW) translucent Cleartran® ZnS glass to control the surface temperature (TS) and [absorptivity (α)/emissivity (ε)] at different heat source temperatures. These NPs were selected for their chemical, optical, and thermal properties, which make them suitable for a wide range of applications, including radiative cooling. By capturing IR images with a thermal imaging camera (wavelength range 7.5-14 µm), heat flux measurements were experimentally performed for both window and Cleartran® glasses with NPs randomly distributed on one side of the glass surface. The amount of NP (in g/m^2) and surface coverage by NPs (in %) were determined by SEM analysis of the window samples in this work. The results showed that the average amount of TiO_2-SiO_2 NP and ZnS-SiO_2 was 0.25 mg/m_2 and 0.3 mg/m^2, respectively, and the surface coverage was ≈ 8% for TiO_2-SiO_2 NPs and ≈ 15% for ZnS-SiO_2 NPs. Increasing the surface coverage of NPs and optimizing the interspersed NPs could still increase the net cooling performance of a skylight to be effective for daytime use. While for simple glass the heat flux is increased by a factor of 2 to 4, for the Cleartran® glass the change is only a few %. However, this work needs to be extended to shortwave radiation (SW) for further investigation to verify the performance of the material under solar irradiation.

Keywords**:** Radiative transfer, Atmospheric window (8-13 µm), Skylight, Daytime passive radiative cooling, TiO_2-SiO_2 NPs, ZnS-SiO_2 NPs, Absorptivity, Emissivity.

1. Introduction

The need for the cooling of buildings is anticipated to rise in many regions as a result of the higher temperatures brought on by climate change. In order to maintain pleasant indoor conditions, air conditioning and cooling systems in buildings may be used more frequently during warmer summers and prolonged heatwaves. The heat gain of the roof, which accounts for 70% of the overall heat gain, aggravates this issue. Passive cooling techniques are innovative practices that aim to provide comfortable indoor conditions in buildings using natural processes, without relying heavily on mechanical or active cooling systems. These techniques can help to reduce energy usage and greenhouse gas (GHG) emissions associated with cooling, as well as to promote sustainable and environmentally friendly building design. Both reflective and radiative processes can be used in a combination to reduce overall heat gain by facilitating the expulsion of excess heat from a building's interior to maintain a comfortable environment. Reflective processes involve reflecting solar irradiation effectively in the short wavelength (0.2-3 µm) away from the building's interior, while radiative cooling processes involve releasing heat from the interior to the (upper) atmosphere and space (at 3 K) through the atmospheric window (8-13 µm) wavelengths [1]. The research at Åbo Akademi University (ÅAU) on radiative cooling since 2007 is innovative and promising, combining heat convection and thermal radiation rather than heat conduction and thermal radiation, using a skylight roof window. The use of such a combination of heat transfer mechanisms in the passive skylight can for instance be used in residential or commercial buildings where effective cooling strategies are needed. The first ÅA prototypes showed substantial PRC of around 100 W/m^2 at night [2]. It is, however, more challenging to realise PRC also during daytime. A radiative cooling coating incorporating NPs is presently thought to be the most practical method, also given the simplicity of its manufacturing methods and straightforward design. The glass material used for the ÅA skylight so far is Cleartran® (ZnS). Cleartran® was chosen due to its excellent optical properties, primarily the high transmittance ($\tau = 0.75$) for long wavelength (LW) heat radiation, reasonable cost, and availability in large quantities. This work compares the results obtained when different NP materials are randomly dispersed on different substrates of simple glass window as well as on Cleartran®. For this study, TiO_2-SiO_2 and ZnS-SiO_2 NPs were used for the tests, and their radiative cooling performance was investigated experimentally. The major role of these NPs is to absorb a considerable part of the solar light that reaches to the top of a window and re-emit it as infrared radiation. This enables the window to remain at a low temperature, largely blocking the passage of short wavelength (SW) solar irradiation while, in the case of Cleartran®, allowing LW radiation to pass.

1.1. PRC characteristics of TiO_2-SiO_2 /ZnS-SiO_2 NPs

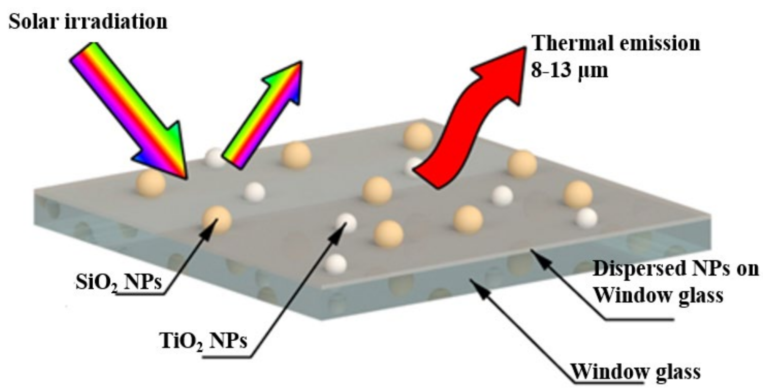

Fig. 1: Radiative cooling coating with mixture of TiO_2 or ZnS particles together with SiO_2 particles.

