



Research Article

Dual-Layered Titanium Oxide-Acrylic Film on Carbon Black for Passive Radiative Cooling below Ambient Temperature under Direct Solar Irradiance

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Abstract: Cooling is an energy-intensive process; and with the effects of global warming, space conditioning poses more load on the scarce and costly energy available for consumption in facilities. Thus, the use of a passive sub-ambient diurnal radiative cooler, which is an effective natural space cooling technology, can help reduce the total energy demand for buildings. A diurnal passive radiative cooler of 40×40 cm, has been designed, fabricated, and experimentally investigated. The cooler comprises a photonic solar reflector and thermal emitter made of titanium dioxide embedded in an epoxy resin that reflects 96% of insolation and emits strongly in the $8\text{-}13 \mu\text{m}$ atmospheric window. Design calculations were made under Owerri climatic conditions. The results obtained showed that when the cooler was exposed to direct insolation well above 950 W/m^2 , it achieved a drop in temperature of $1\text{-}2 \text{ }^\circ\text{C}$ below ambient between 6 am and 9 am. As the solar radiation increased, the temperature of the cooler increased until it reached $41 \text{ }^\circ\text{C}$, which was above the ambient air temperature ($30 \text{ }^\circ\text{C}$). The cooler temperature decreased as the solar radiation decreased, dropping below the ambient temperature from 5:45 pm, by $3 \text{ }^\circ\text{C}$ during the remaining hours of the investigation. The photonic radiative cooler has an estimated cooling power of 56.8 W/m^2 under a clear night sky and achieved sub-ambient cooling during the early and late hours of the day under low solar radiation. Therefore, passive cooling through the photonic approach offers prospects for energy efficiency. Further works may have the prospects of achieving improved passive sub-ambient daytime cooling.

Keywords: photonic radiative cooler, thermal radiation, solar radiation, solar reflector, experiment, sub-ambient cooling

1. Introduction

The cooling of indoor space is commonly used as a means of reducing excessive heat in the built environment. Cooling demand in buildings has been predominantly met by the use of conventional energy generating systems [1-3], leading to local and global climate changes [4]. According to [5], building energy demand is on the increase and likely

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will surpass the heating demand by 2040; and if the current trend continues, it will culminate in about a 78% increase in residential building consumption alone in comparison with the current value [6]. Some of the attendant consequences include increased cooling costs, excessive ambient temperatures exceeding comfort and health levels, increased pollutant and greenhouse gas emissions, and intensification of tropical overheating arising from global warming and climate change [7]. Consequent to these, devices using different space cooling methods that are environmentally friendly such as daytime passive radiative cooling systems are gaining wide recognition in recent times and have the promise of supplementing or entirely replacing the conventional energy systems [8].

The passive radiative cooling strategy operates solely without electric power input and also ensures the cooling of objects without any external resources. On its own, it selectively radiates thermal energy accumulated in objects to the cold outer space through the atmospheric window spectrum, 8-13 μm [8-9]. It has been identified that the performance of passive radiative coolers much depends on the prevailing climatic conditions such as solar radiation, ambient temperature, humidity, wind velocity and cloud cover in a particular location [10-11]. This, therefore, supports location-specific assessment before deployment.

Nocturnal or night-time passive radiative cooling has received extensive attention [11]. Harrison and Walton developed a radiative cooler, which was spectrally selective [12]. The device was produced using an aluminum plate coated with an optical layer on top of a white paint having relatively high emissivity with the atmospheric window, which resulted in a nocturnal cooling of 15 °C below the ambient air temperature. According to [13], a thin film of polydimethylsiloxane (PDMS) aluminum was used in the development of a radiative cooler which was simulated to have a passive cooling of about 12 °C lower than the ambient temperature during the night sky period. According to [14], radiative cooling largely depends on the conditions of the ambient weather. Thus, changes in humidity, winds, and cloudiness influence night-time cooling intensity. A strong ambient wind of more than 3.6 m/s associated with cloud cover and high humidity can delay or entirely repress cooling [14-15]. Nigeria's climate is characterized by three distinct zones, namely, a monsoon tropical climate predominant in the southern part, a savannah tropical climate for regions in the central part, and a climate that is semi-arid and hot in the northern part. Bokor et al. reported that an arid climate condition supports nocturnal radiative cooling due to its dryness [16]. The potential of nocturnal radiative cooling in Nigeria has been reported [17-23].

The development of metamaterials has revolutionized the diurnal or daytime passive radiative cooling technology [11]. Diurnal passive radiative cooling can cool effectively under direct insolation when the majority of the insolation within the visible and near infra-red spectrum is reflected at the same time [8]. Catalanotti et al. developed a radiative cooling device using a coated evaporated aluminum plate with plane Polyvinyl Fluoride (PVF) film having an average emissivity of 0.85 in the atmospheric window, and reflectance of 0.85 outside the region, thus leading to nighttime cooling [24]. The effectiveness of Lithium Fluoride (LiF) as a radiative cooler for sub-ambient radiative cooling has been investigated by [25], obtaining a net cooling power of about 85 W/m² in a clear night sky. The first use of a photonic structure for daytime radiative cooling was published by [26]. The system is composed of a solar reflector with several sets of bilayers of Magnesium Ferrite (MgF₂) and Titanium Oxide (TiO₂) lying under two thermally emitting layers of Silicon Carbide (SiC) and quartz. The prediction of the generated models showed that under real conditions of the atmosphere and direct sunlight, the system could achieve a temperature range of 4 to 6 °C below the ambient temperature conditions, or a cooling power density greater than 100 W/m², at an ambient temperature of 300 K.

