

Supporting Information

Homogeneous Polymer Films for Passive Daytime Cooling: Optimized Thickness for Maximized Cooling Performance

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SI 1. Convergence of the resulting calculated cooling power

To verify the convergence of the resulting cooling power as a function of the upper integration boundary, we map out the corner points of the thickness-temperature space shown in Figure 4. Namely, those are thicknesses of $1 \cdot 10^{-7}$ m and $2 \cdot 10^{-3}$ m and temperatures differences of 0 K and 12.5 K relative to ambient.

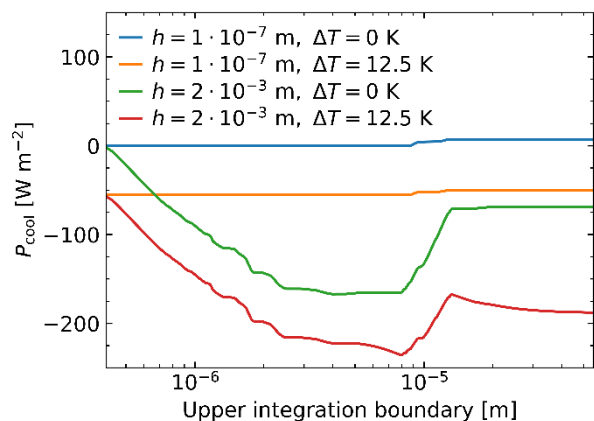


Figure S1: Convergence of the integrated total cooling power as a function of upper integration boundary for the points enclosing the analyzed temperature and thickness space.

Convergence of the resulting cooling power as a function of the upper integration can be seen. For high thickness and temperature differences, maybe higher integration boundaries might be needed, but for the calculations performed in this work, the upper integration boundary of 55 μm is sufficient.

SI 2. Influence of the non-radiative heat transfer coefficient

To analyze the influence of the comprehensive heat transfer coefficient due to convection and conduction, several coefficients are used spanning two orders of magnitude. Regardless of the absolute value, an optimum thickness is always apparent. As intrinsic losses increase, smaller temperature differences to ambient are apparent, as to be expected.

