Colored and paintable bilayer coatings with high solar-infrared reflectance for efficient cooling

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Solar reflective and thermally emissive surfaces offer a sustainable way to cool objects under sunlight. However, white or silvery reflectance of these surfaces does not satisfy the need for color. Here, we present a paintable bilayer coating that simultaneously achieves color and radiative cooling. The bilayer comprises a thin, visible-absorptive layer atop a nonabsorptive, solar-scattering underlayer. The top layer absorbs appropriate visible wavelengths to show specific colors, while the underlayer maximizes the reflection of near-to-short wavelength infrared (NSWIR) light to reduce solar heating. Consequently, the bilayer attains higher NSWIR reflectance (by 0.1 to 0.51) compared with commercial paint monolayers of the same color and stays cooler by as much as 3.0° to 15.6°C under strong sunlight. High NSWIR reflectance of 0.89 is realized in the blue bilayer. The performances show that the bilayer paint design can achieve both color and efficient radiative cooling in a simple, inexpensive, and scalable manner.

INTRODUCTION

Cooling terrestrial objects, such as buildings, vehicles, and data centers, is a critical challenge that we face today. However, cooling is often energy intensive, as predominantly used compression-based coolers consume a substantial amount of electricity. For example, cooling indoor spaces contributes to ~15% of total household electricity usage in the United States (1). Moreover, these cooling designs have a heating effect and often require ozone-depleting or greenhouse gas-releasing coolants. Hence, alternative approaches with low energy consumption and a net cooling effect are desirable (2, 3).

One such promising alternative is radiative cooling using surfaces with high solar reflectance (Rsol) and high emittance (ε) including in the long-wavelength infrared (LWIR) atmospheric transmission window. The high Rsol minimizes solar heating, while the high ε enables radiative heat loss to the cold outer space,allowing the object to stay cool even under sunlight. Because of its passive and eco-friendly operation and its net cooling effect (4–7), radiative cooling designs have been widely investigated. Examples of these designs include white paints (8–10), porous (11) or metatized (12–14) polymers, polymer-dielectric composites (14–18), photonic architectures (19–22), and natural materials (23–25). Usually, these designs maximize radiative cooling by using metal mirrors or white materials with high Rsol. However, their broadband reflectance in visible wavelengths restricts their use in real-life situations. For instance, white colors are often not desirable as coatings on buildings or other objects for aesthetic or functional reasons (26–28). Furthermore, the white or silvery glare from these designs can harm human eyes. Colored radiative coolers (CRCs) have been explored to address this issue (29–34). In a CRC, part(s) of the visible spectrum (VIS; 0.4 to 0.74 μm) is selectively absorbed to exhibit the desired color, while other solar wavelengths, in particular, the near-to-short wavelength infrared (NSWIR; 0.74 to 2.5 μm), are reflected (Fig. 1A). Since NSWIR wavelengths carry 51% of total solar energy, a high NSWIR reflectance (RNSWIR) considerably reduces solar heating. In addition, the cooler also has a high, broadband ε to effectively radiate heat to the cold sky. However, existing CRCs are limited in either their performance or their scope. For instance, multilayer photonic CRCs (29, 30) have a high cooling performance but currently are rather expensive and difficult to apply on buildings or cars, which have various shapes, sizes, and textures (35, 36). Colored paints containing TiO2 and colorants (31, 34), on the other hand, are scalable but usually absorb NSWIR wavelengths to become hot under sunlight. Therefore, simultaneously achieving color and a cooling performance in a highly scalable manner remains a challenge.

Here, we report a bilayer CRC paint coating (Fig. 1, B and C) that consists of a top layer containing a colorant, and an underlayer made of porous poly(vinylidene fluoride-co-hexafluoropropene) [P(VdF-HFP)] or TiO2/polymer composite paint. The top layer selectively absorbs visible wavelengths complementary to the desired color but not others, while the underlayer maximizes the backscattering of any sunlight transmitted by the top layer (Fig. 1B). Consequently, the bilayers exhibit near-identical colors and visible reflectances (RVIS) to those of commercial monolayer paint, but a considerably higher RNSWIR, as illustrated in infrared images in Fig. 1D. The increased RNSWIR translates to a higher cooling performance and substantially lower temperatures for the bilayers under sunlight (fig. S1). The performances, especially those of porous P(VdF-HFP)–based bilayers, are considerably better than the control monolayer coatings made using commercial paints. The results are among the best reported so far (29, 30) but were achieved with the simplicity and scalability of paints. For instance, a black porous P(VdF-HFP)–based bilayer that we designed has a nearly identical RVIS (0.07) to that of a monolayer black coating (0.05), but a much higher RNSWIR (0.81 compared with 0.30). Consequently, it can attain a 15.7°C cooler temperature under a mid-day summertime solar intensity of ~1025 W m−2. Note that the cooling achieved by the bilayer designs is not relative to the ambient air as targeted by white or silvered radiative coolers but to commercial paint coatings of the same color.
RESULTS
Principle of the bilayer design