The first diurnal radiative cooling process using a multilayer photonic device consisting of seven dielectric layers placed at the top of a silver mirror, showing a good reflection for the entire solar spectrum and reliable selective thermal emission within the transparency window of the atmosphere has been experimentally demonstrated [9]. In the design of the dielectric layers, an optimization process was considered which took cognizance of the real global fabrication challenges. Thus, the layers made of Silicon Dioxide (SiO₂) and Hafnium Oxide (HfO₂) with thicknesses in the order of hundreds of nanometers, cause the generation of strong thermal radiations. The layers of SiO₂ and HfO₂, serve to improve the reflectivity of the silver mirror mainly in ultraviolet wavelengths. When the system was positioned at the top of a roof, it attained a temperature of 5 °C lower than the ambient air temperature having 900 W/m² of sunlight directly falling upon it.

The use of polymer materials to achieve diurnal radiative cooling of 2 °C lower than the ambient temperature when it was exposed to direct solar irradiation of 1060 W/m² without a convection shield has also been demonstrated [27]. This happened when a pile of alternating birefringent polymer layers was used with a refractive index in the absence of

a polymer mirror within the solar spectra in the range of 0.4 to 1 μm . An achievement of 11 °C cooling using this device was recorded when placed below a white painted roof whose solar reflectivity and Infrared emissivity were moderate. Thus, showing the potential of the polymer radiators for passive cooling applications. Chen et al. developed a radiative cooling device with a selectively operating thermal emitter using a vacuum system, eliminating most convective and conductive heat gains, and achieving a reduction in the temperature of more than 40 °C [28].

The use of a 2D metal-dielectric photonic structure system which was made up of a selectively operating thermal emitter placed at the top of a broadband mirror has been reported [29]. The reflector had three sets of several bilayers of Magnesium Fluoride (MgF_2) and Titanium Dioxide (TiO_2) over a silver (Ag) substrate. The emitter was made of two 2D layers of Silicon Carbide (SiC) and Quartz presenting a strong resonance within the atmospheric window. The emissivity of the structure within the atmospheric window was enhanced using a phonon-polariton approach. The simulations carried out showed that the device could achieve a cooling power of about 105 W/m^2 and a diurnal temperature fall of about 8 °C lower than the temperature of the ambient air for a coefficient of heat transfer (h) of 12 $\text{W/m}^2\text{K}$ or 15 °C for h of about 6 $\text{W/m}^2\text{K}$, respectively. Also, Bao et al. proposed the use of a double-layer radiator that has both a down emissive layer of Silicon dioxide (SiO_2) and a top emissive layer of TiO_2 . It recorded ambient temperature reduction of about 17 °C at night and 5 °C during the day [30].

Huang and Ruan developed a double-layer coating of acrylic resins embedded with TiO_2 and carbon black parts for reflection of solar irradiation and emission in the atmospheric window respectively [31]. From the examination conducted on the titanium dioxide particles, it was found that the 0.2 μm radius of the particle gave the best performance. Another development that uses polydimethylsiloxane (PDMS) was coated with fused silica, achieving a diurnal cooling of about 8 °C below the ambient temperature. Liu et al. reported that an epoxy resin film coated with silver recorded an infrared emissivity of 0.96 and solar reflectivity of 0.97 in the atmospheric window, and fused Quartz and PDMS of 100 μm thickness showed a temperature fall of 8.2 °C from the ambient and cooling power of about 127 W/m^2 under direct solar irradiance [32]. And Yang et al. proposed the use of millimeters of sintered polytetrafluoroethylene (PTFE) with a reflector made from a silver film which achieved a reflectivity of 0.991 and refrigeration of 11 °C below the ambient temperature [33].