The current work focuses on materials that can limit the (SW) solar irradiation into a building through a passive cooling skylight while it is at the same time sending LW thermal radiation out of the building. TiO_2-SiO_2 and ZnS-SiO_2 composite NP materials have unique properties that make them promising for passive cooling applications, such as skylights and cool roofs. Some key PRC characteristics of these NPs are discussed below:

High refractive index (RI): TiO_2 and ZnS both have a high RI of approximately ≈ 2.59 and ≈ 2.42, respectively [3], making them suitable for optical applications such as thin-film coatings, optical bandpass filters, and waveguides. When these materials are combined with SiO_2, which has a lower RI of ≈ 1.45 [4], the resulting NPs can have a tunable RI, which can be useful for controlling the propagation of daylight in passive cooling transmittive skylights such as ÅA skylight. This combination of NP materials can also be used to create nanoparticle coatings that can reflect IR radiation.

Transmittance**:** The TiO_2-SiO_2 coating is generally known for its lower transmittance in the UV and visible light spectrum, which reduces solar heat gain and prevents UV light from entering the interior. ZnS-SiO_2 coatings also reduce glare by minimizing the reflection of light at the surface, which allows more light to pass through the skylight. They also reduce solar heat by reducing the amount of solar energy entering the building.

Reflectance: TiO_2 and ZnS NPs can both contribute to greater reflectivity in the SW area, due to their strong UV absorption characteristics. SiO_2 NPs are transparent in the visible spectrum, thus they have less of an impact on reflection. Both composites are unlikely to have a major impact on LW reflection due to their absorption qualities. These NPs, however, are expected to have high emission in LW region. It is important to note that the actual absorption, emission, and reflection properties of these NPs are affected by a variety of parameters including NP size, concentration, and layer thickness [5].

One significant criterion for the deposited NPs on the skylight top window (see below) is that the transmittance of LW heat from the building space below should not be compromised while enabling sufficient visible light to enter during the day.

2. ÅA Transmittive Skylight as a Passive Cooling Concept

A sideview impression of the passive cooling skylight developed and put through proof-of-concept testing at ÅA is shown in Figure 2a. The PRC capability is comparable to that of roof paint or other covering materials that emit strongly in the wavelength range (8-13 μm) i.e., the atmospheric window, as illustrated in Figure 2b, which when partly covered with NPs 1) reflects (much) solar irradiation, 2) emits at its own temperature and 3) lets out passive cooling radiation from the building space below the skylight. A participating greenhouse gas (GHG) transfers the heat inside the skylight. In comparison to conductive heat transfer (Q_{cond}) through a building roof, the skylights benefit from a higher heat transfer rate (per square meter) from a building envelope to the outside surface, controlled by convective circulation (Q_{conv}) inside the skylight.

