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1 Ultrahigh Passive Cooling Power in Hydrogel with Rationally 2 Designed Optofluidic Properties

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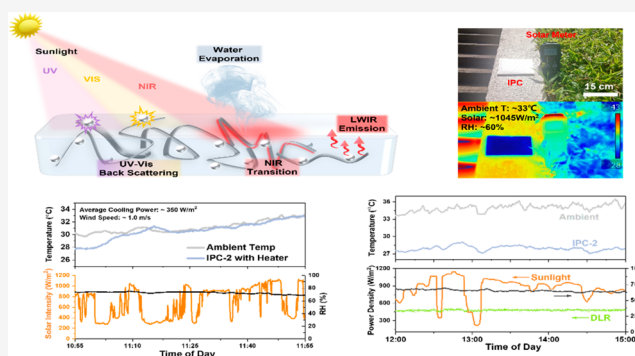
Article Recommendations



Supporting Information

6 **ABSTRACT:** The cooling power of a radiative cooler is more
7 than halved in the tropics, e.g., Singapore, because of its harsh
8 weather conditions including high humidity (84% on average),
9 strong downward atmospheric radiation (~40% higher than
10 elsewhere), abundant rainfall, and intense solar radiation (up to
11 1200 W/m² with ~58% higher UV irradiation). So far, there has
12 been no report of daytime radiative cooling that well achieves
13 effective subambient cooling. Herein, through integrated passive
14 cooling strategies in a hydrogel with desirable optofluidic
15 properties, we demonstrate stable subambient (4–8 °C)
16 even under the strongest solar radiation in Singapore. The
17 integrated passive cooler achieves an ultrahigh cooling power of
18 ~350 W/m², 6–10 times higher than a radiative cooler in a
19 tropical climate. An in situ study of radiative cooling with various hydration levels and ambient humidity is conducted to understand
20 the interaction between radiation and evaporative cooling. This work provides insights for the design of an integrated cooler for
21 various climates.

22 **KEYWORDS:** *optofluidic design, passive cooling, radiative cooling, hydrogel, integrated cooling structure*



23 **T**o achieve a sustainable development, COP26 set the
24 ambitious target of net-zero carbon emission by the
25 middle of the century, which urgently calls for low-carbon
26 technologies.^{1–4} Energy-related carbon emissions totaled 31.5
27 billion metric tons in 2020. Active cooling consumes a
28 considerable amount of energy, especially in the summer
29 season of most countries and in all seasons of tropical zones
30 such as Singapore. For instance, cooling is responsible for over
31 60% of the electricity consumption in nonresidential buildings
32 in Singapore, where 95% of the electricity comes from burning
33 natural gas.⁵ Thus, developing an energy-saving cooling
34 method is crucial for achieving the COP26 goal of carbon
35 neutrality and sustainability.

36 Passive cooling, free of energy consumption, can be realized
37 by various strategies such as radiative cooling, evaporative
38 cooling, high solar reflection, heat insulation, etc.⁶ Recently,
39 passive radiative coolers (PRCs) have attracted ever-increasing
40 attention, which achieve cooling by emitting long-wave
41 infrared (LWIR) radiation to outer space through the
42 atmospheric window (8–13 μm).^{7–12} Daytime subambient
43 cooling in many temperate regions is achievable when the PRC
44 has an ultrahigh solar reflectivity (R_{solar}) of more than 94% to
45 greatly reduce solar heat gain.⁷ However, effective thermal
46 radiation to outer space in the tropical region is significantly

impeded by the high relative humidity (RH) and abundant
47 thick clouds, leading to more than halved cooling power,¹³ as
48 evidenced by the exceptionally high downwelling radiation
49 from the sky (up to 500 W/m²). As a result, effective cooling
50 power through the atmospheric window is significantly lower
51 (down to <15 W/m²) than its theoretical upper limit of ~150
52 W/m².¹⁴ Thus far, subambient radiative cooling has hardly
53 been achieved in a tropical climate with >1000 W/m² solar
54 irradiance even using the best reported PRC ($R_{\text{solar}} \approx 97\%$ and
55 long-wavelength infrared emissivity $E_{\text{LWIR}} \approx 96\%$).¹⁵ 56

As a very effective method, evaporative cooling has been
57 employed from ancient times (e.g., an Egyptian qullah) to
58 modern days (e.g., a Zeer fridge in Africa). However, it is far
59 from providing optimal cooling in terms of cooling power per
60 unit of water consumption. Very recently, a combination of
61 evaporative and radiative cooling has been attempted in a 62

