

Additional file

**Highly efficient flexible structured metasurface by roll-to-roll printing
for diurnal radiative cooling**

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Note S1: Numerical simulation.

In simulations, we set the plan waves with wavelengths from 0.28 to 25 μm propagating from 40 μm above the three-dimensional (3D) trench-like polymer metasurface radiative cooling (PMRC) film. To further study the absorption/emission characteristics, we set up two detectors for each structured polymer-based radiative cooling film: the first, 1 μm below the Ag–Air interface (in the air); the second, 1 μm above the incident light (in the air). These two detectors recorded the transmittance (T) and reflectance (R) of the 3D trench-like structured polymer-based radiative cooling film, respectively, from which we could calculate the absorbance/emissivity (A) by the equation $A=1-R-T$. In the simulations, we obtained the optical constants for silver (Ag) and polymers from the literature [1-3]. The absorption/emissivity of the 3D trench-like PMRC film under the unpolarized light was calculated by averaging the absorptions of the absorber under illumination with transverse electric (TE)- and transverse magnetic (TM)-polarized light.

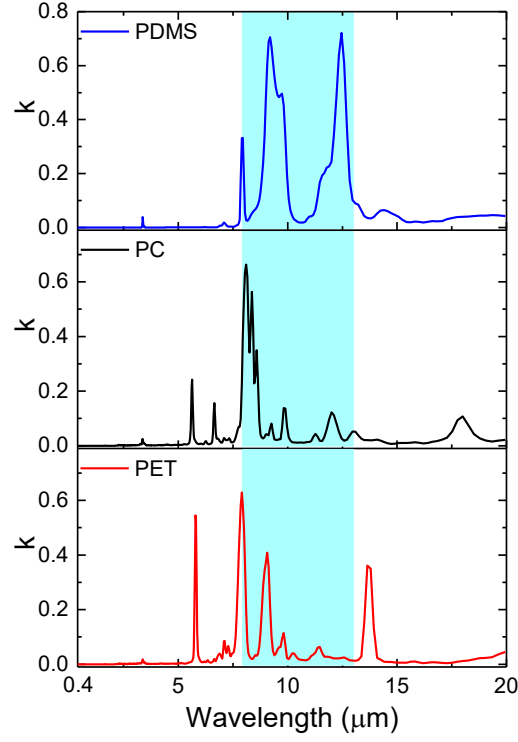


Figure S1. Extinction coefficients (k) of polydimethylsiloxane (PDMS), polycarbonate (PC), and polyethylene terephthalate (PET) [1, 2]. In general, the absorption coefficients ($\alpha = \frac{4\pi k}{\lambda}$) of a material are proportional to its extinction coefficients. Therefore, these polymers possess absorption capability in the main atmospheric transparency window (i.e., 8–13 μm) while low absorption in most of the solar spectral range.

