Supplementary Information

Self-adaptive and large-area sprayable thermal management coatings for energy saving

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Supplementary Note

1. Calculation of the average solar reflectivity ($\bar{R}_{Solar}$) and average atmospheric window emissivity ($\bar{\varepsilon}_{LWIR}$)

The definition of average solar reflectivity ($\bar{R}_{Solar}$) is given by:

$$\bar{R}_{solar} = \frac{\int_{0.3\mu m}^{2.5\mu m} I_{solar}(\lambda)\times R_{solar}(\lambda)d\lambda}{\int_{0.3\mu m}^{2.5\mu m} I_{solar}(\lambda)d\lambda} \tag{1}$$

where λ is the wavelength, $I_{solar}(\lambda)$ is the reference direct normal spectral irradiance ASTM 1.5G, representing the global solar intensity, and $R_{solar}(\lambda)$ is the measured reflectivity of the CCTM film.

The definition of average atmospheric window emissivity ($\bar{\varepsilon}_{LWIR}$) is given by:

$$\bar{\varepsilon}_{LWIR} = \frac{\int_{8\mu m}^{13\mu m} I_{BB}(T,\lambda)\times \varepsilon(T,\lambda)d\lambda}{\int_{8\mu m}^{13\mu m} I_{BB}(T,\lambda)d\lambda} \tag{2}$$

where T is the temperature of a blackbody, $\varepsilon(T,\lambda)$ is the thermal emissivity spectrum of the CCTM film. $I_{bb}(\lambda,T)$ is the irradiance intensity of the blackbody at temperature T, according to Planck's law:

$$\mathrm{I}_{BB}(\lambda,T) = \frac{2hc^2}{\lambda^5}\frac{1}{e^{\frac{hc}{\lambda kT}}-1} \tag{3}$$

where h is the Planck constant, c is the velocity of light in vacuum, k is the Boltzmann's constant.

2. Calculation of the radiation cooling power of the CCTM film

The steady state heat transfer equilibrium process of the CCTM film illustrated is in Fig. 4a, where P_{rad} is the energy emitted from the surface of CCTM film, P_{sky} is absorbed energy from the atmospheric radiation, P_{solar} is the absorbed solar energy, and $P_{cond\text{-}conv}$ is the non-radiative losses from the surrounding environmental conductive and/or convective heat exchange.

For a radiative cooling CCTM film, the net cooling power (P_{net}) can be calculated by:

$$P_{net} = P_{rad}\left(T_r\right) - P_{sky} - P_{solar} - P_{cond-conv} \tag{4}$$

where T_r is the radiation temperature (the surface temperature of the film). Hence, P_{rad} can be described as follows:

$$P_{rad} = \pi \int_0^{+\infty} \int_0^{\frac{\pi}{2}} \varepsilon_r(\lambda,\theta) I_{bb}(\lambda,T_r) \sin(2\theta) d\theta d\lambda \tag{5}$$

where $\varepsilon_r(\lambda,\theta)$ is the emissivity of the CCTM film in the range of MIR. $I_{bb}(\lambda,T_r)$ is the irradiance intensity of the blackbody at temperature T_r.

According to the thermal radiation principles, the absorbed sky radiation energy can be calculated by:

$$P_{sky} = \pi \int_0^{+\infty} \int_0^{\frac{\pi}{2}} \alpha_r(\lambda,\theta) I_s(\lambda,\theta,T) \sin(2\theta) d\theta d\lambda \tag{6}$$

where $\alpha_r(\lambda,\theta)$ is the absorbance of the CCTM film and equals to the emissivity of the film $\varepsilon_r(\lambda,\theta)$ according to Kirchhoff's law of thermal radiation. In addition, the radiation energy of sky can be given by:

$$I_s(\lambda,\theta,T) = \varepsilon_s(\lambda,\theta) I_b(\lambda,T_\alpha) \tag{7}$$

where T_a is the ambient temperature, $\varepsilon_s(\lambda,\theta)$ is the emissivity of sky, which can be taken as:

$$\varepsilon_s(\lambda,\theta) = 1 - [\tau_s(\lambda,0)]^{1/\cos(\theta)} \tag{8}$$

where $\tau_s(\lambda,0)$ represents the sky transmissivity at vertical direction.