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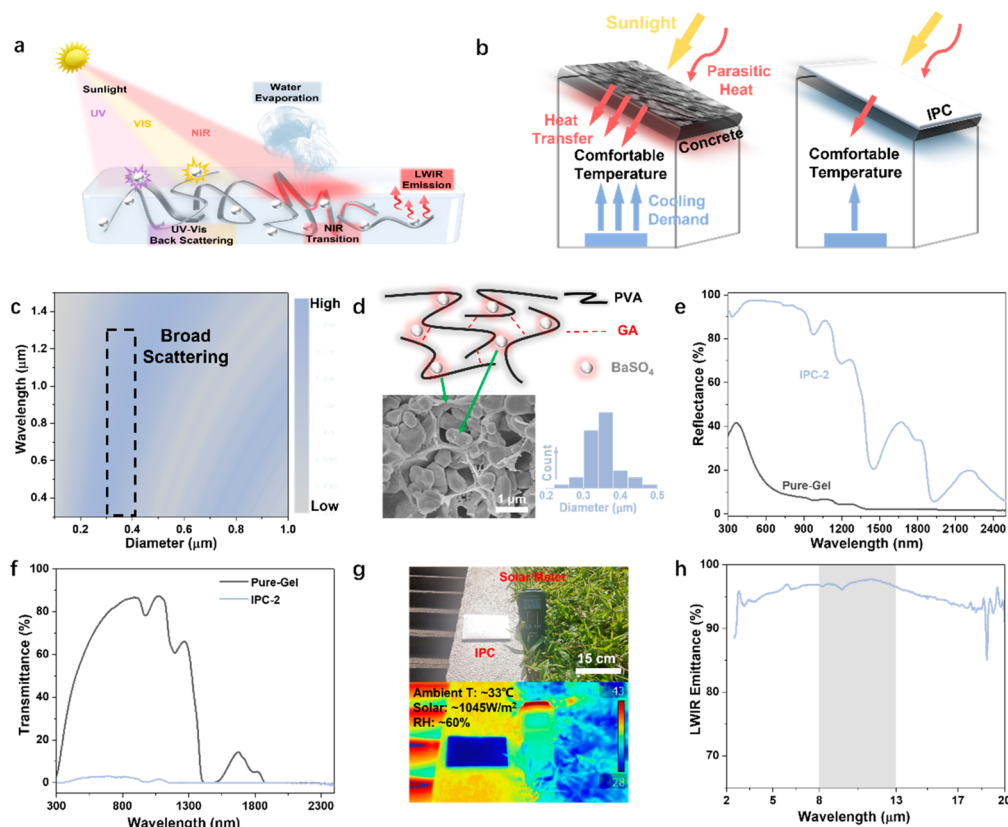


Figure 1. IPC design and characterizations. (a) Schematic illustration of cooling mechanisms. (b) Illustration of active cooling demand (ACD) difference with bare (left) and IPC-covered (right) concretes. (c) Simulated scattering efficiency (300–1300 nm) with varying BSP sizes. (d) Structural illustration (upper) and scanning electron microscopy image (lower left, scale bar is 1 μm) of IPC and BSP size distribution (lower right). (e) Solar reflectance and (f) transmittance spectra of pure-Gel and IPC-2. (g) Optical (upper) and IR image (lower) of IPC-2 under sunlight taken after 30 min stabilization. (h) LWIR emittance spectrum of IPC-2. The shaded area labels the atmospheric window wavelength range.

63 double-layer hydrogel–hydrophobic PRC structure.¹⁶ A cooling
64 power of $\sim 250 \text{ W/m}^2$ has been achieved, which is much
65 higher than that of an ideal radiative cooler. However, such a
66 design has a few outstanding issues to address. Most
67 importantly, the cooling power is not optimized due to the
68 reduction of evaporative surface arising from the capping PRC
69 layer. Moreover, the structural integrity caused by a problem-
70 atic interfacial contact between the hydrated polymer (hydro-
71 gel) and the hydrophobic radiative cooling layer, especially
72 upon severe water loss, needs to be improved. Additionally, the
73 multilayer structure, which has more complex fabrication and
74 coating processes than those of a single composite, limits its
75 application.

76 Nevertheless, when both radiative cooling and evaporative
77 cooling are integrated into a single composite system, the
78 understanding becomes challenging since the light (solar)
79 propagation and liquid (water) transportation would affect
80 each other because water interacts strongly from near-IR to
81 LWIR. Thus, the design of such a composite with optimum
82 cooling power requires a fundamental understanding of the
83 interaction between evaporative and radiative cooling in the
84 same system. The optofluidic properties of such a system
85 should be carefully designed to achieve rational management
86 of light and water transport, which ideally should be adaptive
87 to varying weather conditions.

88 Herein, we achieved an ultrahigh cooling power of $\sim 350 \text{ W/}$
89 m^2 with integrated cooling strategies in a rationally designed

hydrogel. We carefully engineered the optofluidic properties of 90
a hydrogel to concurrently achieve excellent solar reflection, 91
heat isolation, LWIR emission, and water evaporation. 92
Effective subambient passive cooling (up to 6 $^{\circ}\text{C}$ below 93
ambient temperature) is achieved in tropical Singapore, while a 94
better cooling performance (up to 10 $^{\circ}\text{C}$) is observed in other 95
regions, including Illinois in the USA and Paris in France. Our 96
integrated passive cooler (IPC) maintains a stable temperature 97
adaptively regardless of the fluctuating ambient conditions, 98
attributed to the sensitive self-tuning of evaporative behavior 99
depending on incident energy. An experimental comparison 100
shows that the IPC has 6–10 times higher cooling potential 101
(power output is solar intensity dependent) in a tropical 102
climate than that of a state-of-the-art radiative cooler, while 103
consuming 200 times less water to achieve cooling perform- 104
ance comparable to that of a conventional spray cooling 105
system. Our in situ radiative cooling study under controlled 106
hydration levels and ambient humidity shed light on the 107
interaction between radiative and evaporative cooling in the 108
system. Beyond working as a single composite layer, IPC is 109
proved to have broad engineering potential, serving as a 110
functional composite privileged by its facile fabrication, 111
providing insights for next-generation passive cooler design. 112

Design of IPC with Desired Optofluidic Properties. 113
We hypothesize achieving subambient passive cooling by 114
rational integration of various strategies in a single composite 115
material through one-step formation to tackle the issues above. 116

