Supporting Information
Janus film for passive radiative cooling and solar heating for passive thermal regulation

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Supporting notes

Spectra calcualtion of the Janus film

The overall hemispherical solar reflectance, $\bar{R}$, is functions of wavelengths and incidents angles, which is expressed as:

$$\bar{R}(\lambda,\theta,\phi)=\frac{\int_{0.3\mu m}^{2.5\mu m} I_{solar}(\lambda,\theta,\phi)R(\lambda,\theta,\phi)d\lambda}{\int_{0.3\mu m}^{2.5\mu m} I_{solar}(\lambda,\theta,\phi)d\lambda} \tag{1}$$

where λ is the wavelength of the solar radiation, ϕ is the azimuthal angle, and θ is the polar angle. $R(\lambda,\theta,\phi)$ is the spectral directional reflectance.

The overall hemispherical thermal emittance, $\bar{\epsilon}$, is also a function of the wavelengths and incident angles, which is defined by:

$$\bar{\epsilon}(\lambda,\theta,\phi)=\frac{\int_{2.5\mu m}^{25\mu m} I_{bb}(\lambda,\theta,\phi)[1-R(\lambda,\theta,\phi)]d\lambda}{\int_{2.5\mu m}^{25\mu m} I_{bb}(\lambda,\theta,\phi)d\lambda} \tag{2}$$

where $I_{bb}(\lambda,\theta,\phi)$ means the blackbody radiation intensity described by Planck's law. $\bar{\epsilon}(\lambda,\theta,\phi)$ is the spectral directional absorptance at a certain operating temperature.

Optical response of nanoparticles embedded into thin film

Clausius-Mossotti equation is used to express the effective dielectric function when Al nanoparticles are embedded in polyethylene.

$$\epsilon_{eff}=\epsilon_m\left(\frac{R^3+2\alpha_R VF}{R^3-\alpha_R VF}\right) \tag{3}$$

where ϵ_m is the matrix's dielectric function, α_R is the electric dipole polarizability, and R and VF are the nanoparticles' radius and volume fraction, respectively. Mie theory is an effective way to take doping nanoparticle size into account. Electric dipole polarizability is expressed using the Maxwell-Garnett formula and the Mie theory,[1]

$$\alpha_R=\frac{3jc^3}{2\omega^3\epsilon_m^{3/2}}a_{1,R} \tag{4}$$

where $a_{1,R}$ is the first electric Mie coefficient given by

$$a_{1,R}=\frac{\sqrt{\epsilon_{np}}\psi_1(x_{np})\psi_1'(x_m)-\sqrt{\epsilon_m}\psi_1(x_m)\psi_1'(x_{np})}{\sqrt{\epsilon_{np}}\psi_1(x_{np})\xi_1'(x_m)-\sqrt{\epsilon_m}\xi_1(x_m)\psi_1'(x_{np})} \tag{5}$$

where ψ_1 and ξ_1 are the first-order Riccati-Bessel functions expressed by $\psi_1(x)=xj_1(x)$ and $\xi_1(x)=xh_1^{(1)}(x)$ where j_1 and $h_1^{(1)}$ are first-order spherical Bessel functions and spherical first-kind Hankel functions, respectively. Here, '′' indicates the first derivative. $x_m=\omega r\sqrt{\epsilon_m}/c$ and $x_{np}=\omega r\sqrt{\epsilon_{np}}/c$ are the size parameters of the matrix and the nanoparticles, respectively; ϵ_{np} is the dielectric function of nanoparticles.

Energy balance analysis of the Janus metamaterials

In order to analyze the temperature response of the proposed structures, the thermal balance equation is listed as below:[2,3]

$$\begin{aligned} Q_{\text{total}}(T_{\text{Janus}},T_{\text{amb}}) &= Q_{\text{sun}}(T_{\text{Janus}})+Q_{\text{amb}}(T_{\text{amb}}) \\ &-Q_{\text{re-emit}}(T_{\text{Janus}})-Q_{\text{non-radia}}(T_{\text{Janus}},T_{\text{amb}}) \end{aligned} \tag{6}$$

In this situation, the back of the structure is thermally insulated, which means that our proposed structures are not subjected to any thermal load. Different thermal loads can cause varying temperature responses. Energy transfer takes place among the Sun, outer space, the Janus, and the ambient environment. In this context, we use the terms Q_{sun} to represent the solar heat gain, Q_{amb} to describe thermal radiation from the atmosphere, $Q_{re-emit}$ to indicate the heat flux reemitted by the Janus, and Q_{total} to denote the net flux of the Janus. Furthermore, the thermal conduction and convection heat transfer between the structure and the ambient is referred as to $Q_{non-radia}$.