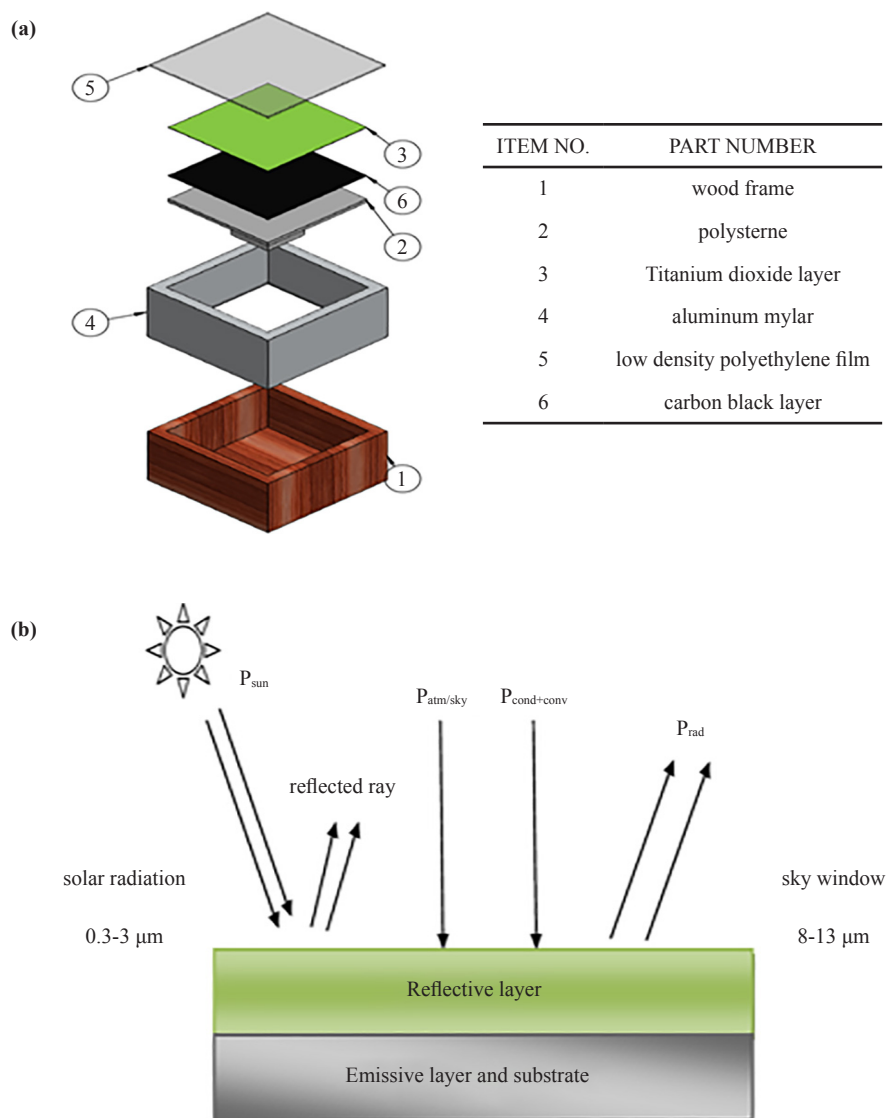
Two radiative coolers, one made from a 3 mm thick ultra-white glass on a 200 nm silver substrate, and the second made from sodium zinc phosphate (NaZnPO_4) particles on an aluminum substrate, in which the first cooler has a spectral surface, and the second, a diffusive surface have been comparatively studied [34]. The first cooler recorded a 2.5 °C temperature below ambient on a daytime period, and 9.8 °C below ambient temperature during nighttime hours; whereas the second radiative cooler obtained a 1.5 °C and 7.3 °C below the ambient temperature during the diurnal and nocturnal hours, respectively.

Bijarniya et al. assessed the potential of ideal, broadband and selective emitters for radiative cooling in different Indian cities, and found that selective emitters outperformed the others in Jaisalmer and Delhi associated with low humidity [10]. However, this may not be the situation in monsoon and high humid locations. Although photonic structures are capable of satisfying the emission and reflection requirements for cooling [9], generally, some of the challenges contending with the effectiveness of diurnal passive radiative cooling include varying cooling power due to varying climatic conditions, excessive material costs, and stability under direct sunlight [8, 11]). Therefore, researches are ongoing to develop simple, cheap, and thermally stable coolers with high cooling power.

From the foregoing, there are no laboratory and field investigations of diurnal passive radiative cooling in Nigeria and this is the first feasibility investigation on the potential of such technique in monsoon, savannah, and semi-arid and hot tropical climates characterizing the southern, central, and northern parts of Nigeria. Since it has been reported that humidity greatly influences the performance of photonic radiative coolers, particularly the cooling effects of titanium oxide coating [35], it becomes necessary to undertake the performance study in other climates with differing humidity. The current study is an effort to develop a simple, cheap, and high-performance diurnal radiator using a bilayer photonic cooler consisting of titanium dioxide embedded in an epoxy resin on a carbon black substrate, and the assessment of its cooling performance under direct sunlight in Owerri (Nigeria) climatic condition. The emitter has high solar reflectivity and near-black absorptivity in mid-infrared. The sub-ambient temperature and radiative cooling power obtained were compared with the reported sub-ambient temperatures and cooling powers from other investigations in the literature to highlight the findings from the novel investigation.

2. Cooler configuration and description

The photonic radiative cooler is made up of a bilayer of titanium dioxide embedded in an epoxy resin atop a carbon black substrate to enhance thermal radiation as a result of material properties and interference effects as shown in Figure 1a. The carbon black for the bottom layer was chosen due to its high level of emissivity (0.95) in the atmospheric window, which makes it a suitable choice. The top layer is a layer of titanium dioxide embedded in epoxy resin. This is responsible mainly for reflecting incident solar radiation on the cooler. The TiO_2 does not absorb in the solar window but reflects a greater percentage of the incoming solar radiation. The essence is to have a system with a combined effect of excessive solar reflectance and highly improved thermal emission. The photonic cooler was mounted on a polystyrene pad borne by a wooden box. A clear low density polyethylene film was placed above the sample as a transparent windshield to allow for strong thermal radiation in the atmospheric window. Figure 1b shows the energy interaction in the cooler.



