# RADIATIVE COOLING Hierarchically structured passive radiative cooling ceramic with high solar reflectivity

Kaixin Lin<sup>1</sup>, Siru Chen<sup>1</sup>, Yijun Zeng<sup>2,3</sup>, Tsz Chung Ho<sup>1</sup>, Yihao Zhu<sup>1</sup>, Xiong Wang<sup>2</sup>, Fayu Liu<sup>2,3</sup>, Baoling Huang<sup>4</sup>, Christopher Yu-Hang Chao<sup>3,5</sup>, Zuankai Wang<sup>3</sup>\*, Chi Yan Tso<sup>1</sup>\*

Passive radiative cooling using nanophotonic structures is limited by its high cost and poor compatibility with existing end uses, whereas polymeric photonic alternatives lack weather resistance and effective solar reflection. We developed a cellular ceramic that can achieve highly efficient light scattering and a near-perfect solar reflectivity of 99.6%. These qualities, coupled with high thermal emissivity, allow the ceramic to provide continuous subambient cooling in an outdoor setting with a cooling power of >130 watts per square meter at noon, demonstrating energy-saving potential on a worldwide scale. The color, weather resistance, mechanical robustness, and ability to depress the Leidenfrost effect are key features ensuring the durable and versatile nature of the cooling ceramic, thereby facilitating its commercialization in various applications, particularly building construction.

nergy used for cooling continues to rise. Carbon dioxide emissions resulting from space cooling have more than doubled to almost 1 billion metric tons over the past 30 years (1). Unfortunately, the carbon footprint of cooling systems contributes to global warming, creating a vicious cycle further exacerbating the need for air-conditioning. To mitigate these environmental concerns, renewable energy sources, such as wind, tidal, and solar power, have been harnessed to meet the ever-increasing energy demand. However, the development and implementation of these energy systems require extensive land use and costly installations. In contrast, passive radiative cooling (PRC), in which surfaces use the cryogenic Universe as a natural heat sink for heat removal, provides an energy-neutral solution for energy transfer and space cooling (2-4).

In recent decades, radiative cooling systems (coolers) of various photonic architectures with subwavelength-scale dimensions have been designed for tuning the thermal spectrum (5–17). Although the tailored long-wave infrared spectrum helps to suppress heat gain from atmospheric radiation, the sophisticated material composition for thermal selectivity sacrifices solar reflectivity ( $R_{solar}$ ). In addition, even though these designs can easily achieve deep nighttime subambient cooling, they struggle to generate cooling during periods of high cooling demand in the daytime.

Atmospheric window (AW) transmittance ( $\tau_{AW}$ ), which serves as the only avenue for ra-

diative heat transfer between the cooler and the Universe, is highly affected by the local climate and environment (18-20). On the other hand, solar radiation dominates the radiative heat exchange of a cooler during daytime given that the Sun (5800 K) emits electromagnetic radiation at an intensity that is 11 orders of magnitude higher than that of a room temperature object (288 K). We numerically solved the steady-state heat transfer model for a skyfacing cooler, enabling estimation of the impact of a cooler's optical properties, that is,  $R_{\rm solar}$  and AW emissivity ( $\epsilon_{\rm AW}$ ), on the cooling performance (fig. S1). The model assumes a constant surrounding air temperature and solar intensity, so that the cooler's cooling power changes according to the cooler's optical properties. The calculation results (fig. S1) clearly indicate that enhancing  $R_{solar}$  can yield a higher cooling capacity than can enhancing  $\varepsilon_{AW}$  by the same amount, regardless of  $\tau_{AW}$ .

However, the development of a daytime PRC material with both high  $R_{\rm solar}$  and practicality for outdoor application is challenging. In contrast to patterned and periodic optical designs that are small-scale and costly, random media, such as particles (21-27) and micropores (28-35), have recently gained attention for achieving desirable daytime PRC. Researchers have investigated disordered designs, such as those of particle-doped polymers (21-27), aerogels (28), porous membranes (29-33), fibers (34, 36), and delignified wood (35). An inherent advantage of these designs is their ability to obviate the need for intricate parameter control, affording simplicity and scalability. However, solar absorption by these materials is inevitable. The dielectric pigments (e.g., titanium dioxide and zinc oxide) and polymers used in these disordered designs absorb sunlight within different wavelength ranges (37, 38). In addition, a high content of organic material potentially leads to degradation during long-term outdoor use (39). For potential application scenarios, such as buildings, this could lead to frequent replacer or maintenance of the building envelope result in exorbitant cost. The trade-off between the optical properties and applicability has always posed a challenge for practical implementation of PRC technology in the real world. Recently, PRC designs using pure inorganic materials to achieve reliable stability offer insight for addressing this challenge (40–42). In this work, we designed a hierarchically porous PRC material in the form of a bulk ceramic, which can be easily obtained using accessible inorganic materials through facile fabrication

processes. The design simultaneously achieves

near-ideal R<sub>solar</sub> and robust applicability, showing

great potential for application in the real world.