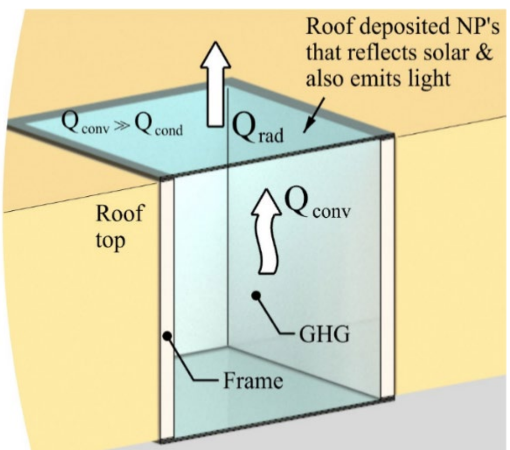

Fig. 2a: Sideview: Passive cooling skylight inside roof

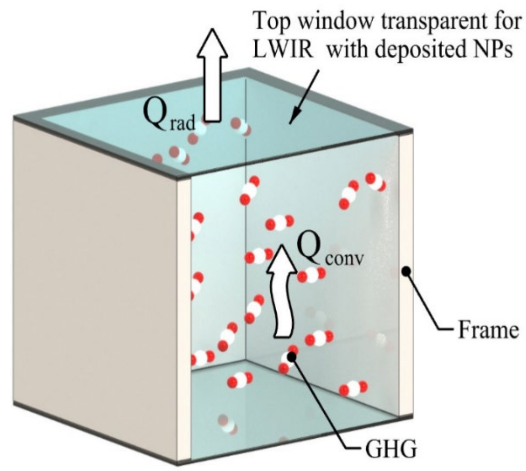

Fig. 2b: A skylight design with dispersed NPs on Cleartran®

With the intention of improving the cooling performance of the ÅÅ skylight roof window while effectively reflecting solar radiation and emitting own surface heat, this work presents the concept of low-cost and easily scalable TiO_2-SiO_2 and ZnS-SiO_2 NPs for an investigation of LWIR thermal radiation. It compares Cleartran® and plain window glass with dispersed NPs on a passively radiating roof cooling system for daylighting application. The purpose of this study was to compare the thermal performance of various window glasses when facing a heat source at various temperatures (303 K, 313 K and 333 K). Using a (LW) heat camera, the heat flow from the backside of a window, with the front facing the heat source was measured 1) without NPs, 2) with NPs on the front side or 3) with NPs on the backside. Heat transmittance was possible with Cleartran® but not with the simple window glass. Further, for the produced glass substrates, the NPs size, the amount of deposited NPs, and area fraction coated with NPs were determined. Therefore, this data can be used in future studies when glass surfaces with deposited NPs are used to investigate the performance under SW (solar) irradiation, and eventually for improving the passive cooling performance of the ÅA skylight.

3. Mathematical Model for Calculating Radiative Heat Flux

This section presents the mathematical model equation used to determine the heat flow (W/m^2) from the (backside) of the glass substrates (simple window and Cleartran®) with or without deposited TiO_2-SiO_2 and ZnS-SiO_2 NPs. Referring to Figure 3, the mathematical model allows the calculation of the heat flux ($Q_{2\text{-}3}$) between the glass substrate and the thermal imager based on a set of parameters. The radiative heat transfer from surface A_2 (glass substrate) to surface A_3 (the heat camera) can be determined by the equation is shown below (see equ.1) [6]:

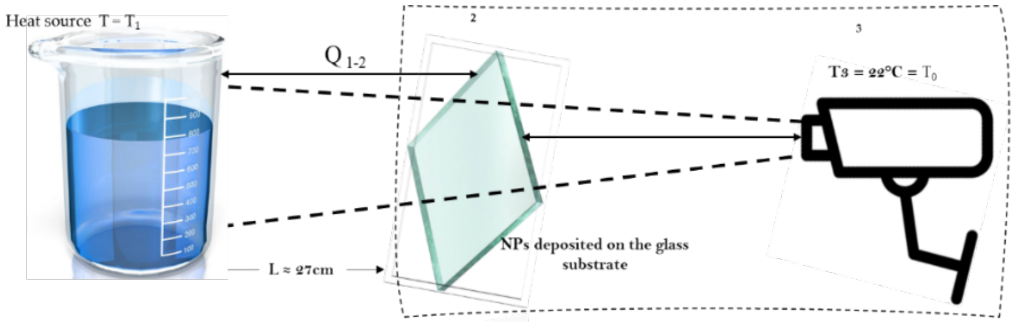

Fig. 3: Experimental setup for determining the radiative heat transfer from a glass substrate to a camera.

$$Q_{2-3} = \frac{\sigma.(T_2^4 - T_3^4)}{\frac{1-\varepsilon_2}{\varepsilon_2}.\frac{1}{A_2} + \frac{1}{A_2.F_{2\to3}} + \frac{1-\varepsilon_3}{\varepsilon_3}.\frac{1}{A_3}} \qquad (Watt) \tag{1}$$

The Stefan-Boltzmann constant σ=5.6704×10^{-8} W/m^2·K, T_2 and ε_2 are the unknown variables in the preceding equation. T_2 stands for the glass substrate's surface temperature (Ts), and ε_2 is the glass substrate's surface emissivity, respectively. $T_3 = T_0$ is the ambient (laboratory) temperature, which is the same as the temperature of a thermal camera. The surface area of the glass substrate is A_2 (0.05 m × 0.05 m = 0.0025 m^2), and the calculable view factor is $F_{2\to3} = 0.08726$. The estimated value $\varepsilon_2 = \varepsilon^* = 0.95$ provided by the thermal imager software, which implies T_2^*, an estimate for but not the real value for T_2. Here, $\varepsilon^* = 0.95$ is used for thermal images. Finally, $\varepsilon_3 = 1$ is the emissivity of the thermal imager. Nonetheless, using a value for ε* which gives an estimate T_2^* for T_2 allows for determining the heat flow.

$$Q_{2-3} = \frac{\sigma.(T_2^{*4} - T_0^4)}{\frac{1-\varepsilon^*}{\varepsilon^*}.\frac{1}{A_2} + \frac{1}{A_2.F_{2\to3}}} \qquad (Watt) \tag{2}$$

Equation (2) can be used to determine heat flows Q_{2-3} at T_1 = 303 K, 313 K, and 333 K for all glass windows. The heat flows obtained are then used to calculate the heat flux through each window sample.