To simultaneously achieve color and cooling performance, a surface should maximize its $R_{\text{NSWIR}}$ to minimize solar heating and have a high, broadband thermal emittance $\varepsilon$ required for efficient heat loss to the sky. Commercial colored paint coatings already fulfill the emittance requirement—since solar absorption of colored coatings usually results in above-ambient temperatures, their high broadband emittance ($\varepsilon \approx 0.9$) (fig. S2) is better suited for cooling than selective LWIR emittance ($\varepsilon \approx 0.85$). However, their solar reflectance remains less than ideal. In existing monolayer paint designed for cooling, pigments or dyes are used to absorb visible light complementary to the target color, and TiO$_2$ particles scatter and reflect other visible and NSWIR light because of the difference in refractive indices between TiO$_2$ and the paint matrix (e.g., polymers). Since NSWIR light, with its longer wavelengths, has a considerably larger penetration depth than visible light (fig. S3), the paint needs to be thick to effectively reflect NSWIR, much thicker than that required for reflecting visible light. However, this leads to an issue that increases the absorption of NSWIR light as follows. Since colorants (dyes or pigments) typically have at least a trailing NSWIR absorptivity (Fig. 2C), the scattering of NSWIR light by the colorant is weak (if the colorants are nanoparticle pigments $\leq 50$ nm) or negligible (if they are dissolved dyes), light in the NSWIR wavelengths are transmitted, along short optical paths and without substantial absorption, into the underlayer. Once in the solar-scattering and nonabsorptive underlayer, the NSWIR light is strongly backscattered into the top layer, through which they pass, mostly unimpeded, back into free space, resulting in a high $R_{\text{NSWIR}}$. Furthermore, the thin top layer has the same concentration of colorant and composition as the conventional monolayer with the target color. Although much thinner than the monolayer coating, it still has enough thickness to ensure a strong absorption of visible wavelengths complementary to the target color, while other wavelengths are reflected by either itself or the solar-scattering underlayer. This ensures that the bilayer appears almost identically colored as the monolayer while attaining a higher solar-infrared reflectance.

The bilayer design in Fig. 2B, with a thin colored layer atop a thick solar-scattering and nonabsorptive underlayer, can address this problem. Since the top layer is thin (Fig. 2D) and scattering of NSWIR light by the colorant is weak (if the colorants are nanoparticle pigments $\leq 50$ nm) or negligible (if they are dissolved dyes), light in the NSWIR wavelengths are transmitted, along short optical paths and without substantial absorption, into the underlayer. Once in the solar-scattering and nonabsorptive underlayer, the NSWIR light is strongly backscattered into the top layer, through which they pass, mostly unimpeded, back into free space, resulting in a high $R_{\text{NSWIR}}$. Furthermore, the thin top layer has the same concentration of colorant and composition as the conventional monolayer with the target color. Although much thinner than the monolayer coating, it still has enough thickness to ensure a strong absorption of visible wavelengths complementary to the target color, while other wavelengths are reflected by either itself or the solar-scattering underlayer. This ensures that the bilayer appears almost identically colored as the monolayer while attaining a higher solar-infrared reflectance.

The bilayer concept is validated by FDTD reflectance simulations of bilayer (5-μm colored top layer +145-μm porous polymer underlayer) and monolayer (150-μm colored porous layer) coatings (Fig. 2D). The colored layer contains a selective black dye in polymer. The complex refractive index of the dyed polymer is shown in Fig. 2C. As simulated, the bilayer design achieves a near-identical $R_{\text{VIS}}$ and, thus,
followed by that of the TiO2-based bilayers. Specifically, are substantially higher for the porous P(VdF-HFP)–based bilayers, (Fig. 3B, fig. S5, and table S1). In the NSWIR, however, the reflectances ilar CIE x values of RNSWIR are closely matched, leading to sim-