Cyphochilus, a beetle native to Southeast Asia, is the whitest known insect on Earth. We learned from the intricate biological structure of this beetle how to design a robust ceramicformat cooler. On the basis of the investigation of the scattering system of the beetle's scales, our cooling ceramic was engineered with a hierarchically porous structure that led to a nearideal  $R_{\text{solar}}$ . This cooler is easily fabricated, requiring neither precision instruments nor meticulous regulation of parameters, and it boasts excellent daytime cooling performance, thus reducing energy consumption for indoor cooling. In the vast forest ecosystem, the beetle's white appearance serves as camouflage against fungal backgrounds, shielding it from predators. The Cyphochilus specimen used for morphological and optical characterization (Fig. 1A) is distinguished by a bright white exoskeleton comprising the head, thorax, and forewings. Upon closer examination using scanning electron microscopy (SEM), we detected the very small scales that densely cover the white areas, with ~15,000 scales/cm<sup>2</sup> (Fig. 1B). These scales, shaped like teardrops, are only 6 µm thick and comprise a highly connected random network of chitin with a filling fraction of  ${\sim}60\%$  and a typical diameter of  $0.25 \pm 0.05 \,\mu m$  (Fig. 1C and fig. S2). Spectroscopic characterization revealed that the scale-covered skeleton exhibits an average  $R_{\rm solar}$  of >60%, in stark contrast to the  $R_{\rm solar}$  value of only 32% for a scale-free skeleton (Fig. 1D and fig. S3). Notably, the scale covering contributed more than 40% to the reflectivity within the visible (VIS) range (0.38 to  $0.75 \,\mu$ m), which is responsible for the beetle's brilliant whiteness. The exceptional visual whiteness of such a thin structure suggests the potential for the development of efficient  $R_{solar}$  in daytime PRC applications.

In the plastic, ink, and paint industries, products are often rendered white by incorporating pigments with high refractive index values into polymers. Similarly, the whiteness of *Cyphochilus* beetle scales can be attributed to a scattering system comprising a chitin filament network



<sup>&</sup>lt;sup>1</sup>School of Energy and Environment, City University of Hong Kong, Hong Kong, China. <sup>2</sup>Department of Mechanical Engineering, City University of Hong Kong, Hong Kong, Objachina. <sup>3</sup>Department of Mechanical Engineering, The Hong Kong Polytechnic University, Hong Kong, China. <sup>4</sup>Department of Mechanical and Aerospace Engineering, Hong Kong University of Science and Technology, Hong Kong, China. <sup>5</sup>Department of Building Environment and Energy Engineering, The Hong Kong Polytechnic University, Hong Kong, China.

<sup>\*</sup>Corresponding author. Email: chiytso@cityu.edu.hk (C.Y.T.); zk.wang@polyu.edu.hk (Z.W.)



Fig. 1. Whitest beetle in the world. (A) Photograph of the *Cyphochilus* specimen demonstrating its white appearance. (B) SEM image of *Cyphochilus* scales.
(C) Cross-sectional SEM image of a *Cyphochilus* scale. (D) Measured R<sub>solar</sub> spectra of *Cyphochilus* exoskeleton surfaces with and without scale covering.
(E) Simulated backscattered power for incident light of four wavelengths (0.3, 0.5, 0.7, and 1.0 µm) as a function of the chitin filament diameter. (F) Simulated scattering center dimension-dependent variation in the ratio of the backscattered power and the total scattered power for 0.5 µm incident light. The orange shading indicates the size distribution of the chitin filament.

and a complementary interconnected structure of air pockets. To better understand the scale whiteness, we modeled the Mie scattering behavior of chitin filament using Maxwell's equations (fig. S4). Considering a refractive index value of 1.56 to simulate chitin (43), we calculated the backscattering intensity at four incident wavelengths covering the ultraviolet (UV) to near-infrared (NIR) ranges (Fig. 1E). The results show that chitin filament with a diameter of 0.3 µm strongly scatters light with blue wavelengths, and as the size of chitin filament increases, the peak of the scattering intensity shifts toward red wavelengths (Fig. 1E). Because most Cyphochilus chitin filaments are between 0.2 and 0.3 µm in size, the scale most efficiently scatters light between 0.5 and 0.7 µm, which results in high opaqueness within the visible range. Additionally, we calculated the ratio of the backscattering intensity to the overall scattering for 500-nm incident light and found that the beetle's chitin filament dimension is notably coupled with one of the backscattering peaks located at 0.25 µm, confirming that the size of chitin filament is optimized for maximum scattering of visible light to achieve high visual whiteness (Fig. 1F).