To investigate the performance of the cooler, the test rig was structured as shown in Figure 2. The device was mounted onto a 400 × 400 mm cavity, made of titanium dioxide and epoxy resin.

The nano-thick embedded layer was fabricated over a layer of carbon black substrate and emitter as shown in Figure 2a. It involves the mixing of two parts of epoxy and one part of epoxy hardener. Epoxy was chosen due to its high transparency within the atmospheric window as this would allow the layers below to interact adequately with the environment. The titanium dioxide was prepared and uniformly mixed with acrylic resin with a volume fraction of about 15%. The TiO₂-resin mixture was sprayed on a 0.5 mm mold and allowed to set and solidify for 24 hours. A hollow square-shaped groove was made in a polystyrene foam to accommodate the carbon black substrate upon which is deposited the TiO₂-resin layer. The surface polystyrene, except the top, was wrapped with aluminum foil. The carbon black substrate was spread evenly on top of the sensors to the brim of the groove made in the polystyrene foam, then the dried layer of TiO₂-resin was deposited on top of the carbon black. The mid-infrared emissivity is unity and zeroes outside this band.

From Figure 1b, it can be seen that there are three components of heat flow into the system and one out, namely, P_{atm} , $P_{\text{cond+conv}}$, P_{sun} and P_{rad} , respectively. The net cooling power of the cooler can be obtained by the heat balance expressed as in Eq. 1:

$$P_{\text{cool}} = P_{\text{rad}} - P_{\text{atm}} - P_{\text{cond+conv}} - P_{\text{sun}} \quad (1)$$

where P_{rad} is the energy radiated by the cooler, P_{atm} is the radiant energy from the sky/atmosphere, P_{sun} is the incident energy from the sun, and $P_{\text{cond+conv}}$ is the heat energy from the surroundings by virtue of conduction and convection and P_{cool} is the system's net cooling power, which is the difference between the energy radiated by the cooler and every other source of heat gain into the system. A positive value of P_{cool} suggests an efficient system which would lead to a drop in temperature in the system.

3. Experimental setup

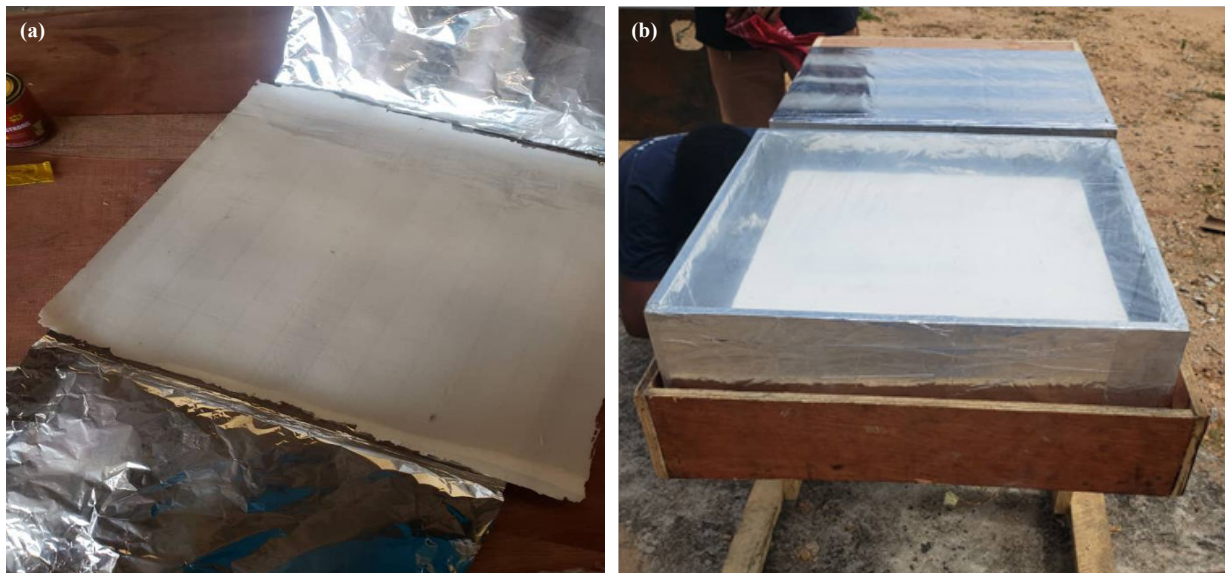


Figure 2. (a) Titanium dioxide and epoxy resin layer (b) The experimental setup

Three DHT11 temperature sensors were mounted at three different locations on the back side of the cooler and

connected to a data logger. The entire apparatus as shown in Figure 2b was placed on an elevated platform in an open field at the Procurement Building, Federal University of Technology, Owerri, Nigeria (5.49°N, 8.67°E) at a tilt of 20.49°. It was exposed to the sky from 8:00 am on the 5th of August, 2021 to 8:00 am on the 6th of August 2021 in order to take the temperature readings of the cooler for the 24 hours. The solar radiation incident on the cooler was measured using a Solarimeter (Solar Power meter) with a range of 0.1-1999.9 W/m². The ambient temperature and relative humidity data were measured using DHT11 sensor probes while the wind speed was measured with the aid of an anemometer. While wind speed and solar radiation data were collected every 30 minutes, the data logger was employed to record the cooler and ambient temperatures at 5 minutes intervals.