4. Results and Discussions

4.1. Production of window glass samples for experimental tests

Simple window glasses were coated with TiO_2-SiO_2 NPs using a dip coating approach [7] and are called WG 1 NP Hot, WG 2 NP Hot, WG 3 NP Hot, and WG 4 NP Hot, as well as WG1 NP Cold, WG 2 NP Cold, WG 3 NP Cold, and WG4 NP Cold, respectively. NP Hot denotes IR images taken with NPs facing towards the hot surface (at T_1), whereas NP Cold denotes IR images captured when NPs face the thermal camera. The procedure was repeated for ZnS-SiO_2 NPs, followed by Cleartran® (ZnS) glass with and without NPs on it. In total, test was done with one plain window glass with no particles, four TiO_2-SiO_2 on simple glass samples, four ZnS-SiO_2 on simple glass samples, a Cleartran® glass sample, and two Cleartran® glass samples with both types of NPs.

4.2. Analysis of the NPs Coverage Area and Particle Size on Window Glass

Window glass samples with NPs were characterized using SEM (see Figs. 4a and 4b) to determine how the particles were dispersed randomly across the glass surface. Image analysis (ImageJ software) was used to determine the NP coverage on the glass surface. The requirement for this application is that visible light (0.4 - 0.9 µm) can pass through it without being interrupted. TiO_2, SiO_2, and ZnS have primary particle sizes of around 25 nm, 30 nm, and 140 nm, respectively, for the purpose of producing nanocoating's. The average particle (cluster) size and area covered by the NPs lying on the glass surface can be estimated from SEM images. Particle aggregate sizes of the TiO_2-SiO_2 sample averaged

0.4 µm, while the $ZnS-SiO_2$ aggregates averaged 0.1 µm. The particles are somewhat aggregated on the glass surface, and this might influence the scattering performance on the skylight, i.e., decrease the quality of the light that is being transmitted. Aggregated particles can also cause a higher level of reflection, further decreasing the amount of light passing through. These effects need to be evaluated in order to optimize the performance of the skylight.

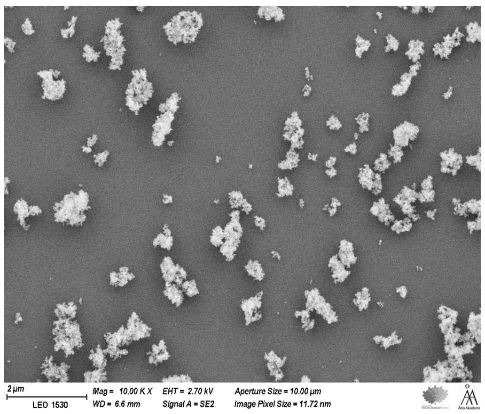

Fig. 4a: SEM images of TiO_2-SiO_2 NPs on glass substrate

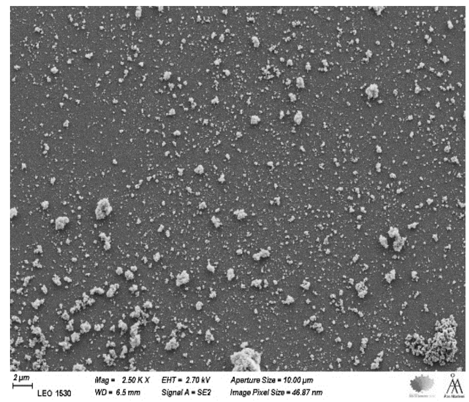

Fig. 4b: SEM images of $ZnS-SiO_2$ NPs on glass substrate

The surface coverage area of the NPs is estimated to be 8.0% for the TiO_2-SiO_2 glass sample and 15.8% for the $ZnS-SiO_2$ glass sample, respectively. Quantitatively determined NP mass for TiO_2-SiO_2 NPs on the WG1, WG2, WG3, and WG4 samples are 0.2 mg, 0.3 mg, 0.3 mg, and 0.4 mg, respectively. Similarly, for $ZnS-SiO_2$ NPs, masses on WG1, WG2, WG3, and WG4 are 0.2 mg, 0.2 mg, 0.3 mg, and 0.3 mg, respectively. Due to mass balance limitations, all weighted NPs have an accuracy of ± 0.1 mg.

4.2. Heat Flux (Q 2-3) Comparison for Two Types of NP Composites at Different Temperatures

Although all the window glasses were made using the same process, the heat fluxes obtained for each glass are different. This is due to the fact that the amount of NPs present on window glasses varies from one another. Apart from affecting heat absorptivity and emissivity, the amount of NPs on the glass surface affects the reflectivity and transmissivity of the glass. This results in different heat fluxes passing through each glass.