#### High-solar reflectivity cooling ceramic

On the basis of the biostructure analysis, we engineered alumina particles through phase

inversion and sintering to obtain the cooling ceramic (Fig. 2A and fig. S5). A three-component homogeneous solution, composed of polvethersulfone (PES), N-methyl-2-pyrrolidone (NMP), and  $\alpha$ -alumina, was cast on a flat substrate and immersed in ethanol. The phase inversion takes place when the ethanol diffuses into the casting and dissolves with NMP. This process results in a polymer-rich membrane, forming an anisotropic porous network carrying alumina particles. Note that the porous alumina-polymer membrane can be flexible and imprinted with patterns or shaped into any form desired. In a high-temperature sintering process, PES starts to combust above 500°C. Both energy-dispersive spectroscopy (EDS) mapping (fig. S6A) and thermogravimetric analysis (TGA) (fig. S6B) confirmed the complete removal of PES through the sintering process. Meanwhile, alumina particles were bonded, resulting in a cooling ceramic with a well-preserved porous structure. Manipulating the alumina concentration in the phase inversion precursor solution vielded a short transport mean free path leading to high R<sub>solar</sub> of the thin cooling ceramic (fig. S7). The as-obtained cooling ceramic shows intense whiteness under natural light (Fig. 2A) and has a structure similar to that of Cyphochilus scales, with a densely packed outer layer and numerous internal voids, but with a wider structural dimension distribution (Fig. 2, B and C). The cooling ceramic exhibits near-perfect reflectivity in the UV, VIS, and NIR ranges, leading to an  $R_{solar}$  value of 99.6%, versus a value of 89.5% for silver, a value of 88.6% for white-pigmented polymer, and a value of 76.2% for white commercial tiles (Fig. 2D). For scattering systems containing a certain volume of pores, a multidispersed pore system is more favorable than a monodispersed system for scattering in a broadband way (Fig. 2E and fig. S8) (26). This explains why hierarchical pores render the cooling ceramic not only white in the VIS range, similar to the appearance of *Cyphochilus*, but also white in the UV and NIR ranges.

The cooling ceramic is composed solely of porous  $\alpha$ -alumina, which has ideal intrinsic electromagnetic properties for PRC applications. Among common white and transparent inorganic materials, alumina has the top-ranked high bandgap with a relatively high refractive index (fig. S9). The bandgap of  $\alpha$ -alumina reaches 7.0 eV, which is well above the upper boundary for photon energy in the solar spectrum (4.13 eV) (44, 45). The high bandgap of  $\alpha$ -alumina results in a low extinction coefficient across the solar wavelength range (fig. S10) and a low absorption of solar photons, which makes the high  $R_{\rm solar}$  possible. On the other hand, arising from the vibrational modes of the Al-O chemical bonds, alumina attains a high extinction peak within the AW range



Fig. 2. Engineered hierarchically porous cooling ceramic. (A) Photograph of the cooling ceramics with flat and curved shapes. Patterns can also be applied on the cooling ceramic surface. (B) SEM images of a fabricated cooling ceramic sample showing the hierarchically porous structure. (C) Volume concentration of pores within the cooling ceramic. (D) Comparison of the optical properties of the white cooling ceramic with a 600  $\mu$ m thickness and a 70% porosity to those of white-

pigmented polymer (Al<sub>2</sub>O<sub>3</sub>-doped polydimethylsiloxane), silver, and white commercial tile. (**E**) Simulated scattering efficiency of porous alumina systems, including three monodispersed systems and one multidispersed system (the total pore volume concentrations are the same). The multidispersed case includes 0.3, 0.5, and 2  $\mu$ m pores with a volume concentration ratio of 1:1:2. (**F**) Angular  $R_{solar}(\theta)$  value of the cooling ceramic. (**G**) Angular  $\epsilon_{AW}(\theta)$  value of the white cooling ceramic.

(fig. S10), resulting in a high  $\epsilon_{AW}$  of 96.5%. Therefore, the cooling ceramic can efficiently radiate heat through the AW, where peak blackbody emissions from terrestrial surfaces coincide with the high atmospheric transmittance into space. Considering the high  $R_{\rm solar}$ and high  $\varepsilon_{AW}$ , the cooling ceramic can be regarded as superior to other state-of-the-art PRC material counterparts (fig. S11). The whiteness of the cooling ceramic was achieved in a diffused way, with high angular  $R_{\rm solar}$  across its full solid angle (Fig. 2F). At the thermal wavelengths, the angular emissivity was also consistent, with only a slight decrease at low zenith angles. We attributed the high  $\varepsilon_{AW}$ value of the cooling ceramic within the wide angular range (Fig. 2G) to the open, porous surface and the effective medium behavior of the nanopores at high wavelengths (33).