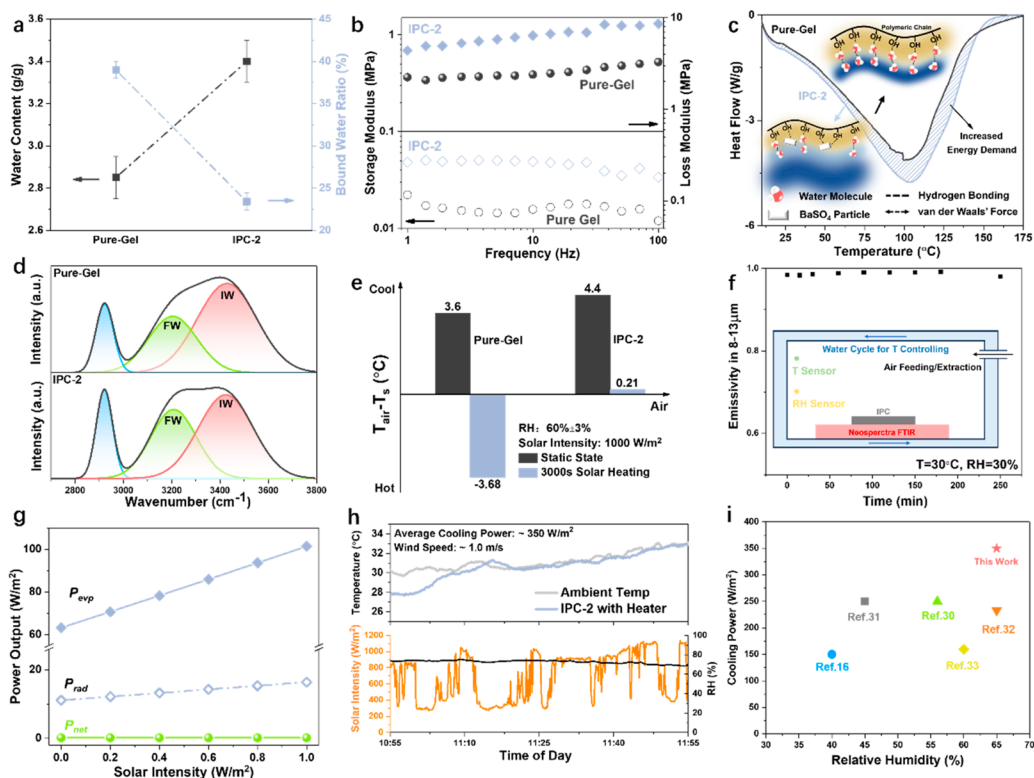


Figure 2. Passive cooling mechanisms and performance evaluation of IPC. (a) Comparison of water content and bound water ratio within Pure-Gel and IPC-2. (b) Storage and loss moduli of Pure-Gel and IPC-2. (c) DSC thermogram of water evaporation within Pure-Gel and IPC-2. Insets: schematic structures. (d) Raman OH stretching mode of Pure-Gel (upper) and IPC-2 (lower). (e) Indoor evaporative cooling comparison and temperature increase comparison (under solar illumination) between Pure-Gel and IPC-2. (f) In situ emissivity change along continuous water evaporation in a constant environment revealing the possible coexistence of radiative and evaporative cooling within an individual panel. The inset is a schematic illustration of the test bed. (g) Solar intensity dependent output power density by evaporative (P_{exp}) and radiative (P_{rad}) cooling and the power difference (P_{net}) between incident and output energies of IPC-2, which indicates the near static state and accuracy of the thermal dynamic model. (h) Experimental cooling power test of IPC-2 in a typical sunny day in Singapore (tropical climate). (i) Cooling power comparison among IPC-2 and state-of-art multifeature passive coolers under different RHs.

117 A structural media that is water maintainable (for evaporative
118 cooling), thermally isolative, solar reflective, and LWIR
119 emissive (for radiative cooling) could act as such an IPC.

120 Water has negligible ultraviolet–visible–near-infrared (UV–
121 vis-NIR) light (300–1300 nm, which accounts for ~87.5% of
122 total solar energy) absorption but partial short-wave IR
123 (1300–2500 nm) absorption in the solar spectrum.¹⁷ Thus,
124 the optical regulation in an IPC should reject most solar energy
125 similarly to a conventional PRC. To cover a tropical climate,
126 UV reflection should be enhanced compared to that of a
127 conventional PRC. The total reflection of NIR absorbance is
128 challenging; it only accounts for ~3% solar energy. With
129 complementary simultaneous evaporation cooling, NIR
130 absorption-induced heating can be well compensated. Besides,
131 the structural design should balance the water maintenance
132 and light scattering, because the stored water leads to
133 weakened light scattering due to a reduced refractive index
134 difference between water and the polymeric matrix, while
135 additive solar scattering also affects water maintenance.¹⁸ To
136 this end, a rationally designed hydrogel consisting of a porous
137 polymeric matrix that effectively regulates water storage,
138 transportation, and evaporation through multistate water
139 bonding is suitable for an IPC framework.^{19,20}

140 Water evaporation, leading to evaporative cooling, can be
141 triggered by several factors including solar irradiance, ambient
142 temperature, relative humidity, wind, etc. Atmospheric

radiation, solar illumination, and parasitic heat transfer can
143 be compensated well by evaporative cooling power generated
144 by water evaporation. Apart from the incident energy, water
145 evaporation continuously takes away internal thermal energy of
146 a passive cooler under the vapor pressure difference between
147 an IPC and the ambient environment. Such a water
148 evaporation driven by unsaturated vapor pressure is inevitable
149 as long as the relative humidity is not 100% regardless of other
150 climate conditions, leading to constant heat reduction. Most
151 important, this environmental-condition-dependent behavior
152 of water evaporation leads to an adaptive cooling performance
153 and thus a steady temperature profile.¹⁵⁴