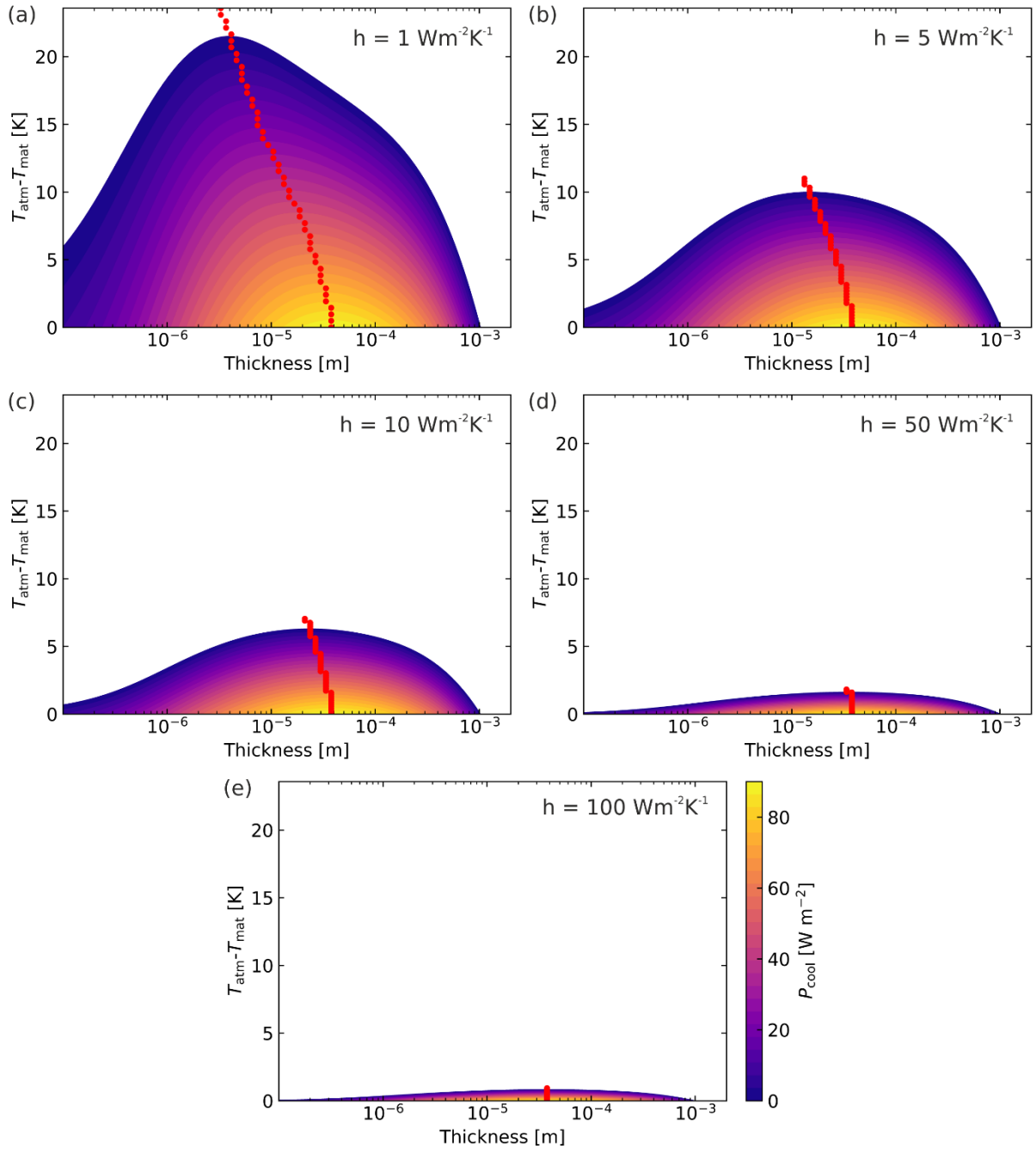


Figure S2: Influence of the non-radiative heat transfer coefficient on the net cooling power for PDMS.

SI 3. Optimum emitter thickness during night-time

Only above a certain temperature difference threshold, an optimum thickness in a similar order of magnitude as for the daytime case becomes apparent. This maximum has a different origin compared to the daytime case. As the emitter temperature decreases, the power radiated decreases due to Planck's law of radiation. Above a certain threshold, for PDMS approximately 3 K below ambient, the derivatives regarding the thickness of the power absorbed and emitted have an intersection point. This intersection point represents a zero crossing in the derivative of the net cooling power, and in this case, a maximum cooling power. To better illustrate this effect, we focus on a temperature difference of 0 K and 10 K relative to ambient temperature.