The efficient scattering by the hierarchical pore system enables the cooling ceramic to achieve the desired optical performance in a materialsaving manner. Cooling ceramic with a thickness of only 150  $\mu$ m can achieve  $R_{solar} > 95\%$ , while high-performance roof cooling coatings typically require >1 mm thickness to achieve the same level of optical performance (46) (fig. S12). As the thickness of the cooling ceramic increases, its solar transmittance decreases because of increased backscattering, leading to continuous enhancement of  $R_{\rm solar}$ . Eventually, when the thickness of the cooling ceramic reaches 600 µm, its  $R_{\rm solar}$  reaches saturation at 99.6% (fig. S12). On the other hand, the cooling ceramic exhibits low reflectivity within the AW range at any thickness. Therefore, a thick cooling ceramic can dissipate heat through its own high emissivity, and a thin cooling ceramic does not hinder the radiative dissipation of its substrate, making it favorable for application on substrates with already adequate  $\varepsilon_{\rm AW}$  values—concrete, for example.

# **Cooling performance assessment**

We demonstrated the cooling performance of our ceramic using a custom-designed thermal setup (27) in Hong Kong and compared it to that of commonly used white commercial tiles as a control ( $R_{solar} = 76.2\%$ ,  $\varepsilon_{AW} = 88.5\%$ ) (Fig. 3A and table S1). Throughout a continuous 84-hour measurement period, the cooling ceramic consistently maintained a temperature below the ambient air temperature, with an average subambient temperature difference of 3.8°C and a maximum subambient temperature difference of 8.8°C (Fig. 3B and fig. S13). In contrast, the white commercial tiles only

provided cooling at night. During the daytime, the cooling ceramic delivered an average subambient temperature reduction of 4.3°C. Given perfect  $R_{solar}$ , the cooling ceramic could achieve >4°C subambient cooling even around midday (between 11 a.m. and 2 p.m.), leading to a  $>8^{\circ}$ C lower temperature than that of the white commercial tiles. At the same time, another identical setup coupled with a controllable heating system was installed in parallel to measure the cooling power of the samples. To ensure repeatability, we conducted cooling power measurements over four short periods (Fig. 3C). During the two measurements at night, the cooling ceramic generated average cooling power of 142 and 125 W/m<sup>2</sup>, as compared with values of 128 and 117.3 W/m<sup>2</sup> for the white commercial tiles (Fig. 3C). To examine the cooling performance of the cooling ceramic under extreme incoming heat flux conditions, the two davtime measurements were conducted around solar noon, at a solar intensity of up to 800 W/m<sup>2</sup>. Despite the challenging conditions, the cooling ceramic demonstrated exceptional cooling, with a cooling power of 134.5 and 133.8 W/m<sup>2</sup> for the two measurements. In contrast, the white commercial tiles, which strongly absorb solar



Fig. 3. Cooling performance characterization. (A) Experimental setup for characterizing the subambient cooling and cooling power. (B) Sample temperatures, ambient air temperature, solar intensity, wind speed, and relative humidity, measured in Hong Kong over 84 continuous hours from 11 to

14 November 2021. (**C**) Cooling power measurements during four periods: 11 November, 20:30–21:30; 12 November, 12:15–14:25; 13 November, 13:45–14:45; and 13 November, 22:15–22:50. (**D**) Temperatures of the samples and ambient air, measured in Philadelphia, Yellowstone National Park, and Boston.

radiation, failed to generate cooling power. For traditional daytime PRC materials, a higher cooling power can normally be obtained at night, while the daytime cooling power is obviously lower than that at night because of solar heat absorption (22, 35). As a result of the ideal  $R_{\rm solar}$ , the cooling ceramic exhibited comparable or even superior cooling performance during the daytime as compared with that at night, which is highly desired for addressing the high daytime cooling energy demand. It should be noted that Hong Kong is a coastal city with a humid climate, which is very unfavorable for heat dissipation through thermal radiation. The average humidity during the field test was around 50%. Considering a 3000 atm-cm water column (47), the local  $\tau_{AW}$  during the field test was calculated at <0.6 (48), which is not ideal for PRC applications (fig. S1). Although direct comparison to the cooling performance determined in other studies is virtually impossible given the notable influence of geography and meteorological variances on the local field test results, the high cooling power exceeded  $100 \text{ W/m}^2$ under these unfavorable test conditions, strongly indicating that the cooling performance of the engineered cooling ceramic is superior to recent results reported in the literature.

Apart from Hong Kong, we also validated the cooling performance of the cooling ceramic in different climates. In Yellowstone National Park (US), the cooling ceramic achieved a cooling effect of subambient temperature reduction of between 1° and 7°C at noon (Fig. 3D). In urban areas, such as Philadelphia (US) and Boston (US), the cooling ceramic also achieved a subambient temperature reduction of about 3°C (Fig. 3D). For the field test conducted in Beijing, the cooling ceramic showed 3.3°C subambient temperature reduction around midday (between 13:30 and 14:30), with an average solar intensity value of 876 W/m<sup>2</sup>, whereas the white porous polymer cooler ( $R_{solar} = 96.2\%$ ,  $\epsilon_{\rm AW}$  = 96.0%) and white porous alumina-polymer membrane ( $R_{\rm solar}$  = 97.6%,  $\varepsilon_{\rm AW}$  = 97.2%) only showed 2.4° and 2.7°C subambient temperature reduction, respectively; the white commercial tile failed to achieve subambient temperature reduction (fig. S14). These results demonstrate the ability of the cooling ceramic to ensure stable cooling performance in various application environments.