4. Results and discussion

The sub-ambient temperature and radiative cooling power from the present investigation compared with the same data from literature is as shown in Table 1.

Table 1. Cooling performance comparison

Sub-ambient Temperature (°C)	Cooling Power (W/m ²)	Cooler Layer Structure	Solar Irradiation (W/m ²)	Source
4-6	100	Multilayer (MgF ₂ + TiO ₂) on (SiC + Quartz)	-	[26]
4.9	40.1	Multilayer (HfO ₂ + SiO ₂)	860	[9]
2	-	Polymer materials	1060	[27]
8	105	Multilayer (MgF ₂ + TiO ₂) on (SiC + Quartz)	-	[29]
5	-	SiO ₂ + TiO ₂	-	[30]
8.2	127	Epoxy + Quartz + PDMS	-	[32]
2.5	-	Ultra-white glass + Ag	-	[34]
1.5	-	Dual (NaZnPO ₄ + Al)	-	
7.2	136.3	Multilayer (TiO ₂ + SiO ₂)	680	[8]
4.3	72.7	Dual (PDMS + Ag)	760	[36]
1-3	56.8	Dual (TiO ₂ + Acrylic Resin)	1050	Present work

The performance of the photonic radiative cooler was investigated experimentally for two consecutive days from 6:00 am, 05/08/2021 to 06:00 am, 07/08/2021 to cover both the diurnal and nighttime thermal performance of the system.

Figure 3 shows the temperature values of a horizontally mounted cooler without any tilt on a cloudy day. At about 8 am, the temperature of the ambient reached a value of about 27 °C, which increased slowly to 29 °C at 9:40 am and maintained almost the same value throughout the day. The cooler and aluminum foil temperatures increased to a maximum of 38 °C and 43 °C at 1:15 pm and 3:40 pm, respectively. Although the cooler temperature remained above the ambient throughout the sun peak period, the difference in the temperatures recorded was not significant. At about 6:00 pm, the cooler and aluminum foil temperatures began to fall while the ambient temperature was noticed to commence an upward rise above the cooler temperature. This, shows that cooling commenced at about 6:00 pm on a horizontally mounted cooler on a cloudy day.

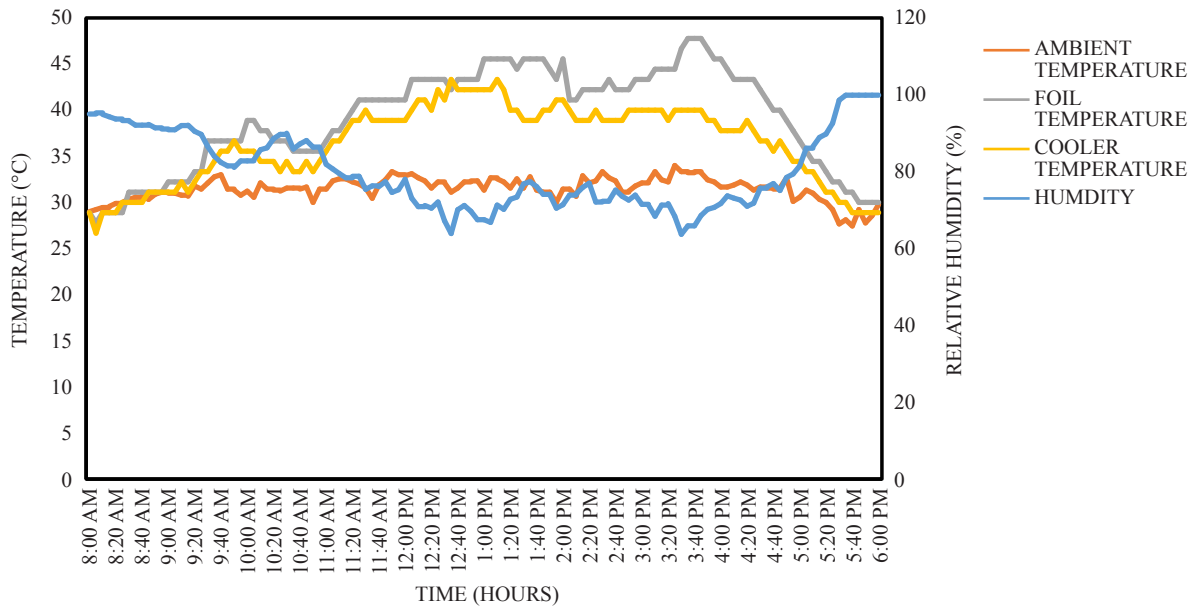


Figure 3. Radiative cooler equilibrium temperature from 8 am to 6:30 pm for a horizontal surface on a cloudy day, 5th August 2021