Optical and Structural Characterizations of IPC. An
155 IPC (Experimental Section in the Supporting Information)
156 was synthesized by dispersing BaSO₄ particles (BSPs) into a
157 typical PVA hydrogel matrix. As illustrated in Figure 1a, PVA
158 polymeric chains (gray) tightly stabilize water (light blue)
159 through hydrogen bonding, where BSPs (white) within the
160 polymeric network backscatter UV–vis–NIR light (300–1300
161 nm, ~87.5% of solar energy).^{17,21} OH bending (water and
162 PVA) and SO₄ vibration endow the IPC with high IR
163 emissivity to facilitate radiative cooling. Compared to bare
164 surfaces (Figure 1b), the IPC-coated surface can effectively
165 minimize incoming solar heat gain. The active cooling demand
166 (ACD) is the energy required to maintain a constant indoor
167 temperature by an active cooling system (e.g., air conditioner),¹⁶⁸

169 which is taken as a comparison factor to evaluate cooling
170 performance.

171 A theoretical light scattering analysis suggests that UV–vis–
172 NIR light is strongly scattered by the added BSPs with
173 diameters of 0.3–0.4 μm , resulting in efficient optical
174 regulation (Figure 1c and Supplementary Note 1). Gluta-
175 raldehyde cross-links polymer chains through an acetal
176 reaction while electrostatic force between $-\text{SO}_4$ and $-\text{OH}$
177 anchors the interpenetrated BSPs (Figure 1d and Figure S2
178 and Supplementary Note 2).^{22,23} BSPs introduce strong Mie
179 scattering to UV–vis light (Figure 1e and Figure S3a–d),
180 leaving negligible transmission (Figure 1f) through IPC-2
181 (Experimental Section and Supplementary Note 3). The
182 strong visible-light backscattering leads to a white appearance
183 (Figure 1g, upper panel). The optical feature of IPC is proven
184 to be stable through a dry–wet process, where negligible
185 reflectance change is observed after full recovery through water
186 absorption (Figure S3e). Moreover, IPC-2 exhibits high LWIR
187 emissivity ($\sim 94\%$, Figure 1h) in a dry state (freeze-drying),
188 attributed to the intrinsic vibration of OH and SO_4 bonds.
189 Notably, 97% LWIR emittance benchmarks that of the state-
190 of-the-art PRC.^{24–27} Thanks to these superior parameters
191 favorable to passive cooling, IPC-2 shows outstanding
192 subambient cooling performance even without effective
193 thermal insulation under extreme climate conditions (~ 1045
194 W/m^2 , RH $\sim 60\%$), as revealed by the IR image in Figure 1g
195 (lower panel).

196 Compared to the reference of a pure hydrogel (pure-Gel),
197 IPC-2 exhibits 19% higher water content with 40% less bound
198 water, providing 51% more evaporable water (Figure 2a and
199 Figure S4 and Supplementary Note 4). Though the hydroxyl
200 groups on polymeric chains are partially occupied by bound
201 BSPs via electrostatic force (Figure S2), the expanded porous
202 matrix owing to a steric hindrance effect (SHE) arising from
203 BSPs enhances intermediate/free water storage compared to
204 pure-Gel (Figure S5). Dynamic mechanical analysis reveals
205 that IPC possesses enhanced mechanical strength (storage
206 modulus) benefiting from the interpenetrated BSPs, and the
207 increased loss modulus in turn suggests a significant SHE,
208 further indicating a balance between mechanical and swelling
209 properties (Figure 2b and Supplementary Note 5).^{19,28} It is
210 worth noting that IPC is proved to be mechanically stable
211 through repeated water cycles, where both the storage and loss
212 modulus are maintained during a dry–wet state transition
213 experiment (Figure S3f). The decreased connections among
214 polymeric chains arising from the SHE further impedes inner
215 heat transfer, leading to lowered thermal conductivity at both
216 saturated and dry states, favoring passive cooling.²⁶ (Figure S6
217 and Supplementary Note 6).²⁹ The endothermic curve (Figure
218 2c and Figure S7) indicates distinct evaporative behaviors in
219 IPC-2 compared to pure water, where the drastically
220 broadened peak suggests the presence of bound and
221 intermediate water with a stronger hydrogen bonding
222 interaction.¹⁹ The presence of BaSO_4 theoretically (Figure
223 S8 and Supplementary Note 7) leads to a stronger system
224 viscosity compared to that of the pure hydrogel, restricting the
225 dynamics of the water network. Meanwhile, BaSO_4 –water
226 hydrogen bonds have an ~ 1.5 times longer lifetime and ~ 5
227 times higher binding energy (Figure S9) compared to a water–
228 water hydrogen bond, which hinders the formation of an
229 ordered hydrogen bonding network and leads to higher
230 evaporative enthalpy. A thermal analysis (Figure S10f and
231 model details in Supplementary Note 8) clearly reveals that