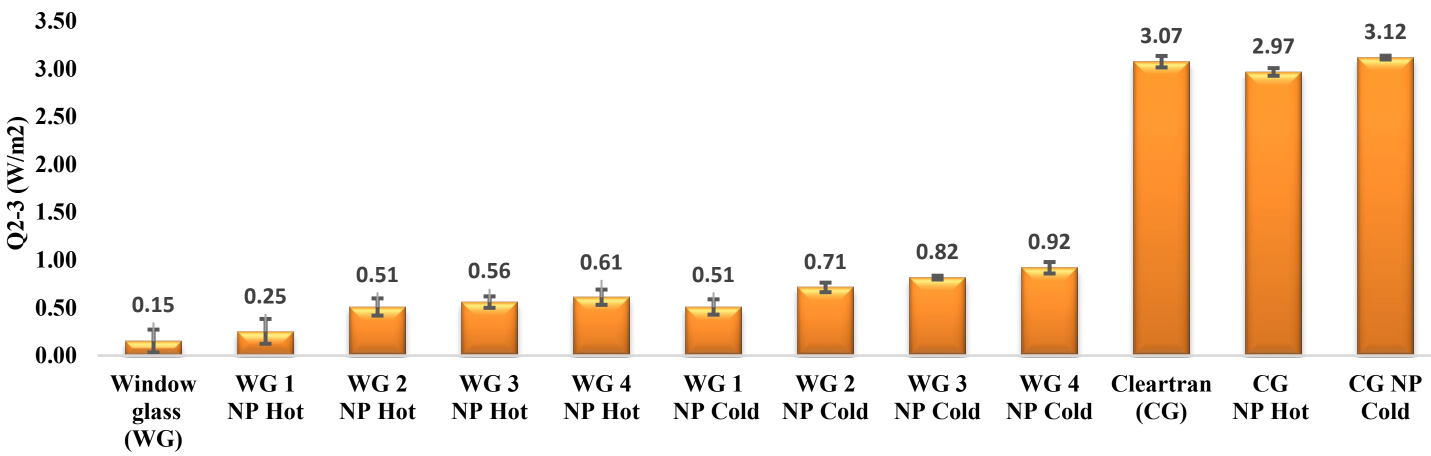

Fig. 5a: The comparison of the obtained heat flux for TiO_2-SiO_2 NPs at 303K

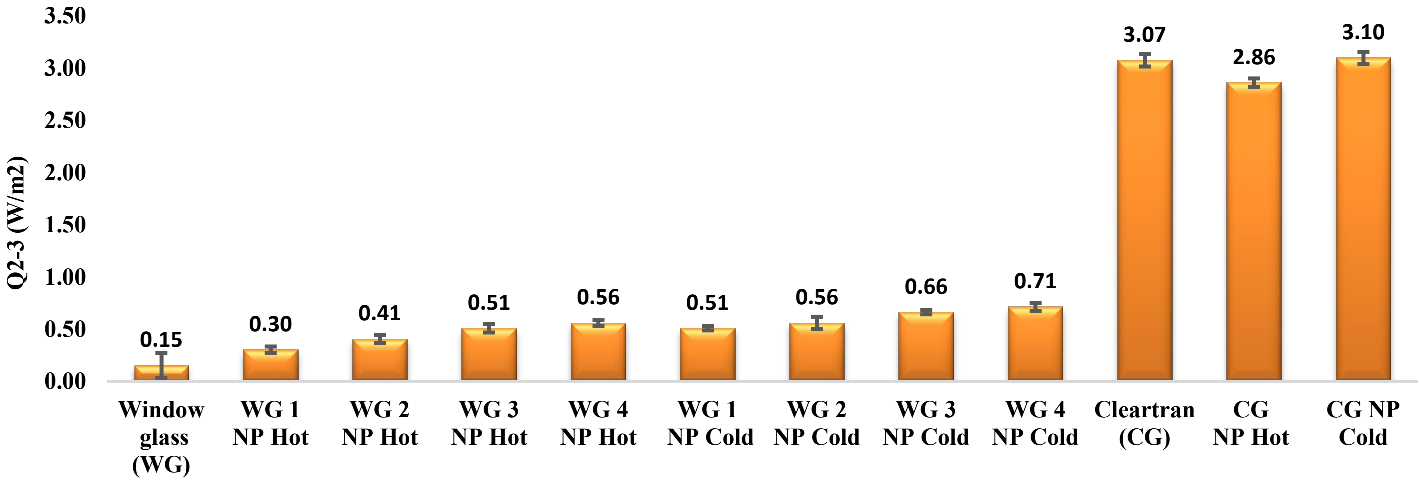

Fig. 5b: The comparison of the obtained heat flows for ZnS-SiO_2 NPs at 303K

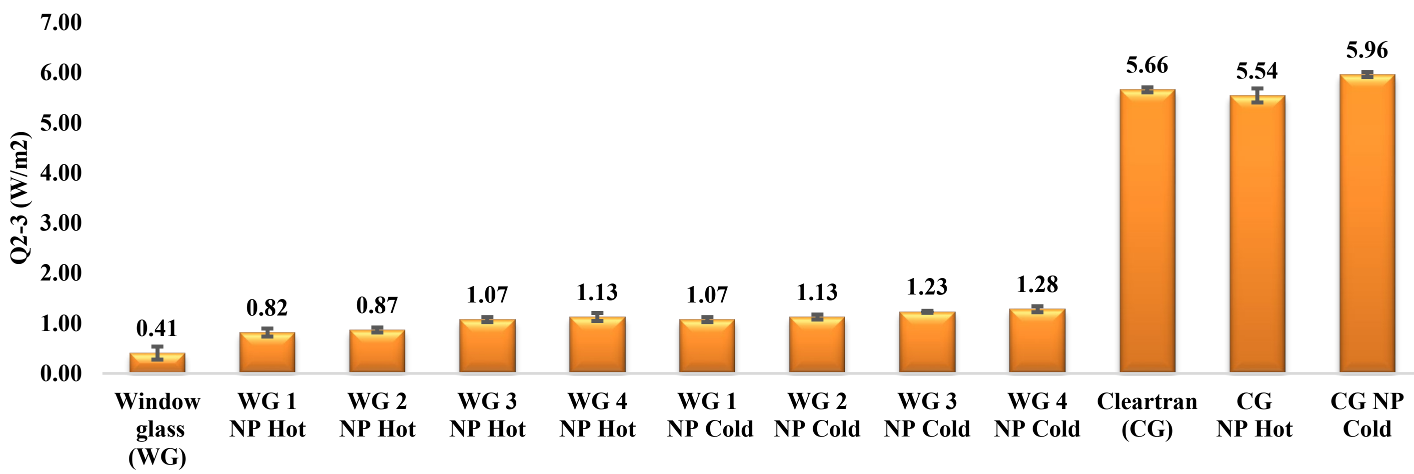

Fig. 6a: The comparison of the obtained heat flux for TiO_2-SiO_2 NPs at 313 K

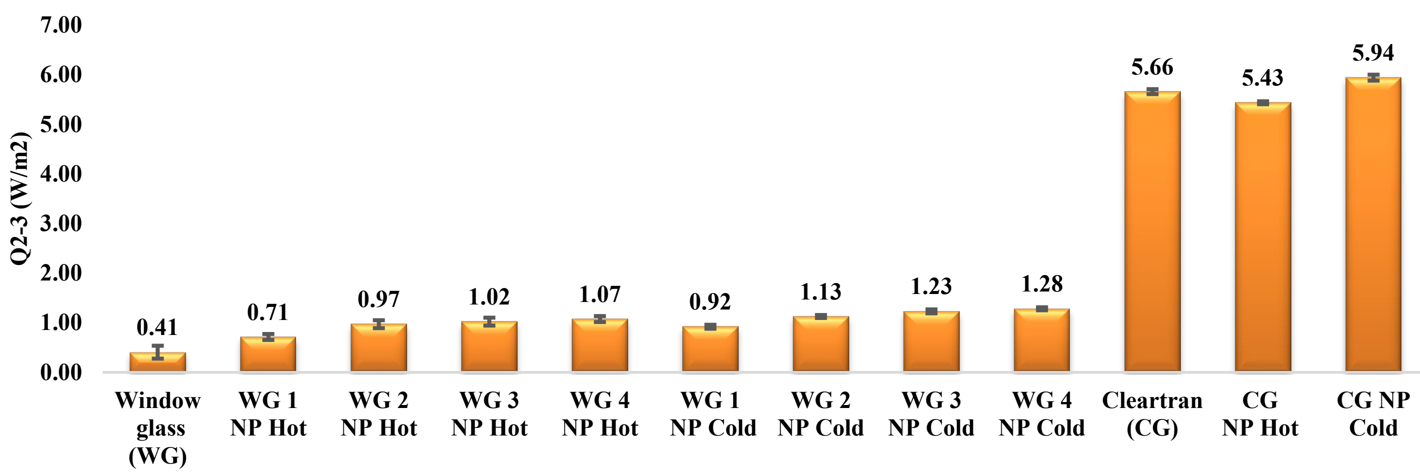

Fig. 6b: The comparison of the obtained heat flux for ZnS-SiO_2 NPs at 31

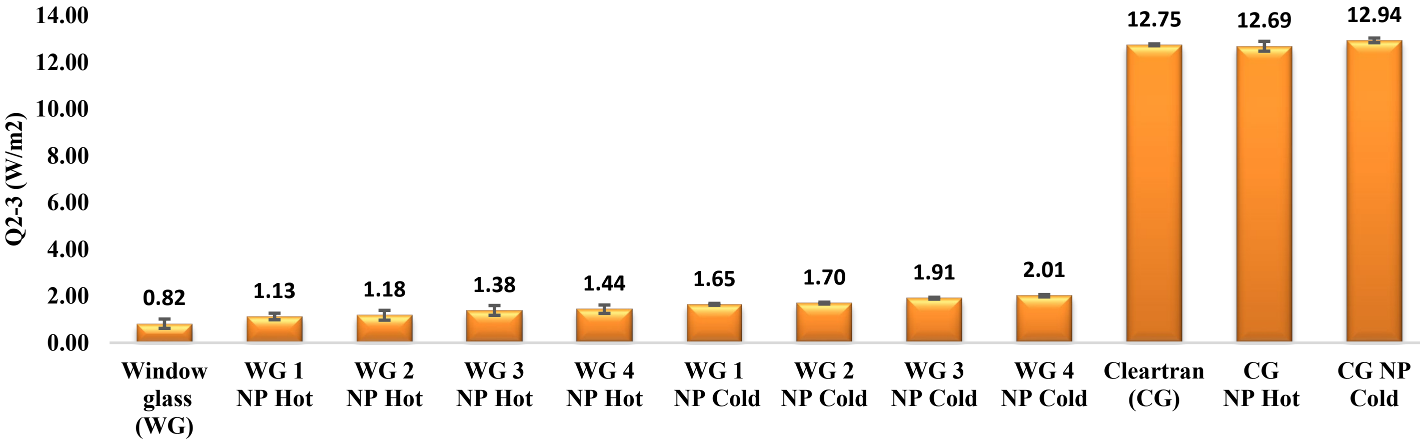

Fig. 7a: The comparison of the obtained heat flux for TiO_2-SiO_2 NPs at 333 K

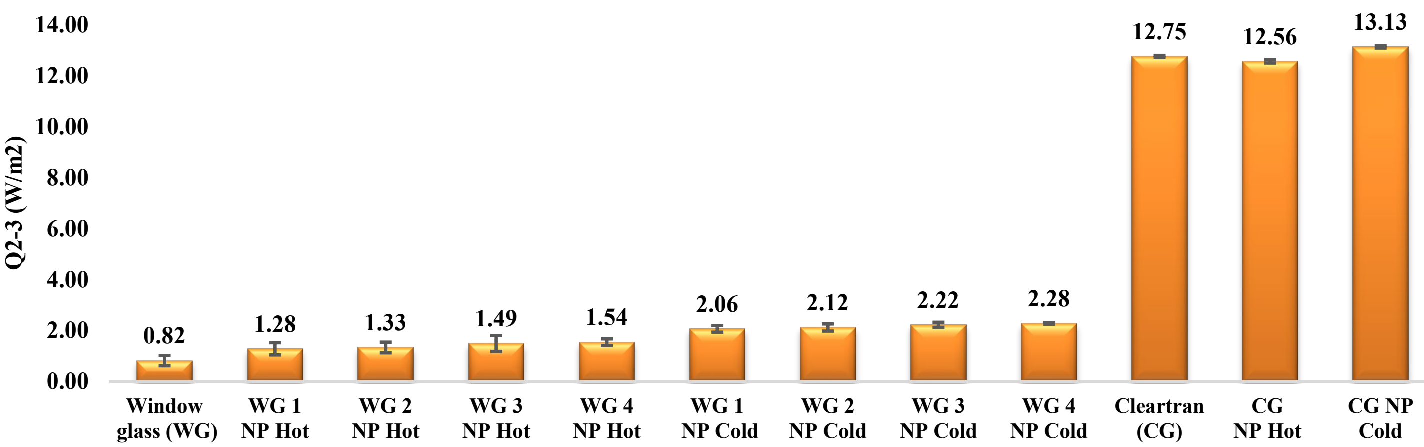

Fig. 7b: The comparison of the obtained heat flux for ZnS-SiO_2 NPs at 333 K