The absorbed solar energy P_{solar} can be described as follows:

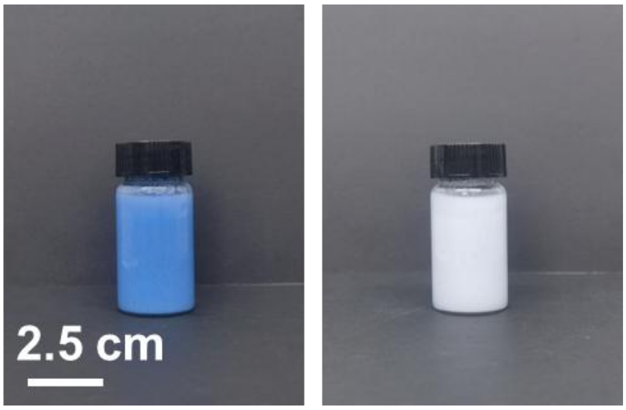
$$P_{solar} = \int_0^{+\infty} \alpha_r(\lambda,\theta_{sun}) I_{AM1.5}(\lambda) d\lambda \tag{9}$$

where θ_{sun} is the incident angle of sunlight on the film, $I_{AM1.5}(\lambda)$ is the reference direct normal spectral irradiance ASTM 1.5G.

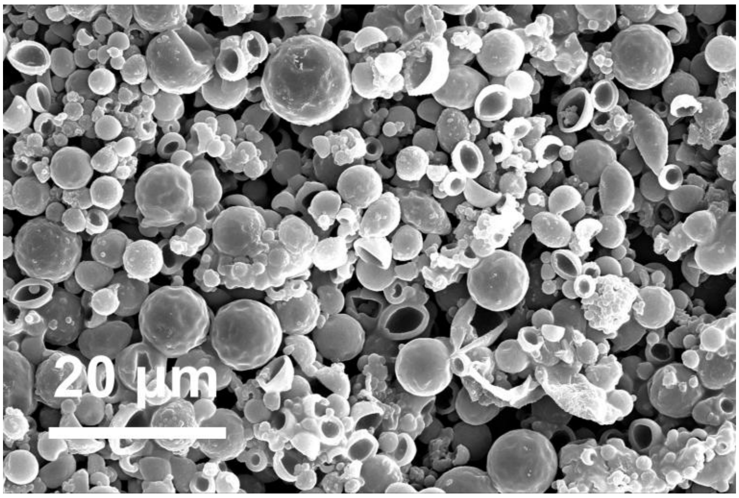
The non-radiative energy losses $P_{cond-conv}$ can be expressed as:

$$P_{cond-conv} = q(T_a - T_r) \tag{10}$$

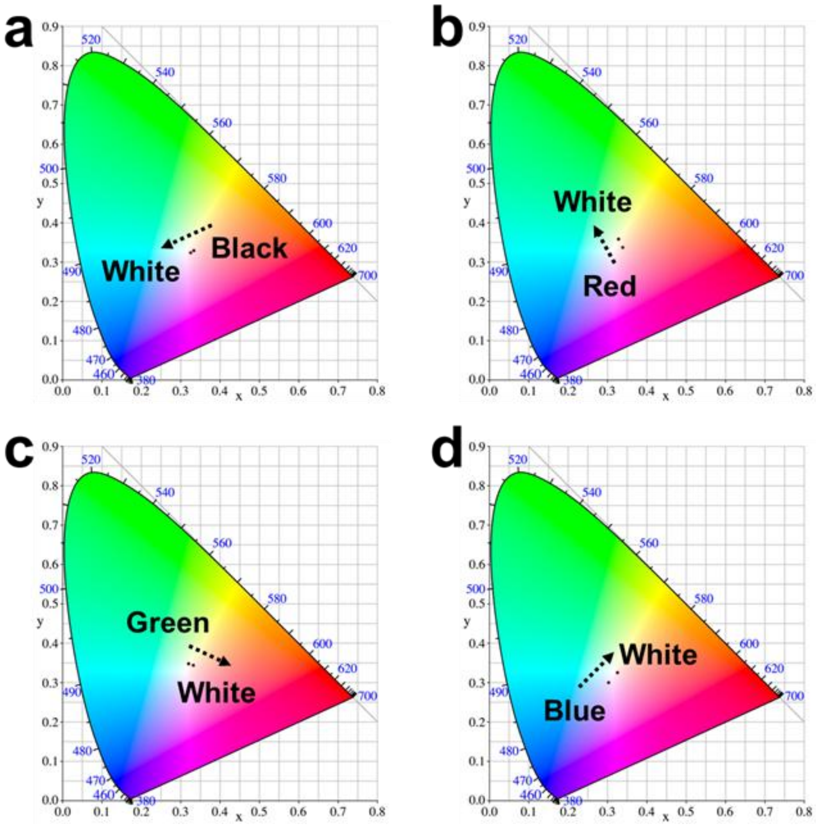
where q is the non-radiative heat coefficient composed of the thermal conduction and convection.