higher evaporative enthalpy is more favorable to cooling since
it can also prolong the cooling time. It is worth noting that
both mass and thermal transfer are considered in the
theoretical model, which is further validated by experimental
results, proving its accuracy. The Raman OH stretching mode
shows a lower intermediate/free water (IW/FW) ratio in IPC-
2 than in Pure-Gel (Figure 2d and Figure S11 and
Supplementary Note 9), suggesting less intermediate water
content that leads to a higher evaporative enthalpy in the IPC
(Figure 2c). The distinct water states fundamentally differ-
entiate the evaporation behavior of water in IPC from that of
normal water, which leads to fine control of evaporation
behavior dynamically, resulting in a drastically increased
evaporative cooling efficiency. Specifically, with the same
amount of evaporated water, IPC-2 saves 92% ACD on a
sunny day (solar intensity, $1000 \text{ W}/\text{m}^2$; RH, 60%; wind speed,
 $\sim 1.5 \text{ m/s}$). Moreover, IPC-2 shows a 50% longer cooling cycle
than normal water, indicating a much more sustainable cooling
effect (Figure S12). Cooling performances are compared
between IPC and conventional spray cooling in Figure S13
(Supplementary Note 10). Though the surficial temperature of
bare aluminum (Al) decreased by $25 \text{ }^\circ\text{C}$ under a continuous
water spray ($13.8 \text{ mL}/(\text{m}^2 \text{ s})$), its temperature is still $\sim 2 \text{ }^\circ\text{C}$
above that of IPC-2 under high solar irradiance ($800\text{--}1000$
 W/m^2). Moreover, the fluctuating Al temperature is in obvious
contrast to the stable temperature of the IPC. Importantly, the
water consumption is >200 times higher than that of IPC-2
($\sim 0.05 \text{ mL}/(\text{m}^2 \text{ s})$). It is worth noting that the surface
temperature rose immediately after most of the water
evaporated, suggesting nonsustainable cooling by spraying
water.

IPC Cooling Capability Evaluation. The lower indoor
(to exclude radiative cooling) temperature of IPC-2 than that
of Pure-Gel (Figure 2e and Figure S14 and Supplementary
Note 11) proves the effectiveness of enhanced fluidic
properties on evaporative cooling. Apparently, 3000 sun solar
illumination ($1000 \text{ W}/\text{m}^2$) heated Pure-Gel to $3.68 \text{ }^\circ\text{C}$ above
ambient temperature. In contrast, IPC-2 remained at a
subambient temperature. Notably, the thickness of IPC is
negligible in affecting surface temperatures (Figure S15) but is
responsible for increasing the length of each cooling cycle
(Figures S16 and S17).

An in situ study was conducted to investigate the interaction
between radiation and evaporation, which helps to understand
the rationality of the multifunction surface design in the IPC. It
is clear that continuous water evaporation negligibly affects
surficial emissivity within the atmospheric window, indicating
the coexistence of excellent radiative and evaporative features
(Figure 2f). Correspondingly, a near-steady-state thermal
analysis derived net power density (sunlight and environment)
shows that the IPC-2 coating significantly reduces the ACD of
rooftop by a $>81.5\%$ to maintain a comfortable indoor
temperature ($25 \text{ }^\circ\text{C}$). Compared to the ideal PRC, IPC-2 leads
to a lower ACD when it is thicker than 6 mm and possesses the
potential to fully minimize the ACD when it gets thicker over a
long run (Figure S18a).^{24,25} Heat isolation is enhanced when
IPC thickness increases, leading to a more effective rejection of
incoming atmospheric thermal energy. Though the ACD is
reduced effectively, the cooling efficiency (the ratio of cooling
power density to thickness) starts to decrease because
thickness plays a negligible role in decreasing solar heat gain.
The intersection of cooling efficiency and reciprocal of ACD
indicates 10 mm to be an appropriate thickness for outdoor