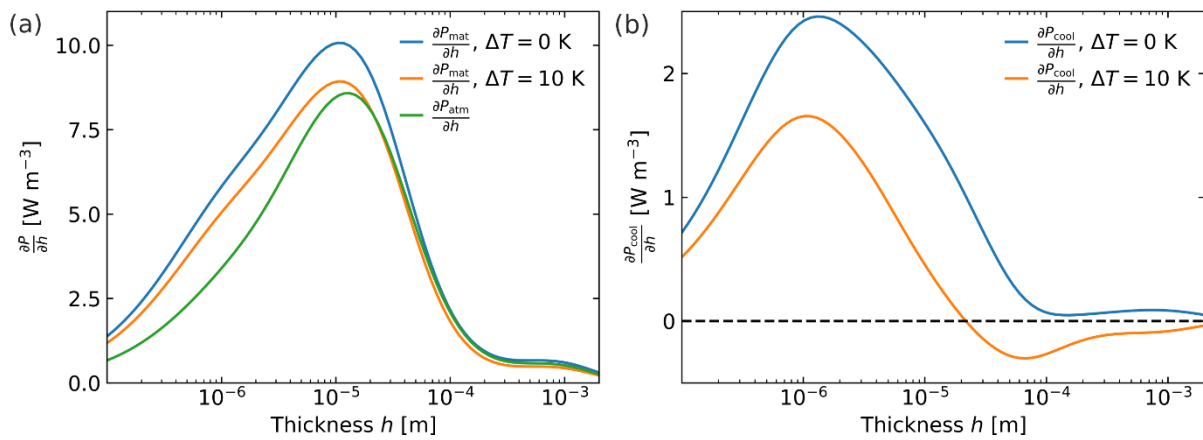


Figure S3: Numerical derivatives of the individual energetic contributions of material and atmosphere as a function of thickness during night-time (a), as well as the total cooling power (b) with respect to thickness. For $\Delta T = 10$ K, there is an optimum thickness apparent, while there is none for $\Delta T = 0$ K.

At ambient temperature, the derivative of the power emitted is continuously higher than the derivative of the power absorbed by the atmosphere. As the emitter temperature decreases, above a certain thickness, here approximately 20 μm , the derivative of the power absorbed by the atmosphere exceeds the derivative of the power emitted. This point represents a zero crossing of the total cooling power, depicted in Figure S3b, and in this case, a maximum cooling power that is not apparent at ambient temperature.

SI 4. Verification of the calculations by means of further materials

We also performed calculations based on the complex refractive index of other polymeric materials to verify the optimum thickness effect for passive cooling in back-reflector geometry.^[1] For polystyrene, polymethyl methacrylate, and polyethylene terephthalate, the cooling powers as a function of thickness and temperature are shown in Figure S4.