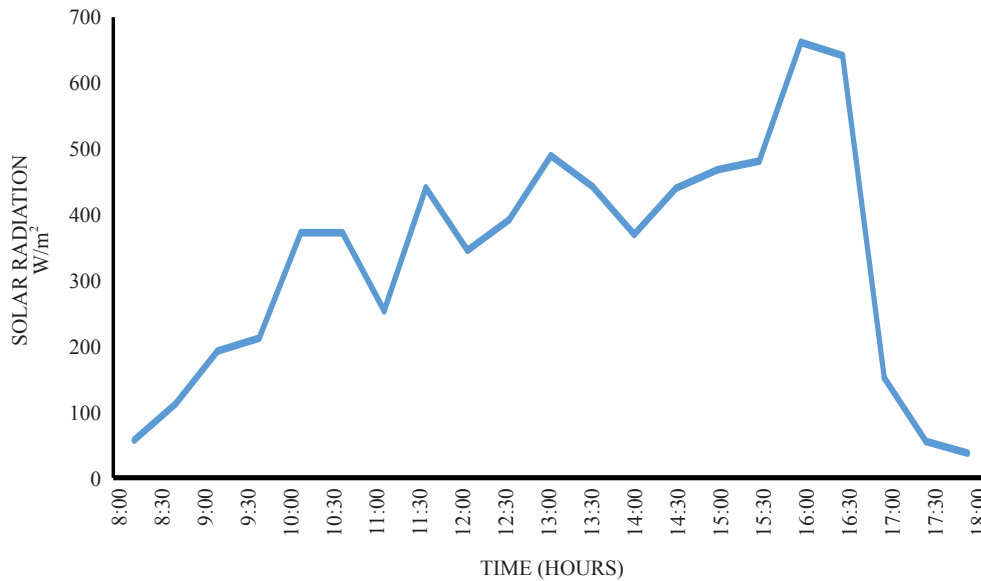


Figure 4. Solar irradiation measurements from 8 am to 6:30 pm for a horizontal surface on a cloudy day, 5th August 2021

At 9:40 am, the humidity of the air dropped from 99.8% to 83%, and further decreased to 63% at the peak cooler temperature around 12:35 pm. The humidity ranged between 62% and 70% from 3:30 to 4:57 pm, after which it experienced a rapid increase, while the temperature of the cooler and aluminum foil decreased significantly.

Figure 4 shows the solar radiation on the same day from 8 am to 6:30 pm for a horizontal surface. The solar radiation increased to 200 W/m² at 9:30 am when the temperatures of the cooler and aluminum foil rose above the ambient temperature (Figure 3). At the peak value of the cooler temperature around 1:15 pm, the solar radiation was 489 W/m², and reached its maximum value of 660 W/m² at about 4:15 pm. The foil temperature attained its maximum value at 3:50 pm, about which time the humidity began to increase rapidly.

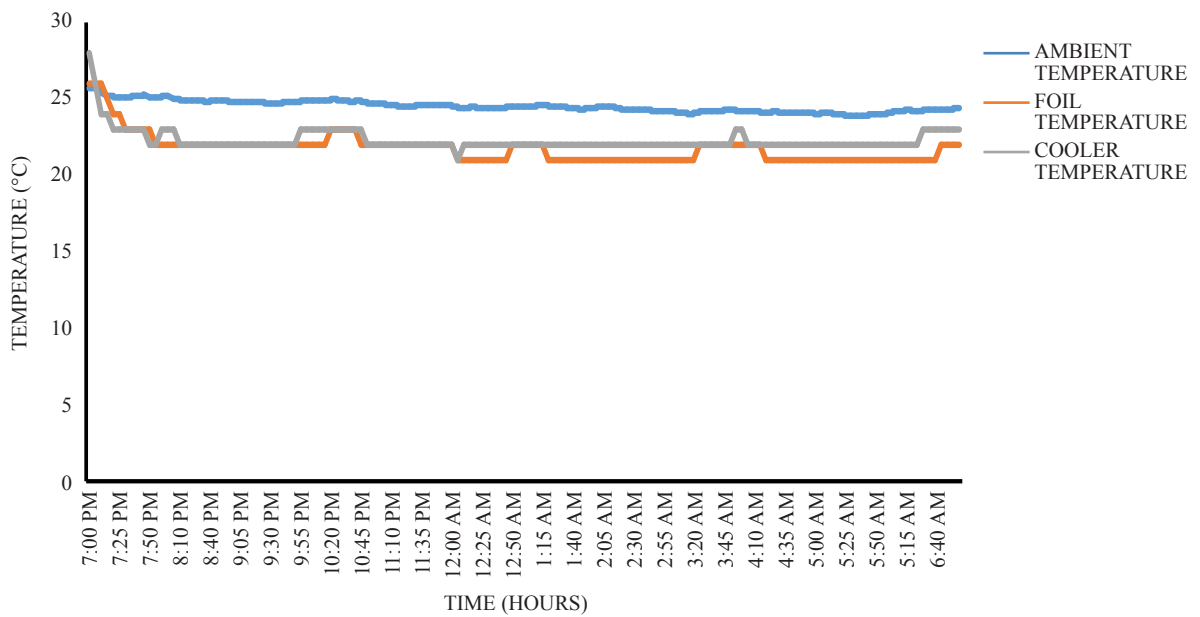


Figure 5. Radiative cooler equilibrium temperature from 7:00 pm 5/08/2021 to 6:40 am 6/08/2021 for a horizontal surface