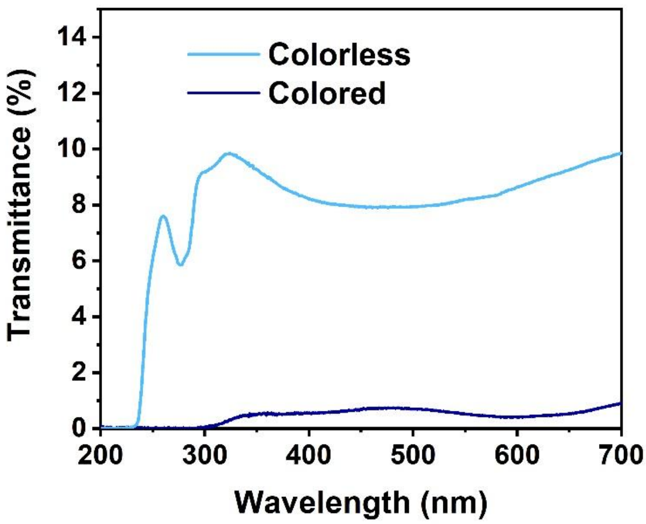
Supplementary Fig. 1. Digital photos of CCTM suspension at low temperatures (colored) and high temperatures (colorless), respectively.



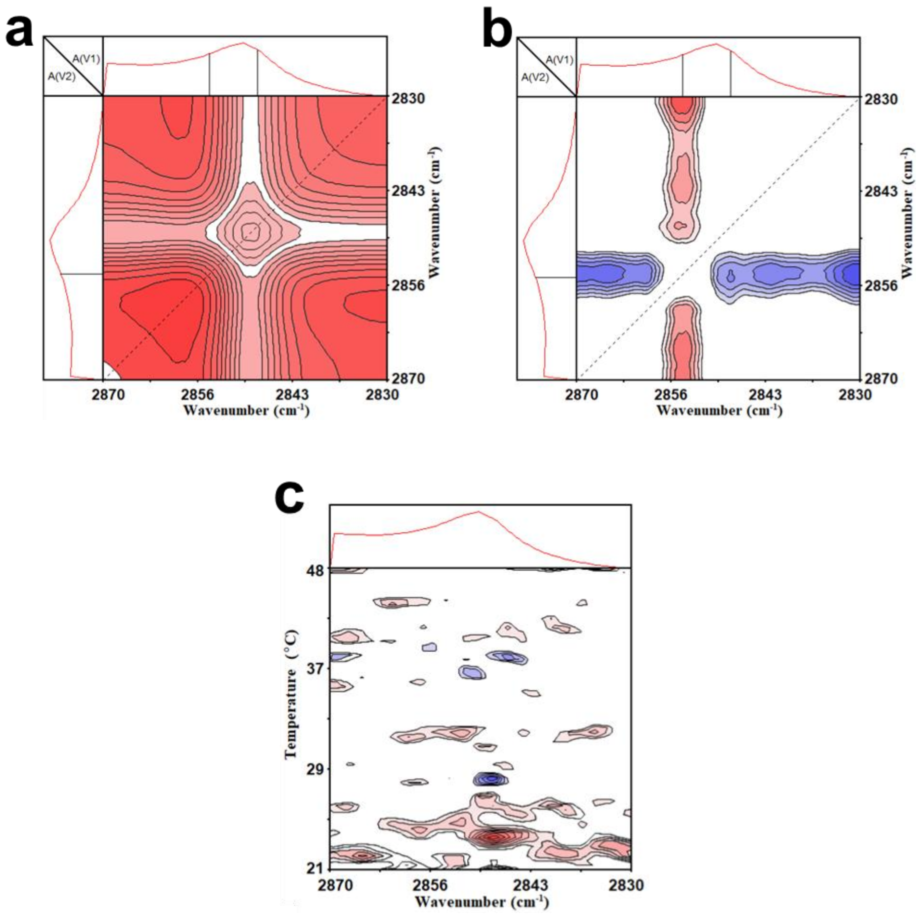
Supplementary Fig. 2. The SEM image of TMP.