# **Energy-saving evaluation**

External building surfaces are the main recipients of solar radiation and contribute notable heat gains to the indoor environment. The most direct and appealing method to exploit PRC technology would be to use the coolers to cover the building envelope surfaces, where the cooler can be directly exposed to the sky to reduce the building thermal load. To investigate the cooling effect in real applications, two identical model houses were constructed (Fig. 4A). One of them was roofed with white cooling ceramic, while the other was roofed with white commercial tiles purchased from the market as control. The model house roofs were designed with 30° pitch angles to increase the projection areas for direct solar irradiation. First, we conducted continuous thermal measurement for the two model houses over 4 days. The roof temperature showed a large daytime difference between the two model houses (Fig. 4B). Specifically, the roof with white cooling ceramic could be almost 5°C cooler than the commercial tiled roof at noon (Fig. 4B). The lower roof temperature meant reduced heat transfer from the roof to the indoor space, resulting in a lower indoor temperature for the model house fitted with the white cooling ceramic (Fig. 4B). The indoor air temperature difference reached a maximum of 2.5°C. The thermal load of a building is directly related to the cooling demand for regulating the indoor environment. To intuitively quantify the energy-saving potential of the engineered cooling ceramic, we conducted additional assessments by operating an air-conditioning unit within the model houses in the summer (July 2022). We continuously monitored the airconditioning electricity usage over three periods, with set temperatures of 25°, 23° and 20°C. With less heat load, the cooling ceramic-tiled model house consumed less electricity, with energy savings of 26.8, 22.6, and 19.6%, respectively, during each set temperature period (Fig. 4C). Moreover, we conducted an energy consumption



**Fig. 4. Application as a building envelope.** (**A**) Photograph of the model houses, with the white cooling ceramic and white commercial tile applied on the roof (area: ~1.15 m<sup>2</sup>). The model houses were installed 2 m apart during the experiment to eliminate interference. (**B**) Differences in the roof and indoor air

temperatures for the two model houses. (**C**) Electricity usage of the two model houses with air-conditioning set points of 25°, 23°, and 20°C. (**D**) Energy-saving performance on a worldwide scale considering the energy consumed by cooling systems, fans, and heating equipment.

simulation of a full-scale building with a model of a typical four-story midrise apartment building (fig. S15 and table S2) to assess the energysaving performance on a worldwide scale by applying the cooling ceramic as the external envelope material on the walls and roof (Fig. 4D). Considering the energy usage of HVAC (heating, ventilation, and air-conditioning) systems, application of the cooling ceramic could benefit tropical regions the most. In particular, the annual energy savings could reach >10% (25 GJ, or ~7000 kWh) per year for indoor air-conditioning in these extremely hot areas.

# Applicability investigation

Beyond cooling, the cooling ceramic could also offer a variety of other functionalities, bringing it closer to practical applications. The interaction between water and surfaces at high temperatures is a critical yet often overlooked phenomenon in the applications involving evaporative cooling. When a building experiences a fire, the heat generated by combustion can cause extensive damage. To extinguish a fire and lower a building's temperature, it is essential to allow direct contact between the envelope surface and water using evaporative cooling. Most commercial tiles prevent water from wetting the

overheated surface at temperatures above 280°C (Fig. 5A, fig. S16, and movie S1), resulting in poor evaporative cooling due to the Leidenfrost effect (49, 50). In contrast, our cooling ceramic exhibits superhydrophilicity enabling immediate droplet spreading, while facilitating rapid impregnation of droplets by its interconnected porous structure (fig. S17). Consequently, the cooling ceramic inhibits the Leidenfrost effect at temperatures above 800°C (Fig. 5A and movie S1) during the evaporative cooling process. The effective evaporative cooling performance of the cooling ceramic was demonstrated by the rapidly decreasing surface temperature, whereas only a negligible temperature variation was recorded for the commercial tiles by infrared camera (Fig. 5B and movie S2).

The cooling ceramic can also be converted from superhydrophilic to hydrophobic by impregnation with organosilicon compounds, which is the most commonly used method to protect porous materials against moisture when applications call for water repellence. Upon treatment with fluorosilane, the cooling ceramic featured high superhydrophobicity with a water contact angle of ~150° (fig. S18). The interconnected porous structure of the cooling ceramic enabled the fluorosilane solution to penetrate the material, resulting in water resistance on both the surfaces and interior (fig. S18). Importantly, this treatment does not affect the high-temperature evaporative cooling performance of the cooling ceramic because the fluorosilane chemical groups start to evaporate at a temperature above 250°C (fig. S19). In terms of the optical performance of the fluorosilane-treated cooling ceramic, its  $R_{solar}$ dropped slightly to around 99.0% owing to the absorption of bonded fluorosilane chemical groups (fig. S20). A dust resistance test using standard pollutants (GB/T 97780-2013) confirmed good antipollution performance of the fluorosilane-treated cooling ceramic with  $R_{\rm solar}$  of >97% maintained after multiple pollutantwashing test cycles (fig. S21).