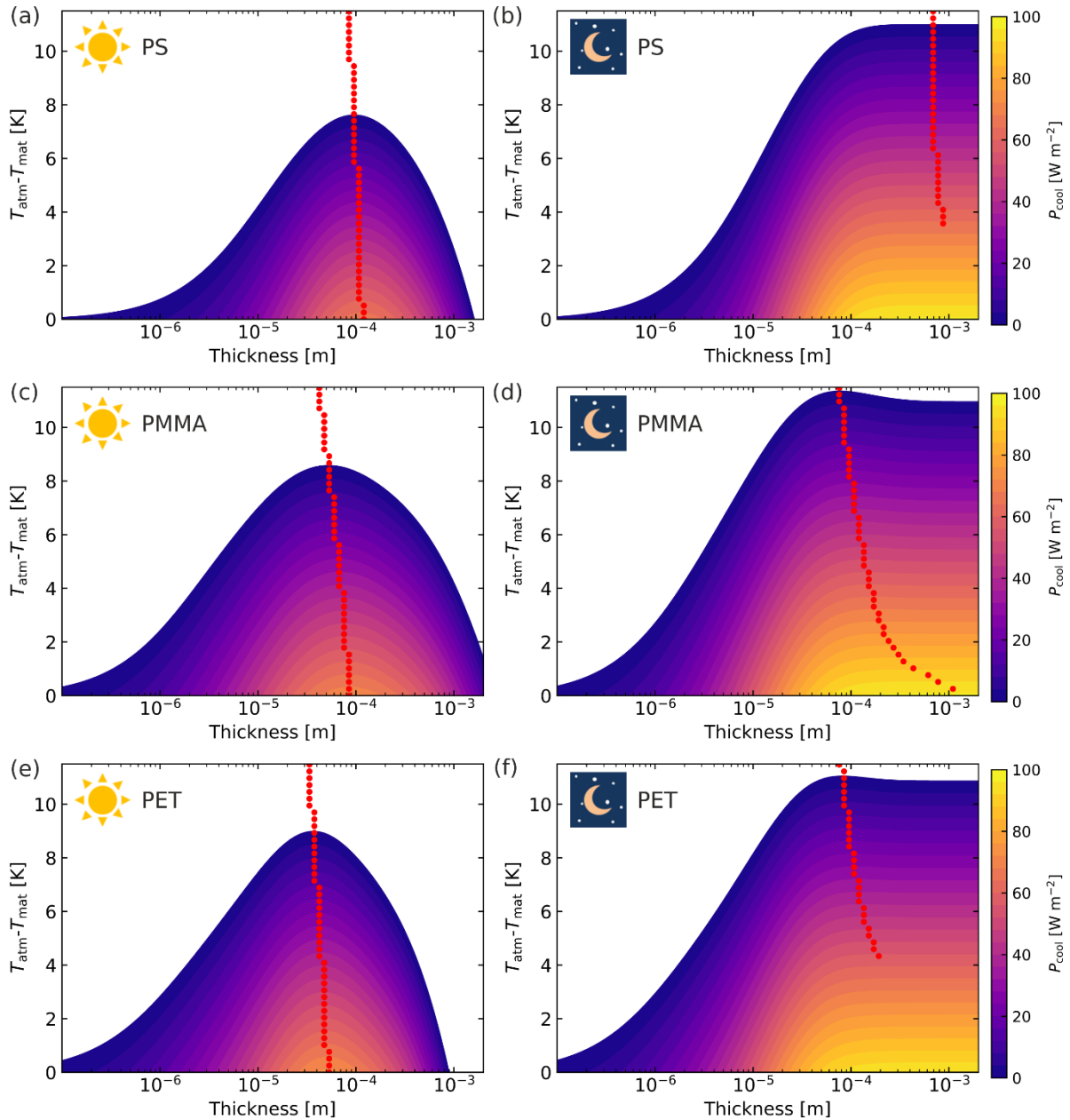


Figure S4: Resulting cooling power as a function of thickness and emitter temperature, as well as highlighted thickness (red dots) with the highest cooling power at the respective temperature, during daytime and night-time for polystyrene (a), (b) polymethyl methacrylate (c), (d) and polyethylene terephthalate (e), (f).

All materials investigated exhibit an optimum thickness during daytime, while the position, as well as the absolute cooling powers, vary. The same can be seen during night-time, whereas the temperature threshold below which an optimum thickness becomes apparent is also dependent on the emitter material. In summary, this can be interpreted as a general confirmation of the effect.

SI 5. Complex refractive index data as a source of error

The primary source of error, in addition to the atmospheric and solar spectrum, are the complex refractive index data of the investigated emitter material. A comparison of the measured absorptance spectrum and the expected absorptance based on the measured thickness and the complex refractive index is shown in Figure S5.