Figure 5 shows the interactions of the ambient temperature, the cooler temperature, and the aluminum foil temperature during the nighttime when there was no more solar radiation. At 7:00 pm the temperatures of the cooler, the ambient air, and the aluminum foil were exactly equal, after which the temperature of the cooler and aluminum foil continued to drop until it reached 22 °C at 8:40 pm. From this time to 6.40 am, the ambient temperature fluctuated between 24 °C and 23 °C, while the cooler temperature ranged between 22 °C to 21 °C throughout the night period. This means a temperature drop of about 3 °C below the ambient in the cooler during the night. This is a result of night-sky-cooler interaction.

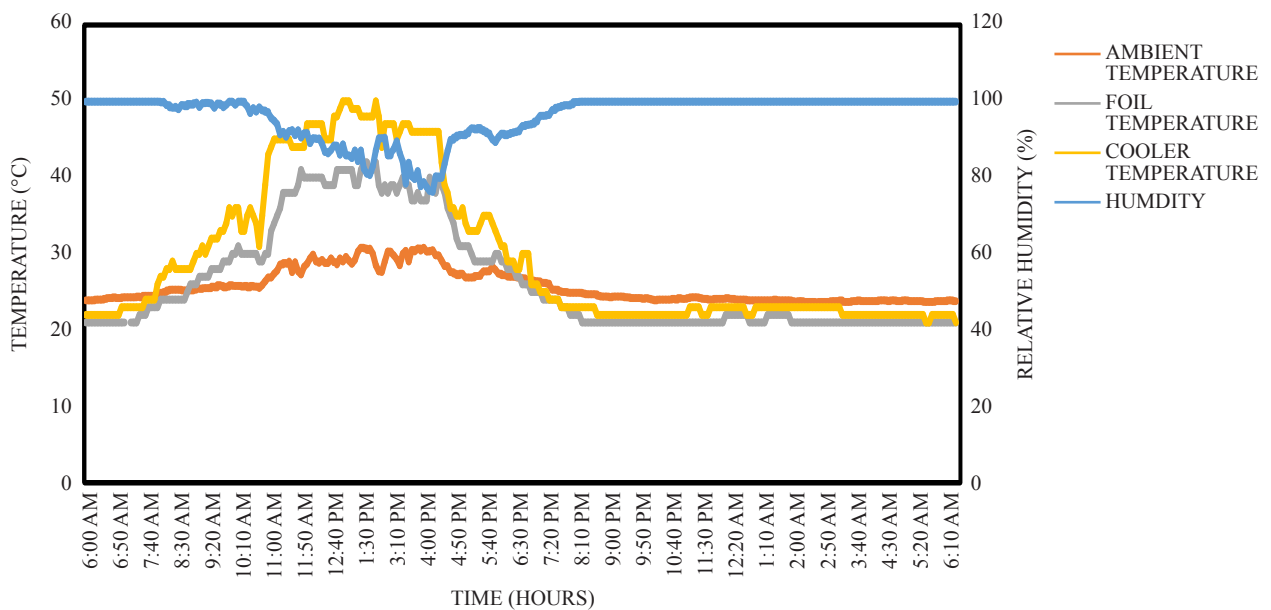


Figure 6. Radiative cooler equilibrium temperature for a 24-hour cycle 6/08/21-7/08/21 on a tilted surface on a clear sky

Figure 6 shows the equilibrium temperature of the cooler, at the tilt angle of 10 °C surface on a clear sky for August 6th, 2021. As shown in Figure 6, the cooler temperature remained below ambient until about 6:45 am when it began to rise. At about 6:00 am, the temperatures of the cooler and foil were 21 °C and 22 °C, respectively, whereas the ambient temperature was 23.5 °C. At exactly 6:45 am, the cooler and aluminum foil began to show noticeable increments in their respective temperatures, although still below the ambient temperature. At 9:00 am, the temperature of the cooler became equal to the ambient temperature when the aluminum foil temperature had increased by 4 °C above it. Also, from 6 am to 8.45 am, the humidity decreased from 98% to 96%. During this period, the cooler temperature remained above ambient till around 4:30 pm when it significantly dropped though still fairly above ambient. The obvious observation is that around 5:40 pm when the solar radiation was still incident on the cooler, it dropped below ambient, signifying sub-ambient cooling under direct sunlight. The temperature drop was about 2 °C till about 6:40 pm when it increased beyond that and gradually progressed into the night-time hours.