The solar heating flux, $Q_{sun}(T_{Janus})$, is defined by:

$$Q_{\text{sun}}\left(T_{\text{Janus}}\right) = A \cdot CF \int_{0}^{\infty} \mathrm{d}\lambda I_{\mathrm{AM1.5}}(\lambda) \alpha\left(\lambda, \theta_{\text{sun}}, T_{\text{Janus}}\right) \tag{7}$$

In this context, A represents the area of the Janus. The term CF denotes the concentration factors. $\alpha\left(\lambda, \theta_{\text{sun}}, T_{\text{Janus}}\right)$ refers to the solar absorptance and it changes with temperature, wavelength, and angle of incidence. However, as shown in subsequent sections, the solar absorptance is independent of temperature and angle of incidence. Hence, we can calculate the solar absorptance when an incident angle is zero.

The power of incident thermal radiation from the atmosphere, $Q_{sun}(T_{Janus})$ is listed as follows:

$$\begin{aligned} Q_{\text{amb}}\left(T_{\text{amb}}\right) = A \int_{0}^{\infty} \mathrm{d}\lambda I_{\mathrm{BB}}\left(T_{\text{amb}}, \lambda\right) \alpha\left(\lambda, \theta, \phi, T_{\text{Janus}}\right) \\ \epsilon(\lambda, \theta, \phi) \end{aligned} \tag{8}$$

$I_{\mathrm{BB}}\left(T_{\text{amb}}, \lambda\right) = 2hc^5\lambda^{-5}\exp\left(hc/\lambda k_B T_{amb} - 1\right)^{-1}$ represents the thermal infrared radiation of a blackbody, where h denotes Planck's constant and k_B denotes the Boltzmann constant. $\alpha\left(\lambda, \theta, \phi, T_{\text{Janus}}\right) = \frac{1}{\pi}\int_0^{2\pi} \mathrm{d}\phi \int_0^{\pi/2} \epsilon_\lambda \cos\theta \sin\theta \mathrm{d}\theta$ denotes the temperature-dependent solar absorptance,[4] which is assumed to be temperature-independent based on the thermal stability test. The air emittance, $\epsilon(\lambda, \theta, \phi)$, is defined by $1 - t(\lambda, \theta, \phi).t(\lambda, \theta, \phi)$, where $t(\lambda, \theta, \phi)$ defines the transmittance of the atmosphere that is extracted from MODTRAN 4.[5]

The heat flux reemitted by the Janus can be calculated as follows:

$$\begin{aligned} Q_{\text{re-emit}}\left(T_{\text{Janus}}\right) = A \int_{0}^{\infty} \mathrm{d}\lambda I_{\mathrm{BB}}\left(T_{\text{Janus}}, \lambda\right) \\ \epsilon\left(\lambda, \theta, \phi, T_{\text{Janus}}\right) \end{aligned} \tag{9}$$

As described by the Kirchhoff's law of thermal radiation,[6] $\epsilon\left(\lambda, \theta, \phi, T_{\text{Janus}}\right)$ is equal to $\alpha\left(\lambda, \theta, \phi, T_{\text{Janus}}\right)$, which represents the emittance of the Janus.

$$Q_{\text{non-radia}}\left(T_{\text{Janus}}, T_{\text{amb}}\right) = h\left(T_{\text{Janus}} - T_{\text{amb}}\right) \tag{10}$$

h represents the thermal conduction and convection heat transfer coefficient. Here, h = 8 W m^{-2} K^{-1} is taken to calculate the convection heat transfer coefficient of natural air.

The change in temperature can be calculated using the following equation:

$$C_{\text{Janus}} \frac{dT}{dt} = Q_{\text{total}}\left(T_{\text{Janus}}, T_{\text{amb}}\right) \tag{11}$$

The Janus has a thickness of approximately 40 μm. For the purpose of calculating the temperature response, the heat capacitance of the Janus, denoted as C_{Janus}, is assumed to be equivalent to that of a 30 μm thick PDMS film and 10 μm thick PE film, since the thickness of the Ag and PE nanograting layer is negligible in comparison and is thus ignored. The substrate thickness primarily influences the thermal load, which in turn affects the time required to reach thermal equilibrium.

References

[1] Doyle, W. T. Optical properties of a suspension of metal spheres. *Physical review B* **39**, 9852 (1989).

[2] Liu, Y. *et al.* Intelligent regulation of vo2-pdms-driven radiative cooling. *Applied Physics Letters* **120**, 171704 (2022).

[3] Yang, P., Chen, C. & Zhang, Z. M. A dual-layer structure with record-high solar reflectance for daytime radiative cooling. *Solar Energy* **169**, 316–324 (2018).

[4] Zhang, Z. M. *Nano/microscale heat transfer* (Springer, 2007).

[5] Berk, A. *et al.* Modtran4 radiative transfer modeling for atmospheric correction. In *Optical spectroscopic techniques and instrumentation for atmospheric and space research III*, vol. 3756, 348–354 (International Society for Optics and Photonics, 1999).

[6] Robitaille, P.-M. Kirchhoff's law of thermal emission: 150 years. *Progr. Phys* **4**, 3–13 (2009).