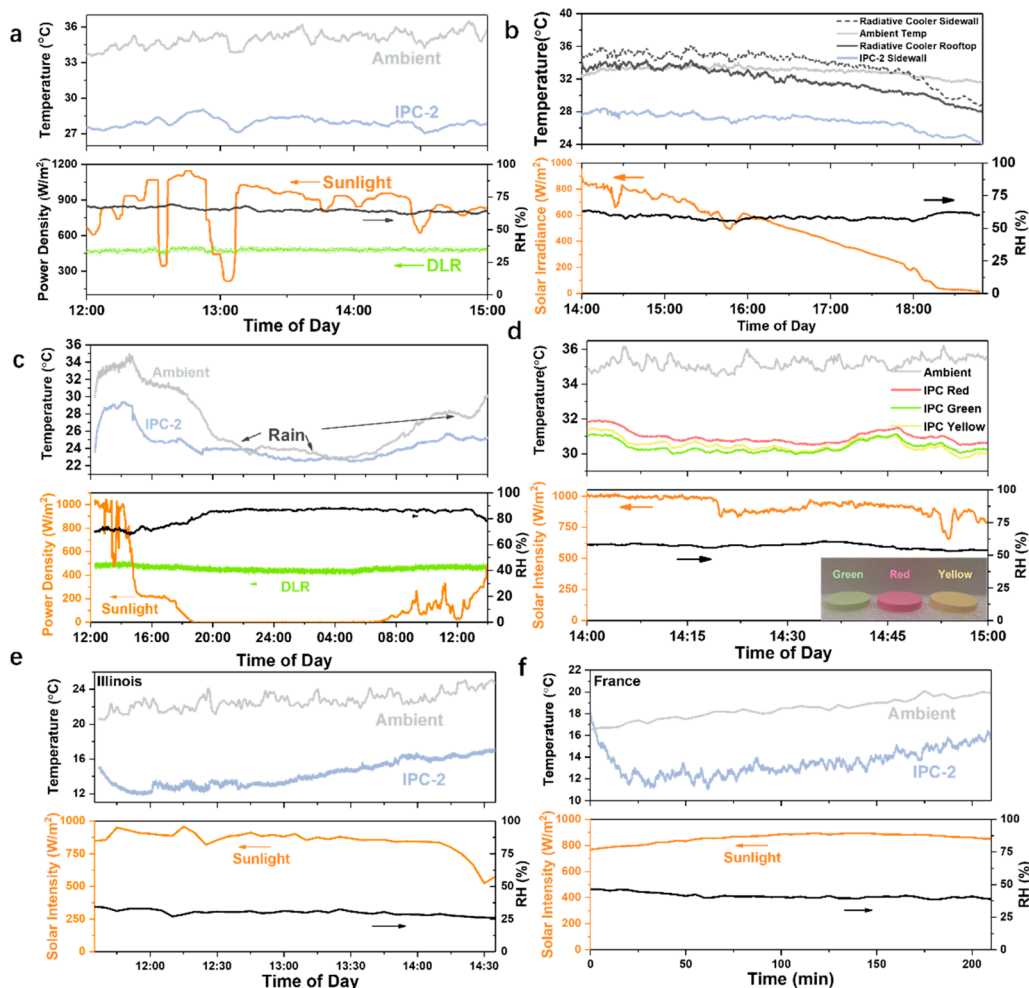


Figure 3. Cooling performances of IPC-2 under various weather conditions. (a) Outdoor cooling performance of IPC-2 in a typical sunny daytime in Singapore. (b) Outdoor cooling performance of IPC-2 compared with the state-of-the-art PRC at different faces. (c) Day-long temperature recording of ambient air and passive cooler, with no water replenishment during the test. The tested IPC-2 sample has dimensions of 25 cm (L) \times 20 cm (W) \times 4 mm (T). (d) Outdoor subambient cooling achieved by colored IPC-2 samples (lower panel inset), indicating effectiveness of the integrated cooling structure. (e) Outdoor cooling performance test of IPC-2 at University of Illinois Urbana—Champaign in Illinois, United States on May 25th, 2023. (f) Outdoor cooling performance test of IPC-2 at Sorbonne University Pierre and Marie Curie Campus in Paris, France, on May 25th, 2023.

295 cooling (under ambient conditions for Figure S18a) with
296 acceptable ACD without apparently compromising cooling
297 efficiency (Figure S18b).

298 To further evaluate the cooling capability of the IPC, the
299 theoretical energy output from the thermodynamic model is
300 provided (Figure 2g). Obviously, the evaporative energy
301 consumption varies significantly with solar irradiance, while
302 the radiative energy output slightly changes and remains at a
303 low level due to the unfavorable atmospheric hindrance (high
304 RH). Notably, $\sim 2\text{--}5\%$ solar absorbance totally compensates
305 for the radiative cooling power of the state-of-the-art radiative
306 cooler in the tropics when the solar intensity reaches $1000\text{ W}/$
307 m^2 . As such, no subambient passive cooling could be achieved
308 by LWIR radiation alone. Interestingly, the higher and stable
309 cooling power exhibited by the IPC under stronger sunlight is
310 obviously different from that of a conventional radiative cooler,
311 which indicates its high performance and adaptive cooling
312 feature. As such, the IPC automatically tunes the contributions
313 of evaporative cooling to maintain a stable temperature,
314 leading to adaptive passive cooling under various conditions
315 regardless of the varying weather or indoor/outdoor

conditions. In comparison with recent reported works, we
316 experimentally achieved a record-high cooling power of
317 approximately $350\text{ W}/\text{m}^2$ on a typical sunny day, which is
318 superior to those of state-of-the-art passive coolers with
319 various designs (Figure 2h,i).^{16,30–33} In comparison with the
320 reported multilayer structure, direct optical modification of the
321 hydrogel matrix ensured mechanical stability since an interfacial
322 adhesion problem between the radiative and evaporative layers
323 may occur when the hydrogel matrix suffers great water loss.
324 The higher cooling power of IPC-2 is attributed to a larger
325 evaporative surface without covering a layer of a radiative
326 cooler.

Cooling Performance Test. 24 h indoor cooling ($\sim 3\text{ }^\circ\text{C}$
328 below ambient temperature) was achieved with 4 mm thick
329 IPC-2 (Figure S19), in apparent contrast to a PRC that does
330 not work indoors due to a blocked atmospheric window and
331 indicated the effectiveness of evaporative cooling driven by
332 unsaturated vapor pressure. Strikingly, outdoor cooling with
333 $\sim 6\text{ }^\circ\text{C}$ below ambient temperature in the early afternoon of a
334 sunny day (solar irradiance up to $1140\text{ W}/\text{m}^2$ at RH $> 60\%$)
335 was achieved, in apparent contrast to the near-ambient 336