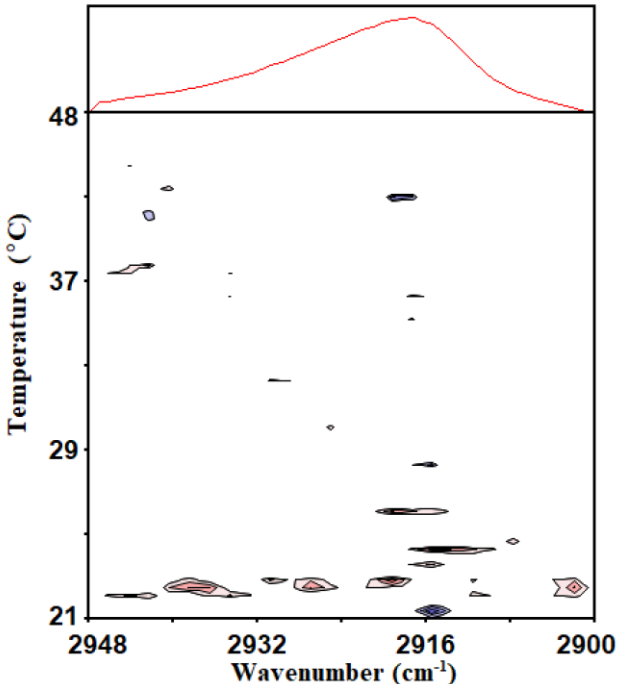
Supplementary Fig. 3. The CIE chromaticity coordinates of four samples.



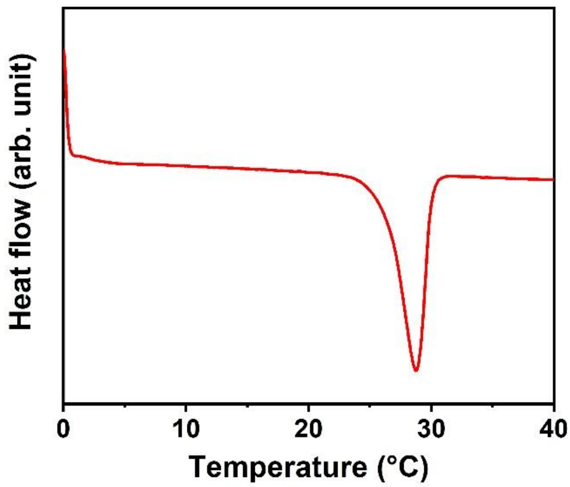
Supplementary Fig. 4. UV spectrums of the TMP suspension at high temperatures (colorless) and low temperatures (colored), respectively.



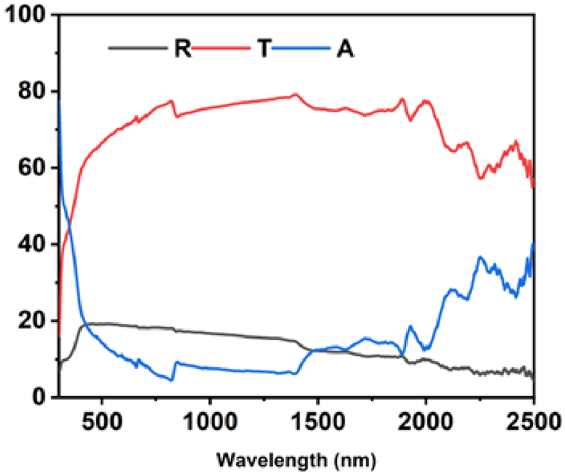
Supplementary Fig. 5. a and b Moving-window two-dimensional correlation spectra and **c** accurate association temperature of–OH groups at 2852 cm^{-1}.