The results showed that the NPs on the backside of the window glass (NP Cold) provided better heat flux than the NPs on the front side (NP Hot). The heat fluxes were increasing with heat source temperature t 333 K > 313 K > 303 K. An interesting observation from the above results is that the heat fluxes of substrates with NPs deposited on the side facing the thermal camera IR (NP Cold) are larger than those of substrates with NPs facing the heat source (NP Hot). Apparently, the NPs facing the heat source reflect heat away from the camera, while the NPs on the other side do not reflect heat away but are heated by conduction through the glass (note that plain window glass cannot transmit LW radiation from the heat source). For the Cleartran®, the LW transmittance of 75% does give an additional heat flow equal to $[\tau/(1-\tau) * Q_{1-2}]$ through the substrate, reaching the camera together with Q_{2-3}, also described by model equation like (1) for Q_{1-2}. Heat fluxes for Cleartran® glass showed that more heat transfer is possible through Cleartran® glass without NPs compared to the NP Hot side, i.e., the LW heat flux decreased because the NPs certainly have significant reflection. However, the NP Cold showed a higher heat transfer resulting from the emissivity of Cleartran® + NPs being larger than that of using Cleartran® glass alone. Even if the reflectance is non-zero, the emissivity value must be < 0.25 because of the ($\tau \approx 0.75$) for the Cleartran® glass. The standard deviations for heat fluxes displayed from Fig. 5a to Fig. 7b are estimated for 5 observations for all window samples, and temperature deviations while recording surface temperatures with an IR camera are ± 0.1 to ± 0.5°C.