Comparison with commercial paint monolayers
In this report, we compare the bilayer design concept with monolay-
er commercial paints of the same colors, which were both fabricated by a simple painting method (fig. S4). Specifically, we investigate two types of solar-scattering underlayers. One is 500-μm-thick layers of ~50% porous P(VdF-HFP), which contains interconnected micro-

When compared with commercial monolayer paints, the two bi-

Fig. 2. Principle of the bilayer design. (A and B) Schematic showing the interaction of sunlight with monolayer (A) and bilayer (B) coatings, respectively, and how the latter achieves a higher reflectance. (C) Complex spectral refractive index (n+iκ) for a polymer containing a selective black dye, showing strong absorption in the visible wavelengths and trailing absorption in the NSWIR wavelengths. (D) Schematic showing the three simulated setups: (left) a solar-scattering porous polymer, (middle) a monolayer of dyed porous polymer with the refractive index in (C), and (right) a bilayer containing the scattering medium at the bottom and a thin film of the monolayer at the top. (E) Simulated reflectances of the three structures in (D).

color as the monolayer. However, its RNSWIR (0.48) is considerably higher than the monolayer’s (0.29), leading to a large reduction in solar heating (Fig. 2E). The overall effect is enhanced cooling and same color as the monolayer, with lowered colorant usage.

In addition to the high RNSWIR, the bilayer designs also have high thermal emittances (Table 1 and fig. S10) due to the intrinsically emissive nature of the polymers in the top layer and underlayer. The emittance of the underlayers, colored top layers, and bilayers are pres-

The enhancements in RNSWIR achieved through the bilayer approach lead to better daytime cooling performances. We demonstrate this by exposing the bilayer and monolayer samples in Fig. 3 (C to F) to direct sunlight (Fig. 4, A and B). To test the cooling performance in a controlled and consistent environment with similar air convection coefficients, we used solar and infrared transparent 25-μm-thick
poly(ethylene) (PE) films to shield the setup, as has been done in previous studies (20, 25). For the extreme case (the black samples), because of the large contrast in $R_{\text{NSWIR}}$ [0.81 for porous P(VdF-HFP) bilayer, 0.73 for TiO$_2$ bilayer, and 0.30 for monolayer; Table 1], the porous P(VdF-HFP)– and TiO$_2$-based bilayers remain 15.6°C and 13.2°C cooler than the monolayer under $\sim$1025 W m$^{-2}$ solar irradiation (Fig. 4C). For blue/red/yellow colors, on the other hand, the porous P(VdF-HFP)– and TiO$_2$-based bilayers remain 6.6°C/3.0°C/7.3°C and 4.3°C/1.8°C/5.2°C cooler than the monolayer commercial paints (Fig. 4, D to F), respectively. These large temperature differences are consistent with theoretical simulations (fig. S12), assuming a convective heat transfer coefficient ($h_c$) of $\sim$5 to 7 W m$^{-2}$ K$^{-1}$ observed in the literature (20, 36).

We also conducted tests without PE convection shields. Even with larger convective effects, the black/blue/red/yellow bilayers were still 6.4°C/3.4°C/2.6°C/4.5°C [porous P(VdF-HFP)] and 4.6°C/1.9°C/1.5°C/3.8°C (TiO$_2$) cooler than the monolayer coatings (fig. S13), indicating their performance under breezy conditions. In certain situations, where the solar intensities are sufficiently low, light-yellow bilayers with high solar reflectances achieved subambient cooling (fig. S14). These results demonstrate that the bilayer design, especially based on porous P(VdF-HFP), is attractive for reducing temperatures and air-conditioning costs in buildings, cars, and other terrestrial objects. From a practical perspective, the performances are achieved with a simple painting process while satisfying the aesthetic requirement for color.

**DISCUSSION**

**Pushing the bilayer performance**

While the above demonstration shows the superior optical and thermal performance of the bilayers over monolayers, commercial paints are not necessarily ideal as top layers, as they usually contain...
colorants that are NSWIR absorptive and, often, pigments that scatter and absorb NSWIR wavelengths. The ideal top layer should exhibit highly selective visible absorption where required and minimal scattering of NSWIR wavelengths. The highly selective visible absorption has been widely studied, including dielectric film–coated metal flakes as pigments by Smith et al. (32, 33) and a comprehensive investigation of pigments by Levinson et al. (38, 39); minimal NSWIR scattering can be achieved by using small pigments (with sizes <100 nm) or organic dyes dissolved in polymers. Figure 5A shows an illustration of this concept where the colorant is a highly selective blue dye (Sudan Blue II) dispersed in solid P(VdF-HFP) and the underlayer is a porous P(VdF-HFP) film, which has a high $R_{\text{NSWIR}}$ of 0.89. This performance is also much higher than the bilayer with commercial blue top layer ($R_{\text{NSWIR}}$ of 0.63). If $R_{\text{VIS}}$ are the same in the two coatings, the large $R_{\text{NSWIR}}$ could reduce noon-time ($I_{\text{Solar}} = 1000 \text{ W m}^{-2}$) surface temperatures by 12°C under still air ($h_c = 5 \text{ W m}^{-2} \text{ K}^{-1}$) or 6°C under mild winds ($h_c = 15 \text{ W m}^{-2} \text{ K}^{-1}$) (fig. S1). The results indicate that optimizing the colored top layer can further enhance the cooling performance of the bilayer design.