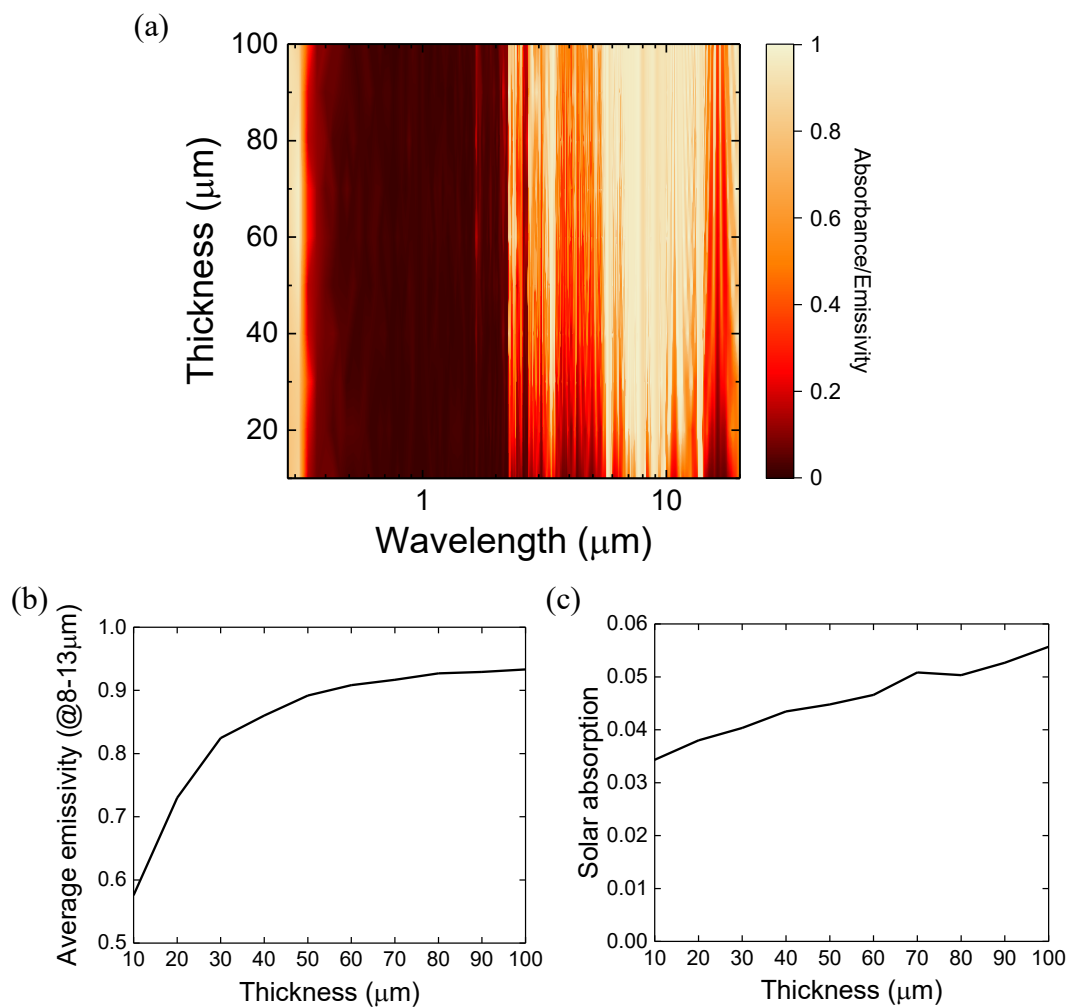


Figure S2. (a) Absorbance/emissivity spectra, (b) average emissivity in the main atmospheric transparency window (i.e., 8–13 μm), and (c) solar absorption of PET film with 100-nm silver (Ag) layer coated on the backside and with different thicknesses.

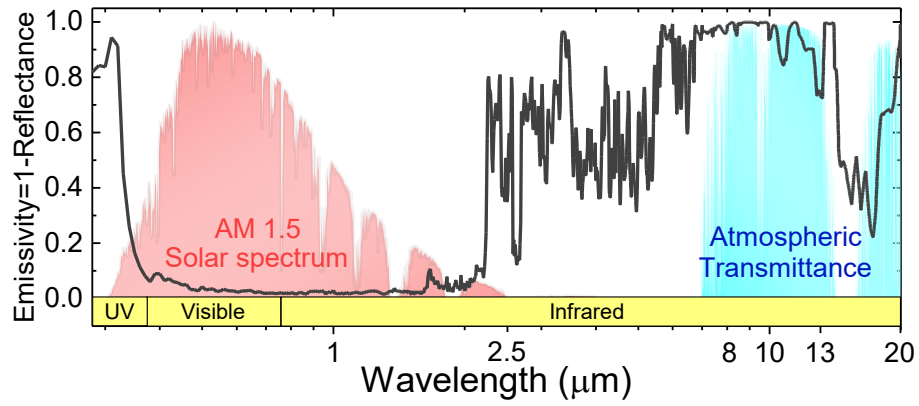


Figure S3. Simulated spectral absorbance/emissivity for the PMRC film

featuring a trench width of 6.5 μm , a periodicity of 8 μm , a depth of trenches of 2.5 μm , and thicknesses of PET film and Ag layer of 50 μm and 100 nm, respectively. An ideal radiative cooler is able to efficiently emit (near unity) thermal radiation in the atmospheric transparency window (shaded blue) and minimize solar absorption in the solar spectral range (shaded red).