The cooling ceramic has a dense all-inorganic structure that inherently endows it with excellent resistance to UV radiation. The extremely low UV absorption and high bond strength of alumina makes the cooling ceramic less susceptible than polymer-based coolers to photodegradation. For verification, the cooling ceramic was exposed to a UV lamp with a 5 W/m<sup>2</sup> UV power for 3 months. The optical properties measured before and after UV exposure showed negligible differences, confirming the high resistance of the cooling ceramic to UV degradation (fig. S22). A durability test of the



**Fig. 5. Applicability of the cooling ceramic. (A)** Water droplets contacting the commercial tiles and cooling ceramic at a surface temperature of 600°C. With the commercial tiles, water cannot directly contact the surface owing to the Leidenfrost effect. After spreading due to the impact force, the droplets bounce and return to a spherical shape. In contrast, water on the cooling ceramic is constantly pinned and remains flattened upon spreading, leading to rapid evaporation. Scale bars, 2 mm. (B) Variation in the surface temperature when the samples are contacted by water droplets at 5-s intervals. The infrared thermal images show the surface temperature of the samples

after thermal stabilization. Scale bars, 5 cm. (**C**) Photographs of the fabricated colored cooling ceramics, along with colored commercial tiles showing identical vivid colors. (**D**) Chromaticity of the colored cooling ceramic and colored commercial tiles in the CIE 1931 color space. (**E**) Comparison of the  $R_{\text{solar}}$  spectra of yellow, red, green, and black cooling ceramics to those of commercialized colored tiles of the same color. (**F**) Outdoor temperature measurements of the colored cooling ceramics around midday, using colored commercial tiles as a control. The line color corresponds to the sample color.

cooling ceramic under real-world conditions was also conducted by exposing it in an outdoor space for 1 year, to solar radiation equaling ~5000 MJ (51). The results show that the  $R_{\rm solar}$ of the cooling ceramic only dropped by 1.3%, still reaching 98.3% (fig. S23). Because alumina is nonflammable, the cooling ceramic can remain intact and undamaged even under extreme fire exposure (fig. S24). The cooling ceramic also demonstrates high mechanical strength, with a breaking strength of >100 MPa (bending test according to ISO 10545-4), which meets the standards for application as building envelope materials (>35 MPa) (fig. S25). Furthermore, the cooling ceramic can be recycled (fig. S26), which is highly desirable for sustainability. When damaged or polluted, the developed cooling ceramic can be ground into raw materials (alumina particles) and then reused in the fabrication of new cooling ceramic with well-preserved optical properties.

High and broadband  $R_{\rm solar}$  is crucial for cooling, yet completely reflecting all visible light would yield a white cooler, which cannot fulfill the aesthetic needs of contemporary urban landscapes. On the basis of the white cooling ceramic, we have developed a colored cooling ceramic that attains equilibrium between chromatic presentation and reduction of thermal load. The colored cooling ceramic was obtained simply by sintering a thin layer of colored glaze on top of the white cooling ceramic. We fabricated four colored cooling ceramics: yellow, red, green, and black. The color of the cooling ceramic was controlled to match that of commercial tile products purchased from the market (Fig. 5, C and D). Both the colored commercial tiles and colored cooling ceramics had high absorption in VIS light for creating vivid color (Fig. 5E and table S3). As the thin color glaze of the colored cooling ceramic enables NIR light to transmit through and be reflected by the underlying highly reflective layer, the colored cooling ceramic attained substantially higher reflectivity in the NIR spectrum (95, 96, 87, and 39% for the yellow, red, green, and black ceramics, respectively) than the colored commercial tiles (76, 57, 66, and 16% for

the yellow, red, green, and black tiles, respectively) (Fig. 5E and table S3). We experimentally verified the thermal performance of the colored ceramics against the colored commercial tiles by exposing the samples to direct sunlight at midday (Fig. 5F). The red cooling ceramic maintained an average temperature 4.7°C lower than that of the commercial red tile, followed by 2.8°, 1.7°, and 1.3°C temperature reductions for the yellow, green, and black cooling ceramics, respectively (Fig. 5F). The temperature reduction obtained for each color corresponded well to the  $R_{solar}$  contrast between the cooling ceramic and commercial tiles. The light absorption in the visible range determines the color intensity, which therefore influences the overall  $R_{solar}$ . With high NIR reflectivity, the colored cooling ceramic is also able to achieve  $R_{solar}$  of >0.9 by tuning the concentration of color glazing (fig. S27). These results confirmed that the colored design based on the white cooling ceramic could provide the desired color along with high NIR  $R_{solar}$  and showcased the great potential in reducing the thermal load and even subambient cooling. Finally, the colored cooling ceramics can be fabricated into curved shapes using the same procedure as for the white cooling ceramics. This makes it a versatile material suitable for a wide range of applications, for example, houses with curved roofing tiles (fig. S28). The ability to shape the material into curved forms allows for more creative and distinctive designs, adding aesthetic value to different applications.