Besides achieving different hues through the choice of colorant, the bilayer design can also achieve different lightness or shades for the same hue. As demonstrated in Fig. 5B, for bilayers with Sudan Blue colorant, a thin top layer can yield a whitish blue color, while thicker top layers show a deeper blue. By mixing with Perylene Black, darker blue-black shades can be achieved. $R_{\text{NSWIR}}$, however, remains appealingly high regardless of the shade (0.86, 0.80, and 0.73 for three curves in Fig. 5B). In applications where the reduction in glare while maintaining hue is important, this can be highly useful. The concept of the bilayer is one that can be generalized to any dye, pigment, polymer, or other paint components to achieve specific attributes. For instance, the pigments in the top layer and the polymer in the underlayer can be chosen to achieve high stability. We demonstrated this by placing porous P(VdF-HFP)–based bilayers outdoors or in an oven at 60°C for 30 days, during which the reflectance and emittance of the coatings showed no appreciable changes (~0.01 or less; fig. S15).

A last point, in relation to the paintability of the bilayer design, is the sensitivity of its optical performance to fabrication parameters. To test for this, we conducted a sensitivity analysis, investigating factors such as replicability of the optical performances over repeated fabrications following the same procedure, changing the composition of the precursor for the P(VdF-HFP) underlayer, and testing the effect of altering the thickness of the top layer. Our results, presented in figs. S16 to S18, show that the optical performances of the coatings are highly replicable for a given fabrication procedure and that the use of different compositions of precursors to fabricate the P(VdF-HFP) underlayer and varying the thickness of the top layer have minimal impact on $R_{\text{NSWIR}}$ and thermal emittance. All the three tests indicate...
the robustness of the fabrication procedure at yielding high cooling performance.

In conclusion, we propose a simple, inexpensive, and scalable method to make a paintable bilayer colored radiative cooling coating, which comprises a top layer to absorb complementary visible wavelengths to the desired colors, and an underlayer to strongly reflect sunlight in NSWIR wavelengths. Theoretical simulations and optical characterizations reveal that the bilayer design can reduce solar-infrared absorption by the colorants and thereby achieve a higher \(R_{\text{NSWIR}}\) than its monolayer counterpart. When compared with conventional colored monolayer paint with the same color, the \(R_{\text{NSWIR}}\) is improved by up to 0.51 by using porous P(VdF-HFP) as a solar-scattering underlayer. As a result, high cooling performances are achieved, e.g., for the black porous P(VdF-HFP)-based bilayer, a 15.6°C lower temperature than that of the monolayer black coating was attained under a solar intensity ~1025 W m\(^{-2}\). By changing the dye components and their amounts, we can further increase \(R_{\text{NSWIR}}\) and change color shades of the bilayer paints. The paintable bilayer designs, which substantially outperform commercial monolayer paints, demonstrate a practical and efficient solution to cooling colored objects in a green and energy-saving manner.