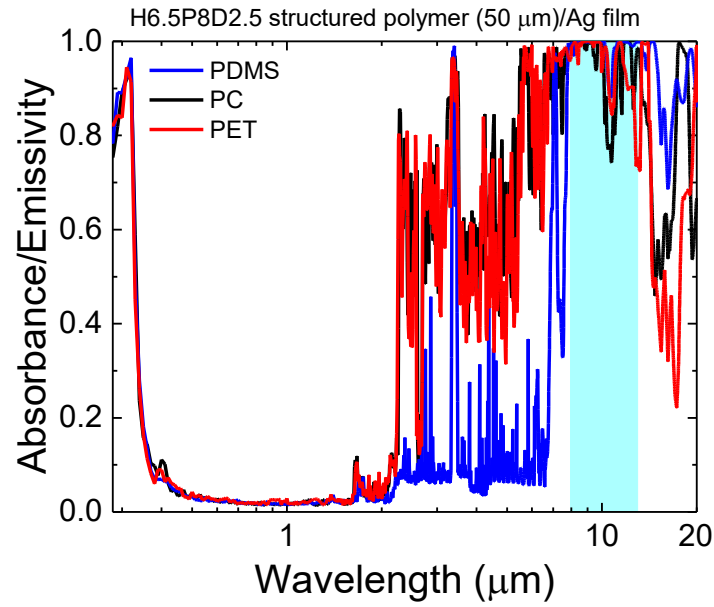


Figure S4. Simulated absorbance/emissivity spectra of the 3D trench-like structured PDMS (blue line), PC (black line), and PET (red line) films with 100-nm Ag layer deposited on the backside. The structural parameters of these radiative cooling films are the width and depth of trenches of 6.5 and 2.5 μm , respectively, the periodicity of 8 μm , and the thickness of polymer film of 50 μm .

Note S2: Thermal resistance.

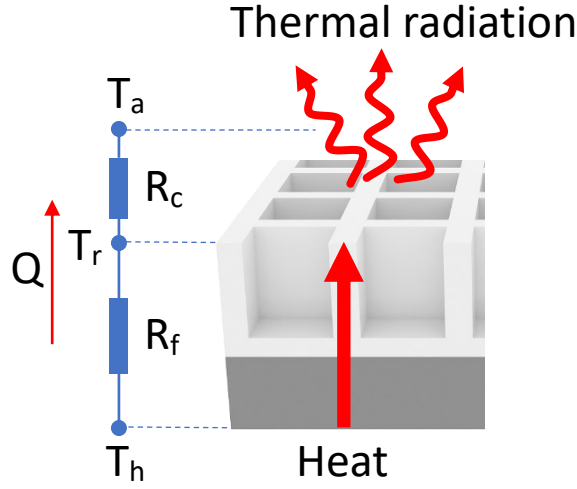


Figure S5. Heat resistance model of the designed PMRC film.

Considering the cooling scheme shown in Supplementary Fig. S5. The thermal process of radiative cooling is that the radiation emits from the surface of the radiative cooler, which depends on the surface temperature of the cooler (T_r). Due to the cooler's thermal resistance (R_f), the temperature at the bottom of the cooler is T_h . In the application, the object's temperature depends on the temperature at the bottom of the cooler, as the cooler will be attached to the object's surface to face the sky. The relationship between T_r and T_h can be expressed as

$$T_r = T_h - P_{net}AR_f \quad (6)$$

A is the area of the cooler. In an ideal situation, the T_r should be as close to T_h as possible to get high cooling power according to Eqs. (1)-(3). On the one hand, it is desired to achieve high net cooling power P_{net} . On the other hand, minimizing the thermal resistance of the radiative cooler R_f is necessary to achieve T_r close to T_h . The thermal resistance can be expressed as

$$R_f = \frac{d}{A \times k} \quad (7)$$

where d is the thickness of the film, and k is the thermal conductivity. By substituting Eq. (7) into Eq. (6), one can get

$$T_r = T_h - \frac{P_{net}d}{k} \quad (8)$$

Therefore, to achieve high cooling power for a given thermal conductivity, minimizing the d of the cooler is necessary. Meanwhile, to cool down to a specific temperature, the heat that needs to be dissipated is Q , which is the sum of the heat of the radiative cooler (Q_1) and the object (Q_2). Therefore, it is necessary to minimize the heat of the cooler (Q_1), which can be expressed as $Q_1 = cm\Delta T$, where ΔT is the temperature difference before and after cooling, c is the specific heat capacity of the material, and m is the mass of the material. For the radiative cooler, the mass is $m = \rho \times A \times d$, where ρ is the density of the material. Therefore, the heat in the film is proportional to the thickness of the film. According to both considerations, decreasing the cooler thickness d is beneficial. Thus, an ideal radiative cooler should be thinner from the thermal extraction point of view.

We design thin polymer metasurface structures in a polymer substrate to allow the incident light to be critically coupled [4] into the polymer and eventually propagate along the film to be absorbed, which can achieve high absorption within ultrathin polymer films (a few tens of microns). As a result, the thermal resistance can be further decreased according to Eq. (7).