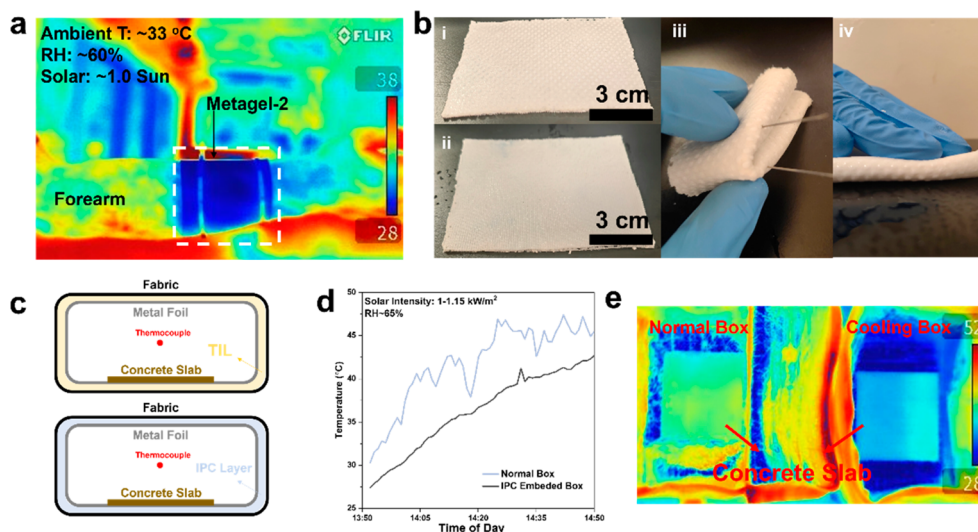


Figure 4. Demonstration of promising applications and engineering potential of IPC. (a) IR image showing outdoor subambient cooling of IPC on human skin (the same sample as that in Figure S22c). (b) Demonstration of direct composition between IPC and commercial fabric (10 cmL × 7 cmW × 4 mmT). (c) Schematic illustration of IPC as a composite cooling layer in a commercial food box. (d) Temperature recording of IPC cooling for a commercial food box. (e) IR image of concrete slabs inside two food boxes at 2:50 pm, August 4th, 2022, in Singapore (Figure 4d).

337 temperature of the state-of-the-art PRC under similar ambient
 338 conditions (Figure 3a,b and Figure S20a and Supplementary
 339 Note 12). Moreover, a greater contrastive cooling performance
 340 was demonstrated between a commercial radiative cooling
 341 paint (Nippon Solarelect Si) and the IPC, where the optical
 342 performances are quite similar between each other (com-
 343 parable solar reflectance of ~88%, Figure S20b–d).¹⁵ This
 344 directly reveals the cooling contribution from evaporative
 345 behavior. Moreover, the temperature of the cooled substrate
 346 remained very stable at 28 ± 0.5 °C, effectively avoiding the
 347 drastic temperature variation caused by fluctuating solar heat
 348 gain (Figure 3a). It is worth noting that a state-of-the-art PRC
 349 shows 2 °C higher temperature on sidewalls than that on a
 350 rooftop, which is at near-ambient temperature (Figure 3b) due
 351 to the halved radiation angle and increased incident thermal
 352 energy from the surroundings.³⁴ In contrast, IPC-2 shows 4–6
 353 °C subambient temperature on sidewalls and a rooftop, which
 354 are attributed to the nondirectional feature of evaporative
 355 behavior. It is proved that a high NIR reflective material for
 356 sidewall application leading to inevitable thermal discomfort
 357 would be alleviated by the IPC concept that partial solar
 358 energy would be directly transferred into inner energy of water
 359 vapor through a phase change process due to solar absorption
 360 of interfacial water.³⁵ Though water evaporation is strongly
 361 affected by relative humidity, nighttime subambient (3–5 °C)
 362 cooling was still achieved despite a high RH of ~80% (Figure
 363 S21). To further illustrate the adaptive cooling feature and
 364 durability of the IPC cooler, we conducted a 24 h continuous
 365 field test, during which various atmospheric conditions
 366 appeared including strong/weak sunshine, raining, cloud, and
 367 nighttime weather conditions. As shown in Figure 3c, 6 °C
 368 cooling was achieved with intense sunlight in the daytime.
 369 Even when it was raining, the passive cooler still reached a
 370 subambient temperature, and the temperature fluctuation was
 371 much smaller than that of the air temperature due to its
 372 adaptive cooling mechanism. This indicates that the IPC
 373 exhibits superior sustainable cooling performance. PRCs
 374 typically appear white to reflect the entire visible solar
 375 spectrum and colored PRCs that have broader applications

are preferred. Coloring inevitably results in increased solar heat
 gain that raises surface temperatures to above ambient.^{36–38} In
 contrast, colored IPC-2 still shows a subambient (~3.5 °C,
 Figure 3d and Figure S22) cooling performance under ~1000
 W/m² thanks to the rationally integrated cooling strategies.

Since the tropical climate is the most challenging for passive
 cooling, and our IPC cooler works well in tropical Singapore,
 we anticipate it would work equally well or better in other
 climates. To verify this, the cooling performance of the IPC
 was measured on two different continents (America and
 Europe). Recorded temperature profiles in Illinois, USA
 (Figure 3e) and Paris, France (Figure 3f) indicate a
 subambient cooling performance up to 10 °C with the IPC.
 These results suggest a generic cooling performance and
 excellent adaptivity.

Next, we further evaluated the applicable potential of the
 IPC design. IPC holds promise as cooling pads due to its
 nonhazardous nature (Figure S23a) and excellent mechanical
 flexibility and strength. 2 mm thick IPC-2 can be conformally
 attached to the skin (Figure 4a and Figure S22b–d, resulting in
 an ~28 °C skin temperature despite the high ambient
 temperature (~33 °C) under 1 sun irradiation. Moreover,
 IPC could be easily composited into a commercial fabric
 through a facile one-step cross-linking process (Figure 4b-i,ii).
 The soaked fabric filled with an IPC precursor spontaneously
 forms a porous network within the framework, holding its
 original appearance but gaining a cooling capability as an IPC.
 As shown, the composited fabric-IPC exhibits great flexibility
 and antipuncture properties (Figure 4b-iii). Thanks to the
 abundant hydrogen bonding interactions between IPC and
 fabric bones, strong adhesion is achieved with supreme
 mechanical performance, which is further confirmed by the
 lack of falling off under strong bending and pressing (Figure
 4b-iv). The demonstrated composition integrated with a
 commercial fabric provides better human comfort. Beyond
 this, we demonstrated an effective cooling performance in a
 commercial food box, where the IPC serves as an interlayer
 (replacing a thermal isolative layer) to provide a cooling
 function (Figure 4c). An obvious temperature reduction of up