**MATERIALS AND METHODS**

**Fabrication of colored cooling paint coatings**

The bilayer colored cooler was fabricated via a two-step process (fig. S4): Porous P(VdF-HFP)– or TiO\(_2\)-based white underlayer was first painted on a substrate, and then, a layer containing a colorant was painted on top. The porous P(VdF-HFP) underlayer was created using a previously outlined phase inversion method (11): A solution of P(VdF-HFP) powder (Kynar Flex 2801)/acetone/water with a weight ratio of 1:8:1 was painted on a plastic substrate to form a white layer with a thickness of ~500 \(\mu\)m. The TiO\(_2\)-based commercial white paint (Sherwin-Williams, 636613 High Reflective White) was painted onto a substrate to form a ~250-\(\mu\)m-thick layer. To make the blue, red, and yellow top layers shown in Figs. 1 to 4, commercial paints (2066-30 Big Country Blue, 2086-30 Rosy Blush, and 2021-30 Sunshine from Benjamin Moore, respectively) were used. For black top layers, Perylene Black (Oakwood Chemical) as the colorant (1 mg ml\(^{-1}\)) and P(VdF-HFP) as the polymer matrix (150 mg ml\(^{-1}\)) are dispersed in acetone to make the paint. For black top layers, Perylene Black (Oakwood Chemical) as the colorant (1 mg ml\(^{-1}\)) and P(VdF-HFP) as the polymer matrix (150 mg ml\(^{-1}\)) are dispersed in acetone to make the paint. The blue top layers shown in Fig. 5 were made in the same way as black, except that Sudan Blue II dye (Sigma-Aldrich) was used as the colorant. The monolayer samples were obtained by coating the same colored paints in the bilayer designs onto poly(methyl methacrylate) (PMMA) substrates, with thicknesses equal to those of the corresponding TiO\(_2\)-based bilayers.

**Optical characterization**

The visible and NSWIR photographs of the samples were taken using Nikon D3300 camera and NIRvana ST 640 camera, respectively. Microscopy was performed using the Zeiss Axio Imager.A2m optical microscope and Zeiss Sigma VP scanning electron microscope. The reflectance spectra were taken separately in two wavelength ranges: visible to near-infrared (0.4 to 1.0 \(\mu\)m) and near-infrared to LWIR.
(1.0 to 15 μm) for incident angles of 30°. In the first range, the reflectance was measured using an integrating sphere (model IS200, Thorlabs) containing a silicon detector and coupled to a high-power supercontinuum laser (SuperK EXTREME, NKT Photonics) and a tunable filter (Fianium LLTF contrast). The sample was put inside the integrating sphere. A calibrated diffuse reflector (item SM05CP2C, Thorlabs) was used as the reference. In the second range, reflectance was measured using a gold integrating sphere (model 4P-GPS-020-SL, Labsphere) coupled with a mercury cadmium telluride detector and a Fourier transform infrared spectrometer (VERTEX 70v, Bruker). A gold-coated aluminum foil was used as the reference. The spectra in the two ranges were then patched to obtain the final reflectance. The transmission spectra were obtained in the same way, except that the sample was placed at the mouth of the integrating sphere.

Outdoor temperature measurements

The thermal tests were conducted using the setup (Fig. 4B) in New York on 23, 26, 27, and 28 June 2019 for blue, black, yellow, and red samples, respectively. For each color, porous P(VdF-HFP)-based bilayer, TiO2-based bilayer, and monolayer samples with an area of 7.5 cm by 7.5 cm were placed in a transparent open-top polycarbonate box. A low-density PE film was tautly drawn above the samples as a wind shield to reduce the convective heat transfer without substantially hindering solar and thermal infrared transmission. All samples were supported by styrofoam, and the box itself was placed on another large white styrofoam to reduce the heat transfer between the samples and ground. The temperature of each sample was measured by a thermocouple pressed to its back face by a black tape, which also served as a solar absorptive layer. A thermocouple shielded from sunlight was used to measure air temperature in the box. A pyranometer (Apogee, SP-510) connected to the computer was placed beside the sample to measure the total (direct + diffuse) solar intensity.

Reflectance and emittance calculation

The reflectance \( R \) is defined as the ratio of the reflected solar intensity within a certain wavelength range (\( \lambda_1 \) to \( \lambda_2 \)) to the total incident solar intensity in the same range, as expressed below

\[
R = \frac{\int_{\lambda_1}^{\lambda_2} I_{\text{sol}}(\lambda) R(\lambda) \, d\lambda}{\int_{\lambda_1}^{\lambda_2} I_{\text{sol}}(\lambda) \, d\lambda}
\]

(1)

where \( I_{\text{sol}}(\lambda) \) is the ASTM G173-03 global solar intensity spectrum, and \( R(\lambda) \) is the sample’s spectral reflectance. Ranges 0.4 to 0.74 μm and 0.74 to 2.5 μm correspond to the wavelength range used in the calculation of reflectance in the visible and NSWIR wavelengths, respectively.