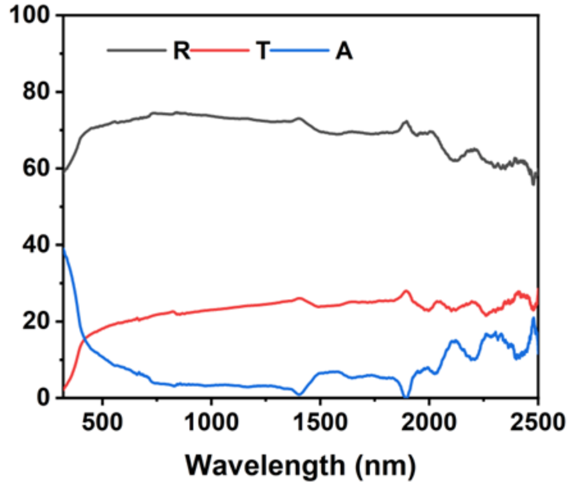
Supplementary Fig. 6. The association temperature of −CH groups at 2915 cm^{-1}.



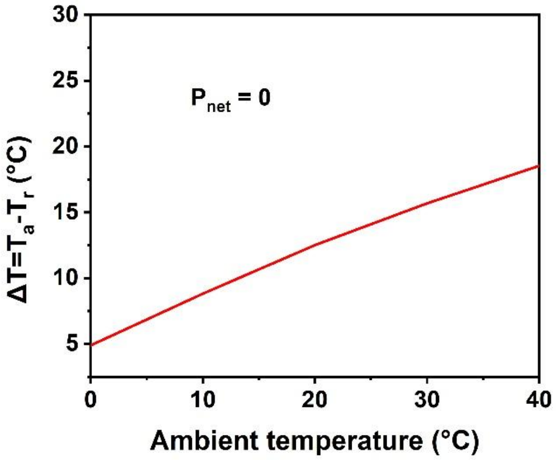
Supplementary Fig. 7. DSC curves of the blue CCTM coating upon increasing temperature.



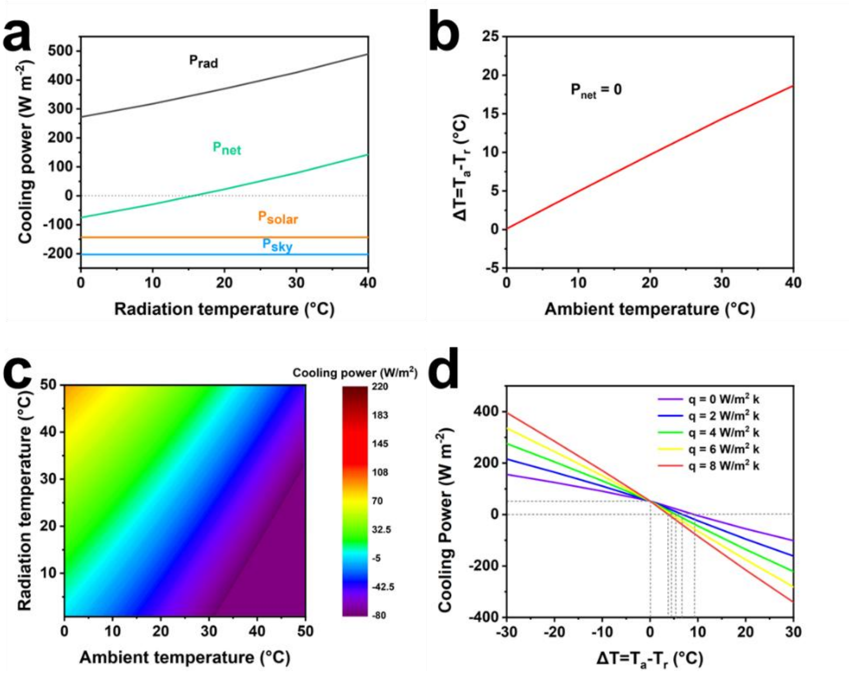
Supplementary Fig. 8. Spectral solar reflectivity and emittance of C-CNF coating. R, T, A represent reflectivity, transmissivity and absorptivity, respectively.