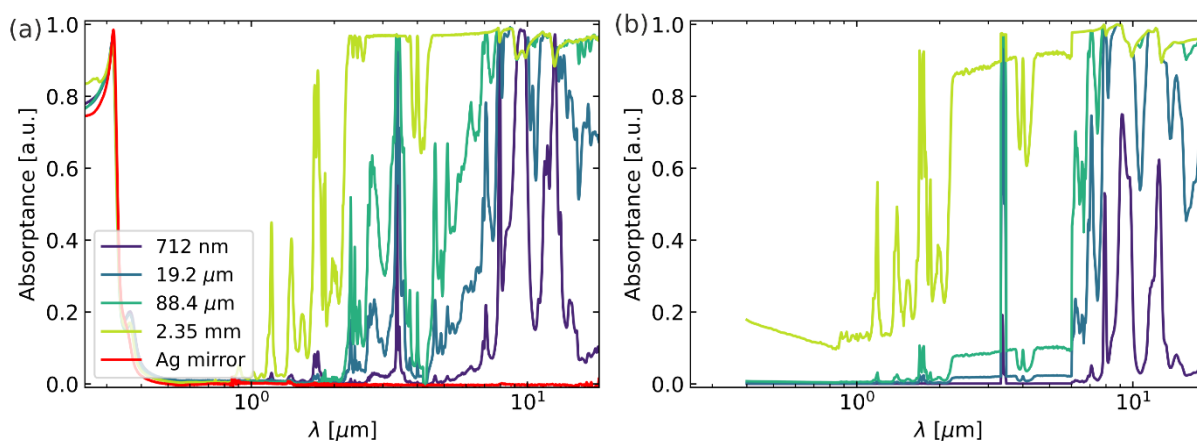


Figure S5: Measured absorptance (a) in comparison to the estimated absorptance based on the complex refractive index data and thickness (b) for an angle of incidence of 8° .

A qualitative agreement between measured and estimated absorptance spectrum is apparent, while the absolute values exhibit clear deviations. The main drawback is the overestimation of the absorption in the solar regime, leading to an optimum cooling power at lower thicknesses. For further work based on this approach, accurate complex refractive index data are, therefore, crucial.

SI 6. Verification of the measurement setup

To determine errors induced by the measurement setup, a measurement with five identical Al mirrors was performed.

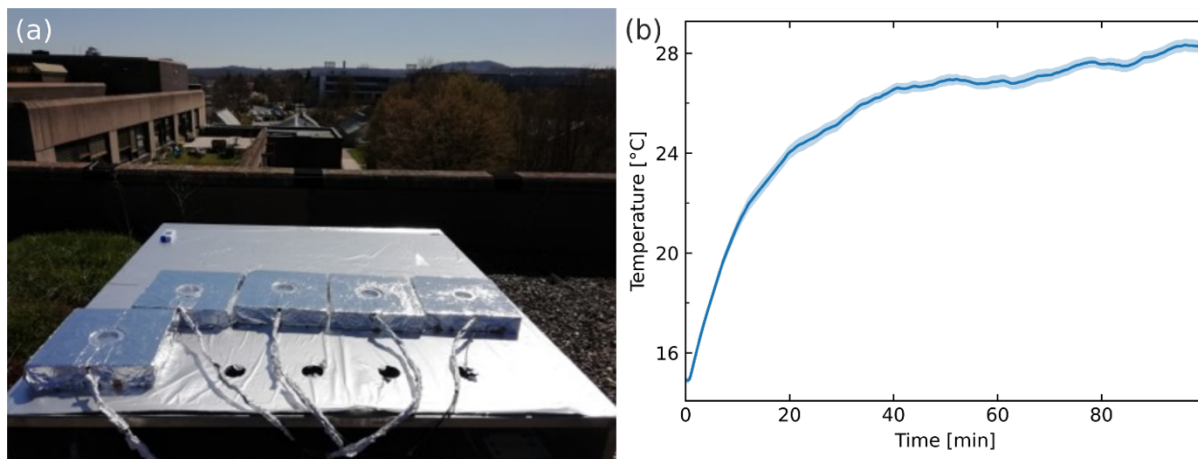


Figure S6: (a) photography of the setups used to measure the samples' daytime cooling performance. (b) Comparison of sample temperatures for the samples during the measurement time. Mean and standard deviation of five identical Al mirrors under the average solar irradiation of approximately 856 W/m^2 . (Start at 12:20, 23rd April 2021, University of Bayreuth, Bayreuth, Germany)

As the identical Al mirrors exhibit a maximum standard deviation of $\pm 0.3 \text{ K}$, not to overstate our results an error of the measurement setup of $\pm 0.5 \text{ K}$ is assumed.

References

- [1] a) X. Zhang, J. Qiu, X. Li, J. Zhao, L. Liu, *J. Quant. Spectrosc. Radiat. Transfer* **2020**, 252, 107063. b) X. Zhang, J. Qiu, X. Li, J. Zhao, L. Liu, *Appl. Opt.* **2020**, 59 (8), 2337-2344.