Similarly, the thermal emittance \( \bar{\epsilon} \) is defined as the ratio of the spectral intensity within a certain wavelength area (\( \lambda_1 \) to \( \lambda_2 \)) to that of a standard blackbody at the same temperature and wavelength same area, as expressed below

\[
\bar{\epsilon} = \frac{\int_{\lambda_1}^{\lambda_2} I_{\text{bb}}(T, \lambda) \epsilon(T, \lambda) \, d\lambda}{\int_{\lambda_1}^{\lambda_2} I_{\text{bb}}(T, \lambda) \, d\lambda}
\]

(2)

where \( I_{\text{bb}}(T, \lambda) \) is the spectral intensity emitted by a standard blackbody with a temperature of \( T \), and \( \epsilon(T, \lambda) \) is the sample’s spectral emittance.

Calculating color from the spectrum

Tristimulus values \( X, Y, \) and \( Z \) are calculated to measure the response of human eyes to the light based on the CIE color-matching functions (40) \( \bar{\epsilon}(\lambda), \bar{\eta}(\lambda), \) and \( \bar{\varepsilon}(\lambda) \) and the sample’s reflectance spectrum \( R(\lambda) \) using the formulas below

\[
X = 100 \frac{\int \bar{\epsilon}(\lambda) R(\lambda) \bar{\eta}(\lambda) \, d\lambda}{\int \bar{\eta}(\lambda) \, d\lambda}
\]

(3)

\[
Y = 100 \frac{\int \bar{\epsilon}(\lambda) R(\lambda) \bar{\varepsilon}(\lambda) \, d\lambda}{\int \bar{\varepsilon}(\lambda) \, d\lambda}
\]

(4)

\[
Z = 100 \frac{\int \bar{\epsilon}(\lambda) R(\lambda) \bar{\eta}(\lambda) \, d\lambda}{\int \bar{\eta}(\lambda) \, d\lambda}
\]

(5)

Here, CIE Illuminant D65 spectrum \( I(\lambda) \) is used to portray the standard open-air illumination conditions. The chromaticity is then specified by the two normalized values (\( x \) and \( y \)) derived from the tristimulus values and located in the CIE 1931 color space

\[
x = \frac{X}{X+Y+Z}
\]

(6)

\[
y = \frac{Y}{X+Y+Z}
\]

(7)

The lightness (\( L \)) of the color is calculated by the Lab-XYZ color space conversion (40)

\[
L=116f\left(\frac{Y}{Y_n}\right) - 16
\]

(8)

where \( Y_n \) = 100, corresponding to the CIE XYZ tristimulus value of the reference white point under Illuminant D65 and

\[
f(t)=\begin{cases} 
0 & \text{if } t \leq \left(\frac{6}{29}\right)^3 \\
\frac{t}{3 \left(\frac{6}{29}\right)^3 + \frac{4}{29}} & \text{if } t > \left(\frac{6}{29}\right)^3 
\end{cases}
\]

(9)

FDTD simulation

FDTD simulations of the structures shown in Fig. 2D were carried out using FDTD Solutions 8.6.1 software by Lumerical. For the scattering medium, a polymer (\( n \sim 1.39 \)) (11) with light scattering air voids (\( n \sim 1 \)) of sizes 0.1 to 0.5 μm was used. The thickness was chosen to be 150 μm. For the dyed monolayer, a 150-μm-thick, optically homogenous, nonscattering polymer-dye mixture with the complex refractive index shown in Fig. 2C, and containing light scattering air voids with the same size distribution as the scattering medium, was used instead. The refractive index was generated using a simple Lorentz oscillator model. The bilayer consisted of 5 μm of the dyed monolayer placed atop of a 145-μm solar scattering layer. A plane wave light source was used, and the spectrally resolved backscattered power and, thus, reflectance were measured by a monitor placed above the aforementioned structures. As shown in Fig. 2E, the simulated NSWIR reflectance for the bilayer is indeed higher than that of the monolayer and validates our bilayer approach.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/17/eaaz5413/DC1
REFERENCES AND NOTES