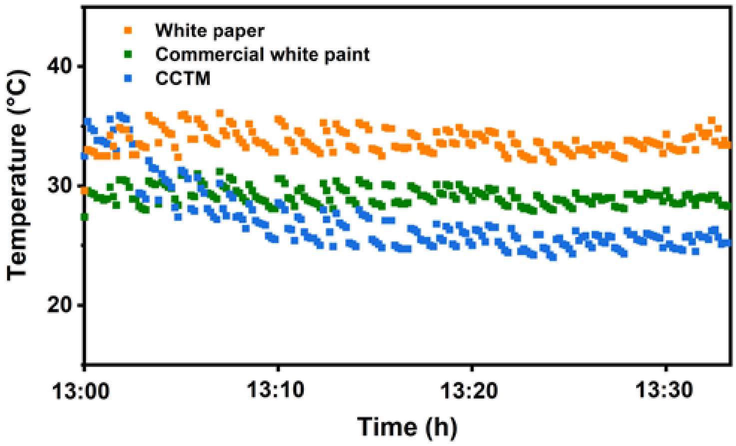
Supplementary Fig. 9. Spectral solar reflectivity and emittance of C-CNF/$CaCO_3$ coating**.** R, T, A represent reflectivity, transmissivity and absorptivity, respectively.



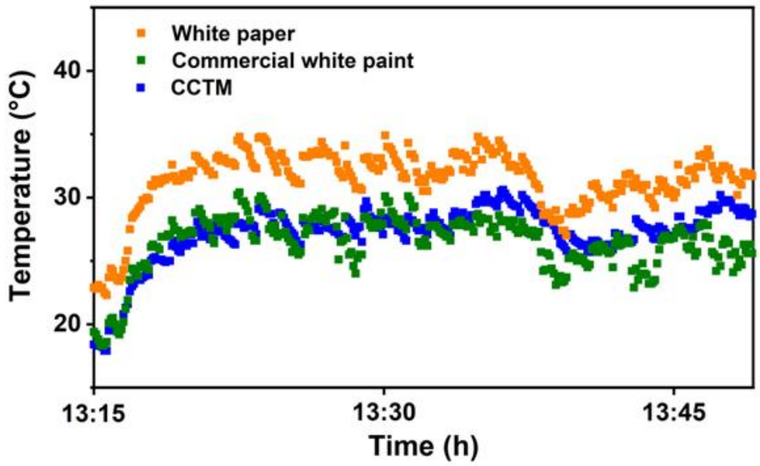
Supplementary Fig. 10. The ΔT as a function of the ambient temperature while the net radiative cooling power is zero.



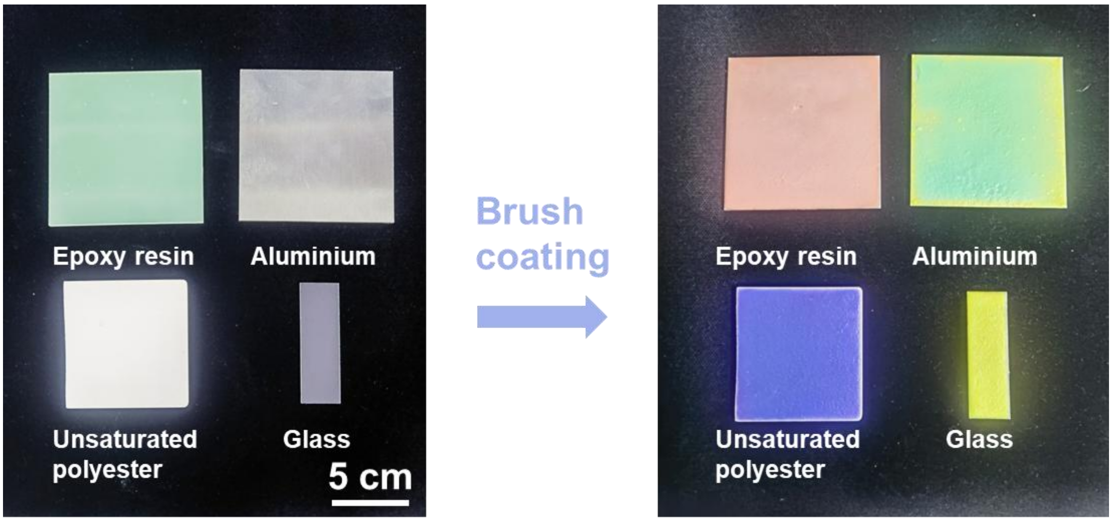
Supplementary Fig. 11. a Estimations of the cooling powers for a CCTM coating under ideal condition. **b** The ΔT as a function of the ambient temperature while the net radiative cooling power is zero. **c** Diagram of the calculated Pnet under different radiation temperatures and ambient temperatures. **d** Diagram of the calculated Pnet as a function of ΔT with different conductive-convective heat transfer coefficients under the circumstance of the ambient temperature of 30 °C.



