Figures of merit of passive daytime radiative cooling materials

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Abstract. Passive daytime radiative cooling (PDRC) materials represent an emerging technology that can provide sub-ambient cooling by dissipating heat as radiation through the long-wave infrared transparency window of the atmosphere. As such, they hold promise to alleviate our growing cooling needs and could find application in a broad range of areas. An increasing number of PDRC materials and applications is reported and tested each year. The fast-paced progress in this field also creates higher demand for reliable and universal methods for comparing the performance of novel materials and predicting their cooling abilities in different environments. However, clear figures of merit and standardised testing methods to evaluate real-world cooling performance are still lacking, so that the cooling performances of various novel PDRC materials presented in literature often cannot be compared. In this work, we review and discuss these issues from the specific viewpoint of the European Partnership on Metrology project PaRaMetriC, which aims at developing a metrological framework to classify and compare these materials.

1 Introduction

Reducing energy demands for cooling technologies has become one of the most relevant scientific challenges for sustainable development. Due to the imminent effects of global warming and growing urban areas, tripling of the cooling equipment installed capacity is expected before 2050 [1]. The prospect of the associated electricity consumption increase generates, among others, an urgent need for researching new cooling strategies and technologies. Passive radiative cooling (PRC) [2] process makes advantage of a spontaneous heat radiation towards a cold heatsink, which can potentially provide cooling of the material below the ambient temperature without the need of an external energy source. Key feature for utilizing PRC in cooling technologies is the ability of some materials to emit infrared radiation selectively in the wavelength range between 8 – 13 μm (long wave infrared atmospheric window, LWIR). Outer space can act as an infinite heat sink for thermal emitted radiation since the atmospheric gases don't have significant absorption bands in this range. An unobstructed access to a clear sky and high emissivity in the LWIR atmospheric window are therefore two main conditions for a successful PRC operation.

Thanks for this principle, reaching below-ambient temperatures by PRC during night is well documented since ancient times. On the other hand, passive daytime radiative cooling (PDRC) is far more challenging and was a subject of intensive research in the past decade. Achieving a net cooling effect under direct sunlight illumination [3] requires little to no absorptivity across the whole solar spectrum (0.3 – 2.5 μm). The well documented progress in both scientific understanding of PDRC phenomenon and development of its practical use (most prominently in cooling in the building sector [4]) brought many novel designs of PDRC materials [5], fabrication techniques and architectures of full-scale cooling systems [6-8]. However, there are still some shortcomings in the scientific coverage of the PDRC field, related to the adoption of inconsistent performance assessment methods and or the reporting of incomplete or incorrectly determined figures of merit, which makes it challenging to summarize the current state of the art [9]. Recognizing and overcoming these problems could help broadening the use of PDRC in real life and applying various scientific findings to address the current energy crisis. Overall, PDRC technology has the potential to reduce cooling energy demands and mitigate the urban heat island effect [10], significantly reducing cooling-related emissions of greenhouse gases while improving human comfort and air quality in urban environments. This study aims to identify the figures of merit for PDRC materials that can help define and expand the potential of this promising technology.

2 Coling performance assessment methods

Every year, more passive daytime radiative cooling materials and applications are reported and evaluated. Significant development in the field of PDRC in recent years creates an increased demand for accurate and universal methodologies for assessing the performance of innovative materials and forecasting their cooling

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capacities in varied situations. However, widely agreed methods have not been identified or validated yet. Bu *et al.* defines three main obstacles that result in inconsistencies in the measured data comparisons [9]: i) different measurement standards, ii) different experimental setups and iii) reporting different figures of merit for measured data. Figures of merit used in the majority of published studies are strongly dependent on both the experimental conditions and thermal properties of the materials, resulting in the current impossibility of comparing the performance of different materials directly against each other in a consistent way. In the following sections, frequently used figures of merit for PDRC materials will be described and various limitations associated with their use will be discussed.

2.1 Spectral properties

Considering the basics of PDRC, the spectral properties of the material are often used to characterise its suitability for these applications. The emissivity spectrum of the material has to be measured and its similarity with the ideal emitter models is then evaluated. Two options are most commonly used for designing PDRC materials: narrow-band emitter with high emissivity within 8 – 13 µm and broadband emitter with high emissivity within 4 – 25 µm. In both cases, the materials should have zero emissivity in the solar spectrum. Two main types of materials present specific advantages in different working conditions. Broadband emitters are able to radiate more heat outward, hence reaching potentially higher cooling powers, but only at a temperature above or slightly below the ambient. On the other hand, narrow-band emitters are suitable to realise larger sub-ambient temperature drops as their cooling power decreases more slowly with decreasing temperatures, and they are more radiatively insulated from atmospheric irradiance. Narrow-band emitters are therefore able to reach lower cooling temperature in absolute terms than broadband emitters [11]. However, broadband emitters can still be used for sub-ambient cooling, mostly in cold and dry areas with high atmospheric transmittance.