1. U.S. Energy Information Administration, Annual energy outlook 2019 with projections to 2050; www.eia.gov/outlooks/aeo/pdf/aeo2019.pdf.
2. G. B. Smith, C.-G. Granqvist, Green Nanotechnology—Solutions for Sustainability and Energy in the Built Environment (CRC Press, 2010).
3. M. Santamouris, J. Feng, Recent progress in daytime radiative cooling: Is it the air conditioner of the future? Buildings 8, 168 (2018).
4. C. G. Granqvist, A. Hjortsberg, Surfaces for radiative cooling: Silicon monoxide films on aluminum. Appl. Phys. Lett. 36, 139–141 (1980).
5. C. G. Granqvist, G. A. Nilsson, Solar energy materials for thermal applications: A primer. Sol. Energy Mater. Sol. Cells 180, 213–226 (2018).
6. M. M. Hossain, M. Gu, Radiative cooling: Principles, progress, and potentials. Adv. Sci. 3, 1500360 (2016).
7. B. Zhao, M. Hu, X. Ao, N. Chen, G. Pei, Radiative Cooling: A review of fundamentals, materials, applications, and prospects. Appl. Energy 236, 489–513 (2019).
8. C. S. Wojtysiak, Radiative cooling surface coatings. Patent WD 2020098996 (2002).
9. B. Bhata, A. Leroy, Y. Shen, L. Zhao, M. Gianello, D. Li, T. Gu, J. Hu, M. Soljačić, E. N. Wang, Passive directional sub-ambient daytime radiative cooling. Nat. Commun. 9, 5001 (2018).
10. Cool Roof Rating Council, Rated products directory; https://coolroofs.org/directory.
11. J. Mandal, Y. Fu, A. C. Overvig, M. Jia, K. Sun, N. N. Shi, H. Zhou, X. Xiao, N. Yu, Y. Yang, Hierarchically porous polymer coatings for highly efficient passive daytime radiative cooling. Science 362, 315–318 (2018).
12. A. R. Gentle, G. B. Smith, A subambient open roof surface under the mid-summer sun. Adv. Sci. 2, 1500119 (2015).
13. J.-L. Kou, Z. Jurado, Z. Chen, S. H. Fan, A. J. Minhch, Daytime radiative cooling using near-black infrared emitters. ACS Photonics 4, 626–630 (2017).
14. N. Yu, J. Mandal, A. Overvig, N. N. Shi, Systems and methods for radiative cooling and heating. Patent WD 2016205717 (2016).
15. A. R. Gentle, G. B. Smith, Radiative heat pumping from the earth using surface phonon resonant nanoparticles. Nano Lett. 10, 373–379 (2010).
16. Y. Zhai, Y. Ma, S. N. David, D. Zhao, R. Lou, G. Tan, R. Yang, X. Yin, Scalable-manufactured randomized glassy-polymer hybrid metamaterial for daytime radiative cooling. Science 355, 1062–1066 (2017).
17. D. Zhao, A. Alii, Y. Zhai, J. Lu, D. Kidd, G. Tan, X. Yin, R. Yang, Subambient cooling of water: Toward real-world applications of daytime radiative cooling. Joule 3, 111–123 (2018).
18. A. R. Gentle, G. B. Smith, Optimized infra-red spectral response of surfaces for sub-ambient sky cooling as a function of humidity and operating temperature. Photon. Sol. Energy, 79, 7273, 72750Z (2018).
19. E. Rephaeli, A. Ramam, S. H. Fan, Ultrabroadband photonic structures to achieve high-performance daytime radiative cooling. Nano Lett. 13, 1457–1461 (2013).
20. A. P. Ramam, M. A. Anoma, L. Zhe, E. Rephaeli, S. Fan, Passive radiative cooling below ambient air temperature under direct sunlight. Nature 515, 540 (2014).
21. Z. Chen, L. X. Zhu, A. Ramam, S. Fan, Radiative cooling to deep sub-freezing temperatures through a 24-h day-night cycle. Nat. Commun. 7, 13729 (2016).
22. C. Zou, G. Ren, M. M. Hossain, S. Ninartar, W. Withayachumnankul, T. Ahmed, M. Bhaskaran, S. Srimat, M. Gu, C. Fumeaux, Metal-loaded dielectric resonator metamaterials for passive cooling. Adv. Opt. Mater. 5, 1700467 (2017).
23. S. H. Choi, S.-W. Kim, Z. Ku, M. A. Visbal-Onufri, S.-R. Kim, K.-H. Choi, H. K. Wu, C. K. Choi, A. M. Urbas, T.-W. Goo, Y. L. Kim, Anderson light localization in biological nanostructures of native silk. Nat. Commun. 9, 452 (2018).
24. N. N. Shi, C.-T. Tsai, M. J. Carter, J. Mandal, A. C. Overvig, M. Y. Sfeir, M. Li, C. L. Craig, G. B. Bernard, Y. Yang, N. F. Yu, Nanostructured fibers as a versatile photonic platform: Radiative cooling and waveguiding through transverse Anderson localization. Light Sci. Appl. 7, 37 (2018).