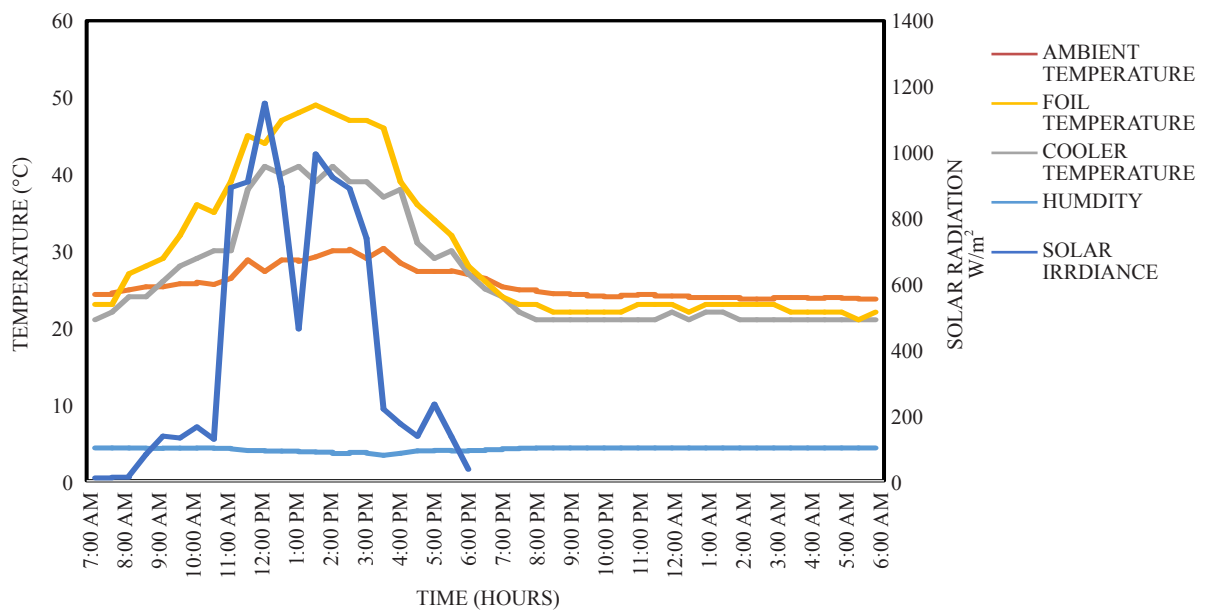


Figure 7. Radiative cooler equilibrium temperature for a 24-hour cycle for a tilted surface on a clear sky with solar irradiation

After 9:00 am, the cooler and aluminum foil temperatures kept increasing, irrespective of a slight increase in the ambient temperature. Between 10:30 am and 12 noon, a rapid increase in the ambient air temperature from 24 to 27 °C occurred, during which time the cooler also experienced an increase in temperature from 28 to 37 °C, respectively. During this period, the solar irradiation reaching the cooler increased from about 140 W/m² to 1125 W/m² as shown in Figure 7. The intensity of the solar radiation attained its peak around 1.00 pm. At 2:00 pm, the ambient temperature rose to its peak value of 31 °C, while the humidity of the air dropped to 80% before reaching its minimum for the day at 75% by 4:30 pm. At the ambient air peak temperature, the cooler reached a peak temperature of 42 °C, while the aluminum foil reached a peak temperature of 49 °C. From Figure 6, the humidity reached the minimum value and the aluminum foil and the cooler temperature began to drop from 47 °C to 41 °C, respectively. The three temperature readings kept dropping as the solar radiation continued to decrease with increasing humidity. At 5:40 pm the cooler temperature became equal to the ambient temperature and thereafter began to drop gradually. While the cooler was below the ambient temperature, the humidity was increasing as the temperature continued to drop. At 7:30 pm the temperature of the cooler and aluminum foil became more stable at 21 °C and 22 °C, respectively while the ambient temperature was 24 °C. At this time the solar radiation was gone and its effect was not felt by the cooler and aluminum foil.

5. Conclusion

The radiative cooling performance of a locally produced photonic radiative cooler made from titanium dioxide embedded in an epoxy resin, and deposited on a carbon black substrate has been carried out. The cooler is simple in structure and capable of implementing daytime and nighttime cooling under mild solar radiation intensity and low humidity, respectively. The results obtained showed that the overall reflectivity of the cooler surface in the solar spectrum (0.25-3 μm) was well below 0.97, thereby inhibiting its cooling power. The system achieved a sub-ambient cooling during the early morning hours and late evening hours, and an average temperature drop of 3 °C was achieved during the nighttime, thus, revealing the prospects of radiative cooling in the tropical climate of Owerri. This, therefore, confirms the strong dependence of the performance of photonic coolers on weather conditions. The current study, which is the first of its kind in Nigeria and the sub-Saharan African region as a whole, has further confirmed the feasibility of night-sky cooling, but much remains to be done in order to successfully achieve daytime cooling in Nigeria under direct sunlight. This is because, under most practical circumstances, the developed radiative cooler of embedded titanium is practicable for daytime radiative cooling when exposed to little solar radiation but its cooler temperature increased above ambient under peak solar irradiance. It is recommended that further field investigations be conducted using the current cooler design under solar shading during peak insolation. Future works are concentrating on improved cooler structural design and fabrication techniques as well as numerical simulations for a more comprehensive evaluation of its performance.

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Conflicts of interest

The authors declare no competing financial interest.

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