Some other ideal emissivity profiles have also been proposed, for instance featuring an additional drop between 9 and 10 µm to avoid radiative exchanges with the ozone absorption band in that wavelength range. However, such fine tailoring of the emissivity profile is difficult to achieve in actual samples [12].

Numeric analysis of different emissivity profiles and PDRC performance of the emitters can be also found in the work of Kecebas *et al.* [13]. According to their simulations, the ideal emissivity for reaching the lowest equilibrium temperature depends on the total heat load q acting on the surface coming from both radiative and non-radiative contributions. In the case of nearly insulated systems, values of q approach zero and lowest temperatures are reached when the emissivity is narrowed between 10 – 12 µm. However, with increasing q, the ideal emissivity broadens first to the usual narrow-band (8 – 13 µm) profile, and eventually to the broadband case.

Besides emissivity, solar reflectance is another parameter strongly correlated with PDRC performance. As suggested by Bu *et al.*, however, optical characterization and solar reflectivity measurements are often affected by common shortcomings in several recent studies on PDRC materials [9]. In ultraviolet-visible-near-infrared spectroscopy, standard reference samples are often measured prior to the actual PDRC material measurement, for calibration purposes. Unfortunately, most studies do not specify whether the reported reflectance values are absolute or relative to the reference standard. The reflectance of the standards is typically below 100 %, even within the specification of the manufacturers, which can lead to a significant reflectance overestimation if only relative values are reported. The approximation of the reflectivity of reference samples can lead to a critical underestimation of the overall solar absorption up to 25 %, introducing a systematic bias which affects the cooling performance prediction. It is therefore important to evaluate and include uncertainty estimation when presenting PDRC materials characterization.

Instead of reporting both infrared emissivity and solar reflectance values, spectral properties can be merged into one figure of merit. For example, Li *et al.* [14] proposed to use a simple metric called RC, which can be computed as:

$$RC = \varepsilon_{\mathrm{sky}} - r\,(1 - R_{\mathrm{solar}}), \qquad (1)$$

where $\varepsilon_{\mathrm{sky}}$ is the emissivity in the sky window, R_{solar} is a total reflectance in the solar spectrum, and r is the ratio of the solar irradiation power over the blackbody surface emissive power transmitted through the sky window. RC should be used to compare different PDRC materials at the same solar irradiation and weather conditions. The authors recommend using the "standard RC" by referring to a standard surface temperature of 300 K to calculate the emissivity, and using a standard r value of 10 (a typical peak solar irradiation of 1000 W m^{-2} and blackbody emissive power 100 W m^{-2}). The RC parameter has been used in various studies to compare different PDRC materials as it conveniently condenses the basic information about the emissivity and reflectance spectrum in one figure which does not depend on environmental conditions. However, the RC value cannot differentiate between broadband/narrow-band emitters, nor does it contemplate important aspects such as the angular dependence of the emissivity.

2.2 Cooling power

Cooling power P_{net} (W m^{-2}) is often reported as a result of a PDRC measurement or simulation. It can be calculated as an outcome of an energy balance of the PDRC surface (Eq. 2).

$$P_{\mathrm{net}} = P_{\mathrm{rad}} - P_{\mathrm{atm}} - P_{\mathrm{sun}} - P_{\mathrm{nonrad}} \qquad (2)$$

Symbols on the right side of the equation represent thermal radiation power from the emitter, atmospheric and solar irradiations and contributions of the non-radiative heat transfer processes between the coating and ambient environment, respectively. The accuracy of P_{net} calculation however depends critically on the ability to correctly measure all contributing terms. When said contributions are approximated or neglected, the resulting P_{net} value might be misleading. Additionally, looking at Eq. 2, the final cooling power depends not only on the spectral properties of the material, but also on the experimental testing conditions and the other factors affecting PDRC performance. Therefore, when comparing various PDRC materials, it is not possible to rely solely on its cooling power as this quantity is largely dependent on the measurement conditions. Also, it should be noted that based on Eq. 2, an upper limit value for the cooling power attainable by an ideal PDRC material under ideal conditions (100% solar reflectance, perfect insulation from non-radiative heat gains) can in principle be estimated, which is typically reported at around 150 W m^{-2} [15].

For the experimental measurement of this quantity, Raman *et al.* [16] proposed a method to obtain P_{net} by heating a PDRC material using an external electric heating pad. Cooling power is then determined as the electric power dissipated by the heater when the temperature of the PDRC material equals the ambient temperature of the environment. To measure cooling power of a PDRC coating in real time, a feedback-controlled loop can be added to continuously tune the current flowing through the electric heater in such a way that the temperature of the coating under test matches that of the ambient air at all times. The measured value of the cooling power is correlated with the temperature fluctuations. This indicates that among other influences such as wind speed and solar irradiance, the accumulation of the heat in the measuring box or at the coating surface can also significantly affect the resulting cooling power, which is often reported as an average value from the whole cooling experiment.