# Conclusions

In this study, we developed an energy-free and robust daytime PRC material in the form of ceramic for reducing the cooling demand of the indoor environment. The cooler features a biomimetic porous structure, which efficiently scatters solar irradiation to achieve an  $R_{solar}$  of 99.6%. In real-world application demonstrations and whole-building energy simulations, our cooling ceramic demonstrated promising energy-saving potential. With high weather resistance, high mechanical strength, favorable recyclability, notable Leidenfrost depression, and optional color features, the cooling ceramic can readily be applied to different scenarios

and outdoor infrastructures on a large scale. With its combination of advanced features and demonstrated performance, the technology holds great potential for contributing to the development of more sustainable and energyefficient building solutions in the future. Although we have not yet explored the cooling power modulation of the cooling ceramic, given its high  $R_{solar}$ , the cooling ceramic may integrate with adaptive optical materials to obtain the temperature-response regulation on optical properties and therefore smartly suppress the cooling effect during cold weather (52, 53). On the other hand, to retain a high radiative cooling power with vivid color, we can combine the cooling ceramics with photoluminescent materials (54), such as carbon dots, to recover the solar absorption in the visible range.

#### **REFERENCES AND NOTES**

- International Energy Agency (IEA), Space Cooling (IEA Paris, 2022); 1. https://www.iea.org/energy-system/buildings/space-cooling.
- 2 D. L. Zhao et al., Appl. Phys. Rev. 6, 021306 (2019).
- X. Yin, R. Yang, G. Tan, S. Fan, Science 370, 786-791 (2020)
- S. H. Fan, W. Li, Nat. Photonics 16, 182-190 (2022). 4.
- E. Rephaeli, A. Raman, S. Fan, Nano Lett. 13, 1457-1461 (2013). A. P. Raman, M. A. Anoma, L. Zhu, E. Rephaeli, S. Fan, Nature 6.
- 515, 540-544 (2014). M. M. Hossain, B. H. Jia, M. Gu, Adv. Opt. Mater. 3, 1047-1051 7 (2015)
- 8. S. Y. Jeong, C. Y. Tso, Y. M. Wong, C. Y. H. Chao, B. Huang, Sol. Energy Mater. Sol. Cells 206, 110296 (2020).
- D. G. Baranov et al., Nat. Mater. 18, 920-930 (2019).
- 10. H. Zhang et al., Proc. Natl. Acad. Sci. U.S.A. 117, 14657-14666
- 11. C. J. Zou et al., Adv. Opt. Mater. 5, 1700460 (2017).
- 12. H. C. Ma et al., Sol. Energy Mater. Sol. Cells 212, 110584 (2020)
- 13. C. Lin et al., Adv. Mater. 34, e2109350 (2022).
- 14. J. L. Kou, Z. Jurado, Z. Chen, S. H. Fan, A. J. Minnich, ACS Photonics 4. 626-630 (2017).
- 15. S. Y. Jeong et al., Renew. Energy 146, 44-55 (2020)
- 16. D. Lee et al., Nano Energy 79, 105426 (2021).
- 17. Y. Fu, J. Yang, Y. S. Su, W. Du, Y. G. Ma, Sol. Energy Mater. Sol. Cells 191, 50-54 (2019).
- 18. J. W. Liu et al., Sol. Energy Mater. Sol. Cells 208, 110412 (2020). 19. D. Han, B. F. Ng, M. P. Wan, Sol. Energy Mater. Sol. Cells 206, 110270 (2020).
- 20. C. Y. Tso, K. C. Chan, C. Y. H. Chao, Renew. Energy 106, 52-61 (2017).
- 21. X. Li, J. Peoples, P. Yao, X. Ruan, ACS Appl. Mater. Interfaces 13, 21733-21739 (2021).
- 22. X. Y. Li et al., Cell Rep. Phys. Sci. 1, 100221 (2020).
- 23. Z. Tong et al., Mater. Today Phys. 24, 100658 (2022)
- 24. J. Mandal, Y. Yang, N. F. Yu, A. P. Raman, Joule 4, 1350-1356 (2020) 25. X. Xue et al., Adv. Mater. 32, e1906751 (2020)
- 26. J. Peoples et al., Int. J. Heat Mass Transf. 131, 487-494
- (2019)
- 27. K. X. Lin et al., Energy Build. 276, 112507 (2022).