25. T. Li, Y. Zhai, S. He, W. Gan, Z. Wei, M. Heidarnejad, D. Dalgo, R. Mi, X. Zhao, J. Song, J. Dai, C. Chen, A. Alii, A. Vellore, A. Martini, R. Yang, J. Srebric, X. Yin, L. Hu, A radiative cooling structural material. Science 364, 760–763 (2019).
26. L. Cai, Y. Peng, J. Xu, C. Zhou, C. Zhou, P. Wu, D. Lin, S. Fan, Y. Cui, Temperature regulation in colored infrared-transparent polyethylene textiles. Joule 3, 1478–1486 (2019).
27. L. M. Lozano, S. Hong, Y. Huang, H. Zandavi, Y. A. El Aoud, Y. Tsurimaki, J. Zhou, Y. Xu, R. M. Osgood, G. Chen, S. V. Borisinka, Optical engineering of polymer materials and composites for simultaneous color and thermal management. Opt. Mater. Express 9, 1990–2005 (2019).
28. A. Synyfa, M. Santamouris, K. Apostolakis, On the development, optical properties and thermal performance of cool colored coatings for the urban environment. Sol. Energy 81, 488–497 (2007).
29. G. J. Lee, Y. J. Kim, H. M. Kim, Y. J. Yoo, Y. M. Song, Colored Passive Radiative Cooler: Colored, daytime radiative coolers with thin-film resonators for aesthetic purposes (Advanced Optical Materials 22/2018). Adv. Opt. Mater. 6, 1870085 (2018).
30. W. Li, Y. Shi, Z. Chen, S. Fan, Photonic thermal management of coloured objects. Nat. Commun. 9, 4240 (2018).
31. H. Konome, M. Nakamura, J. Okajima, S. Murayama, Artificial chameleon skin that controls spectral radiation: Development of chameleon cool coating (C®). Sol. Rep. 8, 1196 (2010).
32. G. B. Smith, A. Gentle, P. Swift, A. Earp, N. Mronga, Coloured paints based on coated flakes of metal as the pigment, for enhanced solar reflectance and cooler interiors: Description and theory. Sol. Energy Mater. Sol. Cells 79, 163–177 (2003).
33. G. B. Smith, A. Gentle, P. D. Swift, A. Earp, N. Mronga, Coloured paints based on iron oxide and silicon oxide coated flakes of aluminium as the pigment, for energy efficient paint: Optical and thermal experiments. Sol. Energy Mater. Sol. Cells 79, 179–193 (2003).
34. J. Song, J. Qin, J. Yu, Z. Song, W. Zhang, Xue, Y. Shi, T. Zhang, W. Ji, R. Zhang, H. Zhang, Z. Zhang, X. Wu, The effects of particle size distribution on the optical properties of titanium dioxide rutile pigments and their applications in cool non-white coatings. Sol. Energy Mater. Sol. Cells 130, 42–50 (2014).
35. X. Lu, P. Xu, H. Wang, T. Yang, J. Hou, Cooling potential and applications prospects of passive radiative cooling in buildings: The current state-of-the-art. Renew. Sustain. Energy Rev. 65, 1079–1097 (2016).
36. M. Zeyhani, D. Y. Goswami, E. Stefanakis, A review of clear sky radiative cooling developments and applications in renewable power systems and passive building cooling. Sol. Energy Mater. Sol. Cells 170, 115–128 (2018).
37. Mikron Instrument Company Inc., Table of emissivity of various surfaces; www-eng.lbl.gov/download-performance-characteristics-data-brochure/https://www.extremematerials-arkema.com/en/product-families/kynar-pvdf-family/download-performance-characteristics-data-brochure/.
38. Arkema, New fluoropolymer latex technology for cool materials solutions across an expanded color space (2008); http://coolcolors.lbl.gov/assets/docs/PAC-2008-03-06-Arkema-slides.pdf.
39. J. Mandal, D. Wang, A. C. Overvig, N. N. Shi, D. Paley, A. Zangiabadi, Q. Cheng, K. Barmak, N. F. Yu, Y. Yang, Scalable, ‘‘dip-and-dry’’ fabrication of a wide-angle plasmonic selective absorber for high-efficiency solar-thermal energy conversion. Adv. Mater. Sci. Rep. 29, 1702156 (2017).
40. H. Karttunen, P. Kröger, H. Oja, M. Poutanen, K. J. Donner, Fundamental Astronomy (Springer, 2016).
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Sci Adv 6 (17), eaaz5413.
DOI: 10.1126/sciadv.aaz5413