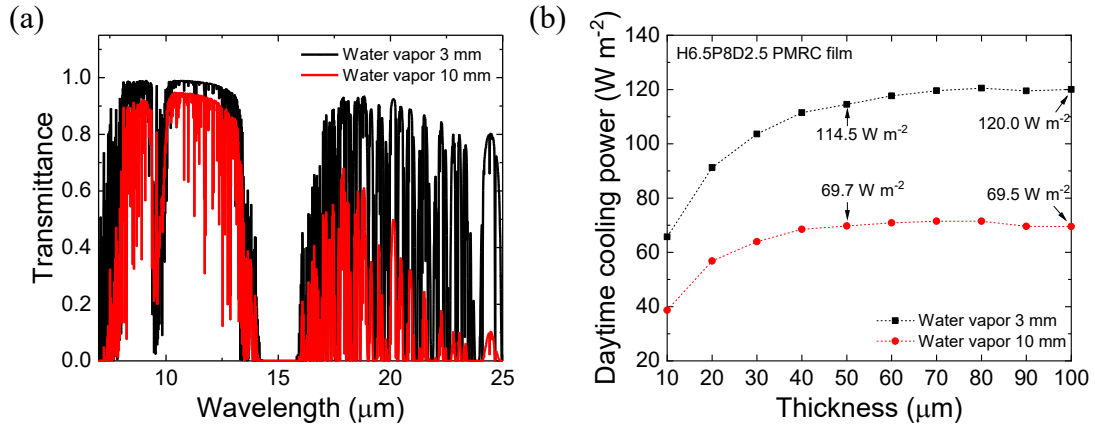


Figure S6. (a) Transmittance spectra of the atmosphere with the amount of water vapor of 3 mm (black line) and 10 mm (red line) under AM 1.5 [5]. (b) Calculated daytime cooling powers of the H6.5P8D2.5 PMRC film with different thicknesses of PET film at T_a and $T_r = 300$ K, $H_c = 6.9 \text{ W m}^{-2} \text{ K}^{-1}$, the power density of the sun of 1000 W m^{-2} , and under the amount of water vapor of 3 mm (black dot) and 10 mm (red dot).

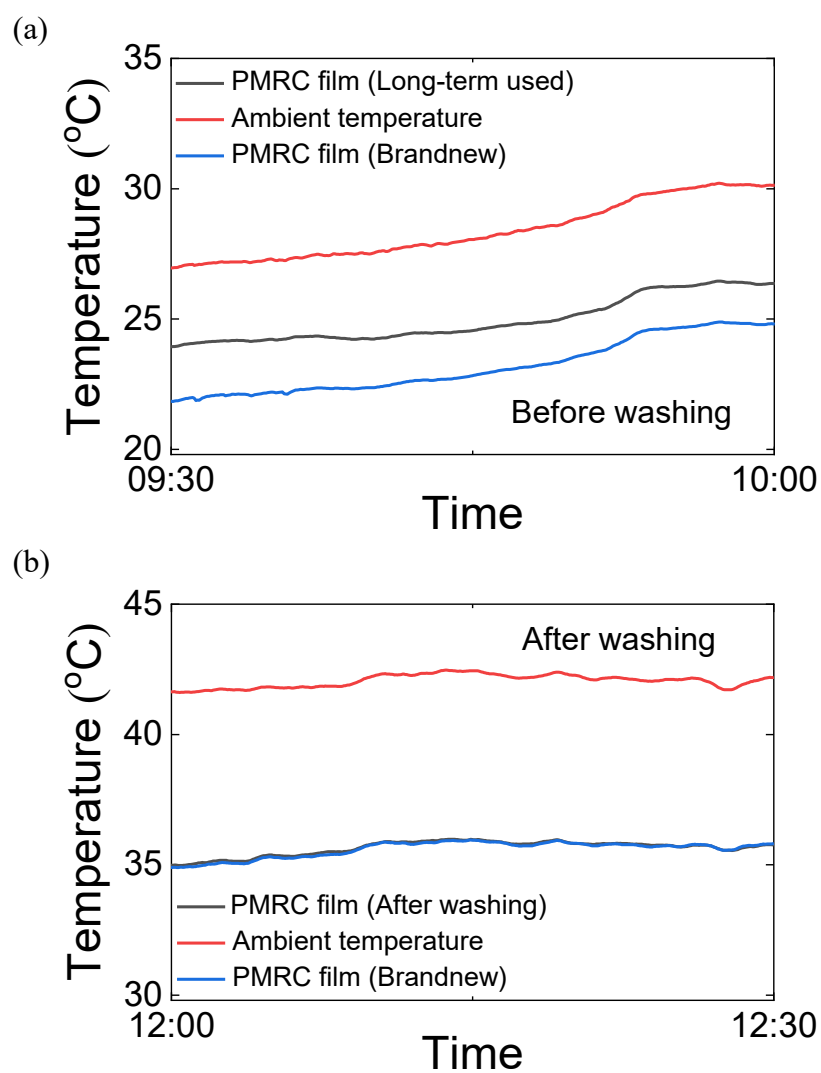


Figure S7. (a, b) Real-time temperatures of long-term used PMRC film

and ambient temperatures under natural sunlight (a) before and (b) after washing alcohol, acetone, and water.