This experimental approach to measure the cooling power of PDRC materials has some disadvantages. When reporting the experimental data, it is generally assumed that 100 % of the heat generated by the electric heater is transferred to the emitter and dissipated radiatively by it, i.e., assuming purely one-dimensional heat transfer from the heater to the cooled substrate. Any deviation from this condition can lead to significant cooling power overestimations and exaggerated cooling power claims. Possible solutions to this problem involve using more advanced heating systems with proper thermal guarding, as applied by Leroy *et al.* [17, 18].

Alternatively, the cooling power could be estimated using the controlled flux of a heat-transfer fluid kept in thermal contact with the backside of the emitter, as first exemplified in the work of Goldstein *et al.* [19]. An advantage of this method is that the inlet and outlet water temperatures can be measured with high accuracy, as well as the instantaneous flux. Moreover, the measured value would likely represent a conservative estimate, since any deviation from the ideal insulation of the system would result in an underestimation of the cooling power, rather than an overestimation.

2.3 Temperature reporting

The most basic characterization used during PDRC experimental tests is a direct temperature measurement. Several thermocouples are often used to monitor the temperature of the PDRC material surface or the substrates cooled down by it (T_s), to be compared with ambient temperature (T_{amb}). Most of the studies provide the results by showing just the lowest achieved temperature or more commonly a maximum temperature difference between T_s and T_{amb} (maximum temperature drop - MTD). The information about the temperature difference is beneficial for comparing various PDRC materials tested in similar working conditions or experiments with similar PDRC materials tested in various working conditions, but cannot be used for a more universal material comparison. Besides temperatures being strongly dependent on the experimental conditions, several shortcomings are also associated with the determination of T_{amb}, since its measurement depends critically on the placement of the thermocouple and its proper shielding from other external factors. Many of the exceptionally high temperature differences reported in the literature can probably be ascribed to significant overestimations of T_{amb}, obtained by measuring it under direct solar radiation, with insufficient shielding from other radiative and convective heat sources or by replacing T_{amb} with the “enclosure air temperature”. When reporting the obtained temperature difference as an indicator of PDRC performance, it is therefore crucial to ensure accurate and consistent T_{amb} measurement [20]. This topic is also discussed in depth in a Viewpoint by Sui *et al.* [21], as well as other thermodynamic fundamentals of PDRC.

The term equilibrium temperature T_{eq} is also used for reporting in the literature. Most studies use this term referring to temperatures obtained under zero net cooling power conditions. Looking at Eq. 2, P_{net} and P_{rad} depend on the temperature of the material surface T_s, whereas P_{atm} depends on the ambient temperature T_{amb}. T_{eq} is therefore equal to T_s when cooling power P_{net} (T_s) = 0. Reaching the equilibrium condition can lead to T_{eq} being either lower or higher than T_{amb}. Ao *et al.* [22] considers the equilibrium temperature as a key parameter for PDRC performance, together with cooling power P_{net} at the ambient temperature T_{amb}. Working conditions of a PDRC material can be set either to reach the lowest equilibrium temperature T_{eq} or to obtain the highest cooling power P_{net} possible. As discussed before, spectral properties are the main parameter that can tune the performance of the PDRC material [23].

As a final remark, it should be noted that values of cooling power at T_{amb} and T_{eq} at $P_{net} = 0$ are typically presented as two alternative methods for the evaluation of the cooling performance of a PDRC material. However, their interpretation can be unified by considering them as two points on a single curve describing the dependence of the cooler temperature on the achieved cooling power. To obtain a more comprehensive characterisation of the cooler, more points of this curve should be reported [16], since the performance of PDRC materials can depend on the temperature of the emitter. Measuring more data points on the curve is on the other hand problematic due to the time needed for the temperature stabilisation of each value, which can lead to changes in external conditions and therefore to a distortion of the data, unless multiple copies of the same sample are tested simultaneously under different heating conditions

3 Conclusions

Summarizing of the current studies published on the topic of PDRC showed that the identification of universal figures of merit for PDRC materials remains as one of the major challenges in the field. Most of the studies suggest considering two basic indicators: maximum sub-ambient temperature drop and cooling power at ambient temperature, but a whole set of experimental conditions is needed to fully describe the PDRC performance of a given material. Some numerical methods have been suggested to approximately compare cooling materials using standard model atmospheres and other simplifications. However, for a more precise and effective comparison, the implementation of standard indoor or outdoor experimental test methods is needed in the scientific community, as well as the identification of a set of established PDRC materials with known properties [24], that should be tested in parallel with the other materials under study.

Acknowledgements

The project 21GRD03 PaRaMetriC received funding from the European Partnership on Metrology, co-financed by the European Union's Horizon Europe Research and Innovation Programme and from the Participating States.

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