However, the obtained heat fluxes are insufficient to improve the thermal performance of the windows for passive cooling. While for simple glass the heat flux is increased by a factor of 2 to 4, for the Cleartran® glass the change is only a few %. The particle coverage for TiO_2-SiO_2 NPs was about 50% less than that of ZnS-SiO_2 composites, according to the SEM investigation (see section 4.2). On the other hand, the amount of NPs that are deposited on the window glass surface is much lower as well. Some work needs to be done and added to this conference paper for it to be appropriate for a journal paper for a dr. thesis.

5. Conclusions

The objective of this research was to examine the LW heat flux through plain window glass, window glass with dispersed NPs, Cleartran® glass, and Cleartran® glass with NPs dispersed on one side in the field of view between a heat source and an IR thermal imaging camera. As shown, the LW heat flux passing through plain window glass is substantially lower than the LW heat flux flowing through window glass with NP on the Cold side, facing away from a heat source. The stronger emission on the NP Cold side increased the heat flux in the simple glass by a factor of two to four. On the other hand, due to the high transmittance ($\tau \approx 0.75$), the heat flux of Cleartran® is slightly (a few %) higher compared to Cleartran® with NP Hot side, as the NP Hot side causes some reflection. Nevertheless, the heat flux flowing through the NPs on the cold side is larger than that of standard Cleartran® glass and Cleartran® glass with NP Hot, which is due to the increased emission in addition to transmission. In terms of heat flux values, the glass with ZnS-SiO_2NPs shows a slightly stronger effect compared to glass with deposited TiO_2-SiO_2 NPs, partly due to different deposits. The heat flux through the Cleartran® glass with NPs is only slightly affected due to various factors, owing to this glass materials' high transmittance in the LW region. So far, the heat flux values measured may not be sufficient for improving thermal performance of the ÅA passive cooling skylight. In the future, parameters such as the NP composition, size, shape, area fraction (in %), and interaction with the glass matrix need to be investigated further and in more detail. These variables have an impact on the heat flux values because they govern how much light is reflected, absorbed, and transmitted. Finally, the data obtained in this work can be verified with a pyrgeometer (4 – 40 μm) with a wider LW radiation wavelength range and for shortwave radiation heat fluxes under solar irradiation using the thermal imaging camera. This will show the effectiveness of deposited NPs on lowering SW irradiation heat flux passing the glass material.

Acknowledgements

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References

[1] R. Zevenhoven and M. Fält, "Radiative cooling through the atmospheric window: A third, less intrusive geoengineering approach," *Energy*, vol. 152, no. December 1997, pp. 27–33, 2018, doi: 10.1016/j.energy.2018.03.084.

[2] M. Fält, The utilization of participating gases and longwave thermal radiation in a passive cooling skylight-. PhD thesis, Åbo Akademi Univ., Turku, Finland, 2016. https://www.doria.fi/handle/10024/125709.

[3] A. Bendavid and P. J. Martin, "Review of thin films materials deposition by the filtered cathodic vacuum arc process at CSIRO," 2014. [Online]. Available: www.austceram.com/ACS-Journal

[4] Z. M. Cheng, Y. Shuai, D. Y. Gong, F. Q. Wang, H. X. Liang, and G. Q. Li, "Optical properties and cooling performance analyses of single-layer radiative cooling coating with mixture of TiO2 particles and SiO2 particles," *Sci China Technol Sci*, vol. 64, no. 5, pp. 1017–1029, 2021, doi: 10.1007/s11431-020-1586-9.

[5] G. Gangisetty and R. Zevenhoven, "A Review of Nanoparticle Material Coatings in Passive Radiative Cooling Systems Including Skylights," *Energies*, vol. 16, no. 4. MDPI, Feb. 01, 2023. doi: 10.3390/en16041975.

[6] D. Kaminski, M. Jensen "Introduction to Thermal and Fluids Engineering", Wiley (2005), Chapter 14.

[7] G. Gangisetty and R. Zevenhoven, "Selection of nano-particulate material for improved passive cooling skylight performance, in Proceedings of ECOS 2022 - The 35th International Conference on Efficiency, Cost, Optimization, Simulation and Environmental Impact of Energy Systems. July 4-7, 2022, Copenhagen, Denmark" 2022.