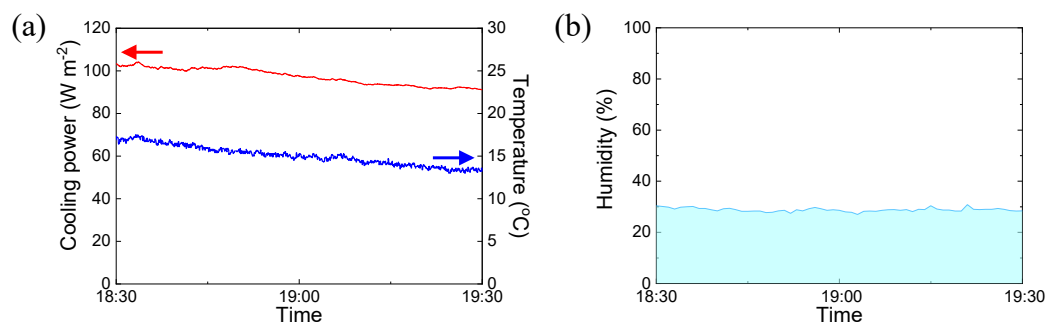


Figure S8. Measured (a) cooling powers of the PMRC film (red line) and ambient temperatures (blue line), and (b) humidity level during nighttime on a clear sky day in Weihai city.

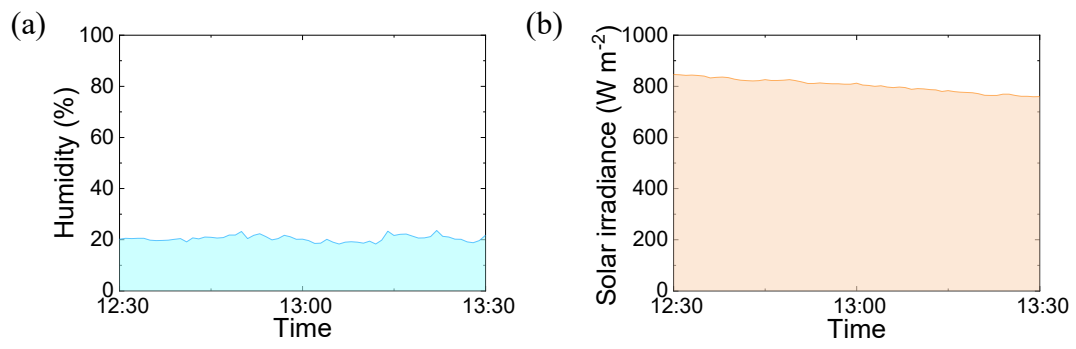


Figure S9. Measured (a) humidity level, and (b) solar irradiance during midday on a clear sky day in Weihai city.

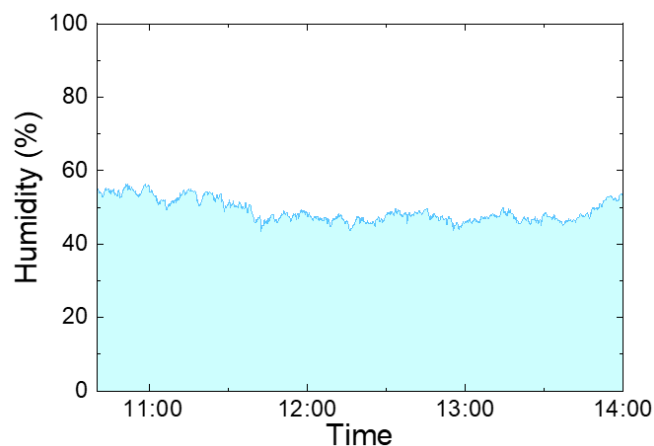


Figure S10. Measured humidity level during the daytime in Weihai city.

In the experiment, the initial water temperature and the amount of water in both fluidic devices with and without attaching PMRC are the same. The water temperatures in the integrated fluidic device are lower than those of the surroundings during the measurement. Moreover, the difference in water temperature between the integrated fluidic device and the fluidic device without attaching PMRC reaches up to 11.5 °C after 3.25-hour solar illumination.

References

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