- 28. A. Leroy et al., Sci. Adv. 5, eaat9480 (2019).
- 29. H. Zhong et al., ACS Appl. Mater. Interfaces 12, 51409-51417 (2020).
- 30 | Li et al. Sci Adv 8 eabi9756 (2022)
- 31. M. Li et al., Small 19, e2301159 (2023).
- 32. W. L. Huang et al., Adv. Funct. Mater. 31, 2010334 (2021). 33. J. Mandal et al., Science 362, 315-319 (2018).
- 34. S. Zeng et al., Science 373, 692-696 (2021).
- 35. T. Li et al., Science 364, 760-763 (2019).
- 36. D. Li et al., Nat. Nanotechnol. 16, 153-158 (2021).
- 37. A. Aili et al., Mater. Today Phys. 10, 100127 (2019).
- 38. J. R. Song et al., Sol. Energy Mater. Sol. Cells 130, 42-50 (2014). 39. H. Zweifel, Stabilization of Polymeric Materials (Springer
- Science and Business Media, 2012). 40. S. K. Jeon et al., Adv. Sci. (Weinh.) 10, e2302701 (2023).
- 41. T. Li et al., Chem. Eng. J. 452, 139518 (2023).
- 42. M.-T. Tsai et al., Nano Today 48, 101745 (2023)
- 43. M. Burresi et al., Sci. Rep. 4, 6075 (2014).
- 44. A. Felicelli et al., Cell Rep. Phys. Sci. 3, 101058 (2022). 45. E. O. Filatova, A. S. Konashuk, J. Phys. Chem. C 119, 20755-20761 (2015).
- 46. APOC, Technical and Safety Data Sheets; https://www.apoc. com/pages/technical-data-sheets.
- 47. Spectral Sciences Inc., MODTRAN (MODerate resolution atmospheric TRANsmission); http://modtran.spectral.com/.
- 48. C. Ziming et al., AIMS Energy 9, 96-116 (2021).
- 49. D. Quéré, Annu. Rev. Fluid Mech. 45, 197-215 (2013).
- 50. M. Jiang et al., Nature 601, 568-572 (2022).
- 51. Hong Kong Observatory, Climatological Information Services Climate of Hong Kong; https://www.hko.gov.hk/en/cis/ climahk.htm.
- 52. S. Wang et al., Science 374, 1501-1504 (2021).
- 53. J. Yang et al., Adv. Mater. 33, e2004754 (2021).
- 54. X. Wang et al., Sci. Bull. (Beijing) 67, 1874-1881 (2022)

# ACKNOWLEDGMENTS

Funding: This work was supported by Hong Kong Research Grant Council via General Research Fund (GRF) accounts 11200022. 11200121, and 11200923 and by the Meituan Green Tech Fund. Author contributions: Conceptualization: K.L. and C.Y.T. Methodology: K.L. C.Y.T., X.W., and F.L. Investigation; K.L., S.C., Y.Ze., Y.Zh., and C.Y.-H.C. Visualization: K.L. and C.Y.T. Funding acquisition: C.Y.T. Project administration: C.Y.T. Supervision: C.Y.T. and Z.W. Writing - original draft: K.L. and C.Y.T. Writing - review & editing: C.Y.T., Z.W., B.H., and T.C.H. Competing interests: A US nonprovisional patent (application no. 17/89.878) related to this work has been filed. K.I S.C., Y.Zh., T.C.H., and C.Y.T. are inventors of this filed patent Data and materials availability: All data are available in the main text or the supplementary materials. License information: Copyright © 2023 the authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. No claim to original US government works. https://www.science.org/ about/science-licenses-journal-article-reuse

#### SUPPLEMENTARY MATERIALS

science.org/doi/10.1126/science.adi4725 Materials and Methods Supplementary Text Figs, S1 to S28 Tables S1 to S3 References (55-64) Movies S1 and S2

Submitted 28 April 2023; accepted 19 September 2023 10.1126/science.adi4725



# Hierarchically structured passive radiative cooling ceramic with high solar reflectivity

Kaixin Lin, Siru Chen, Yijun Zeng, Tsz Chung Ho, Yihao Zhu, Xiong Wang, Fayu Liu, Baoling Huang, Christopher Yu-Hang Chao, Zuankai Wang, and Chi Yan Tso

Science 382 (6671), . DOI: 10.1126/science.adi4725

# Editor's summary

Passive radiative cooling materials emit heat through the atmospheric window and into outer space, providing an attractive way to reduce temperatures in buildings. Zhao *et al.* created a passive cooling glass and Lin *et al.* developed a passive cooling ceramic, both of which are mechanically strong and relatively easy to scale (see the Perspective by Zhao and Tang). Unlike strategies that rely on polymers, these hard materials should be more robust to long-term weathering, which may make them far more useful for outdoor applications. —Brent Grocholski

# View the article online

https://www.science.org/doi/10.1126/science.adi4725 Permissions https://www.science.org/help/reprints-and-permissions

Science (ISSN 1095-9203) is published by the American Association for the Advancement of Science. 1200 New York Avenue NW, Washington, DC 20005. The title Science is a registered trademark of AAAS.

Copyright © 2023 The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. No claim to original U.S. Government Works