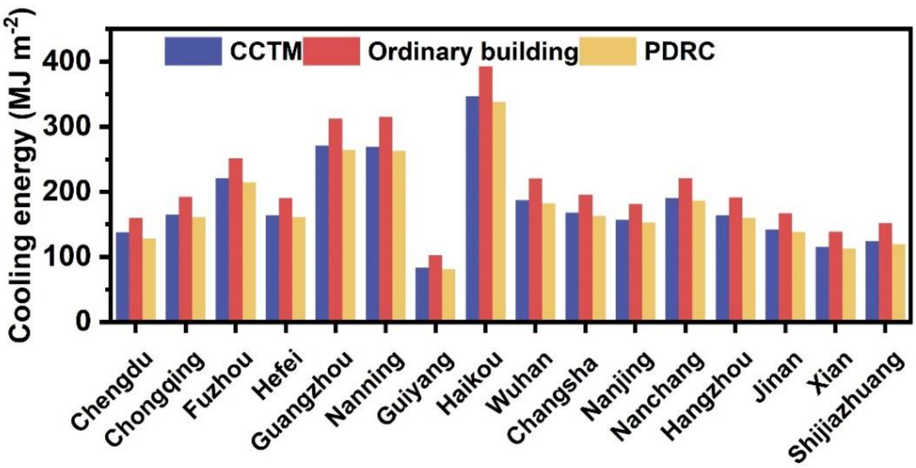
Supplementary Fig. 12. The temperatures of the three samples during the first half hour of the test on 28, October, 2024.



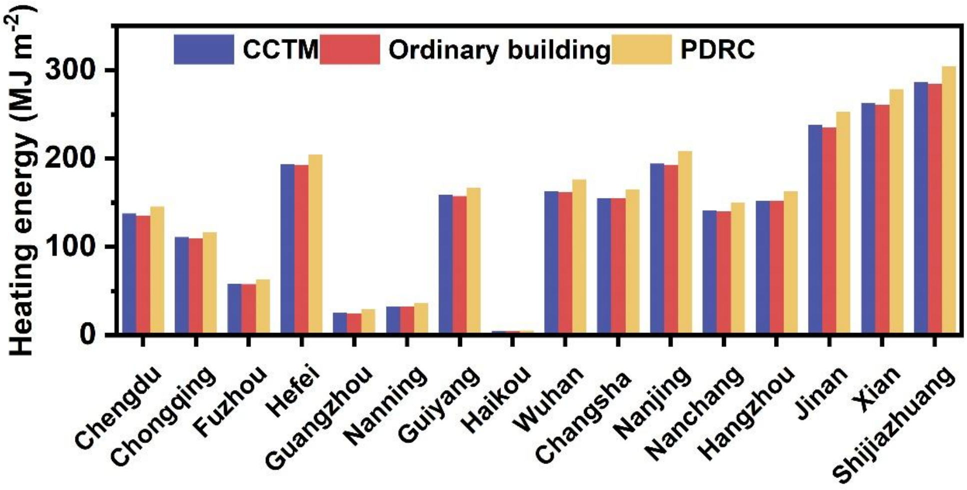
Supplementary Fig. 13. The temperatures of the three samples during the hottest time of the test on 21, November, 2024.



Supplementary Fig. 14. Photographs of different substrates coating with CCTM in different colors.



Supplementary Fig. 15. Simulations of annual cooling energy of different Chinese cities for ordinary buildings with and without PDRC and CCTM coatings.



Supplementary Fig. 16. Simulations of annual heating energy of different Chinese cities for ordinary buildings with and without PDRC and CCTM coatings.