415 to 7 °C was recorded under strong solar illumination outdoors,
 416 where the IR images of concrete slabs (taken when the food
 417 box is open) further prove the cooled atmosphere even in the
 418 absence of inner convection (Figure 4d,e). Additionally, the
 419 IPC shows excellent fire retardant performance (Figure S24
 420 and Supplementary Note 13) thanks to the contained water
 421 and added BSPs.³⁹ Videos 1 and 2 clearly show that the inner
 422 structure of IPC was maintained after long-term burning from
 423 both vertical and parallel directions; only a thin char layer
 424 formed at the surface. Altogether, IPC opens a route for
 425 adaptive passive cooling for various potential applications in a
 426 tropical climate and beyond (Supplementary Note 14).

427 We have successfully engineered a hydrogel-based IPC
 428 composite that provides a solution to the subambient passive
 429 cooling challenge in harsh tropical climates. Incorporating a
 430 variety of mechanisms such as high solar reflection, heat
 431 insulation, long-wave infrared (LWIR) radiation, and evapo-
 432 rative cooling, the composite delivers a remarkable cooling
 433 power of 350 W/m² and achieves 4–8 °C subambient passive
 434 cooling in tropical climates. Demonstrating excellent adapt-
 435 ability to fluctuating ambient conditions, our IPC cooler
 436 maintains stable subambient temperatures despite changing
 437 weather conditions. It exhibits superior cooling performances
 438 in climates beyond the tropics, recording up to 10 °C
 439 subambient temperatures in America and Europe. Additionally,
 440 the IPC can work as a colored passive cooler which could
 441 achieve subambient cooling under strong sunlight, which is
 442 challenging for a pure radiative cooler. Given its cost-
 443 effectiveness, adaptability, abundant choice of spectral
 444 regulation agents, and high engineering flexibility, the IPC
 445 cooler holds great promise for wide application across various
 446 climates.

447 ■ ASSOCIATED CONTENT

448 **SI** Supporting Information

449 The Supporting Information is available free of charge at
 450 <https://pubs.acs.org/doi/10.1021/acs.nanolett.3c03694>.

451 Char test of the IPC under a vertical condition (MP4)

452 Char test of the IPC under a parallel condition (MP4)

453 Details of sample preparation, theoretical simulations,
 454 and supporting experimental results (PDF)

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Author Contributions

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J.F. and H.L. conceptualized the idea of and designed
 experiments. J.F. and D.H. conducted sample fabrication and
 characterization. K.L. and S.L. conducted DFT and MD
 simulation work. D.H. and X.Z. conducted computational work
 for theoretical analysis. S.-W.K., X.W., N.L., E.N., K.Z., J.Y.T.,
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 B.F.N., L.C., T.B., and H.L. provided financial and equipment
 support on the conduction of experiments. J.F. and X.Z.
 drafted the manuscript, which was reviewed by all coauthors.
 The whole work was supervised by H.L.

Notes

The authors declare the following competing financial
 interest(s): J.F. and H.L. are inventors on a patent application
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541 ■ REFERENCES

542 (1) Goldthau, A.; Hughes, L. Protect global supply chains for low-
543 carbon technologies. *Nature* **2020**, *585*, 28–30.
544 (2) Hertwich, E. G.; Gibon, T.; Bouman, E. A.; Arvesen, A.; Suh, S.;
545 Heath, G. A.; Bergesen, J. D.; Ramirez, A.; Vega, M. I.; Shi, L.
546 Integrated life-cycle assessment of electricity-supply scenarios
547 confirms global environmental benefit of low-carbon technologies.
548 *Proc. Natl. Acad. Sci. U.S.A.* **2015**, *112* (20), 6277–6282.
549 (3) Wakerley, D.; Lamaison, S.; Ozanam, F.; Menguy, N.; Mercier,
550 D.; Marcus, P.; Fontecave, M.; Mougél, V. Bio-inspired hydro-
551 phobicity promotes CO₂ reduction on a Cu surface. *Nat. Mater.*
552 **2019**, *18* (11), 1222–1227.
553 (4) Yang, S.; Lin, X.; Lewis, W.; Suyetin, M.; Bichoutskaia, E.;
554 Parker, J. E.; Tang, C. C.; Allan, D. R.; Rizkallah, P. J.; Hubberstey, P.;
555 et al. A partially interpenetrated metal–organic framework for
556 selective hysteretic sorption of carbon dioxide. *Nat. Mater.* **2012**, *11*
557 (8), 710–716.
558 (5) Chua, K. J.; Chou, S. K.; Yang, W.; Yan, J. Achieving better
559 energy-efficient air conditioning—a review of technologies and
560 strategies. *Appl. Energy* **2013**, *104*, 87–104.
561 (6) Taleb, H. M. Using passive cooling strategies to improve thermal
562 performance and reduce energy consumption of residential buildings
563 in UAE buildings. *Front. Archit. Res.* **2014**, *3* (2), 154–165.
564 (7) Raman, A. P.; Anoma, M. A.; Zhu, L.; Rephaeli, E.; Fan, S.
565 Passive radiative cooling below ambient air temperature under direct
566 sunlight. *Nature* **2014**, *515* (7528), 540–4.
567 (8) Zeng, S.; Pian, S.; Su, M.; Wang, Z.; Wu, M.; Liu, X.; Chen, M.;
568 Xiang, Y.; Wu, J.; Zhang, M.; Cen, Q.; Tang, Y.; Zhou, X.; Huang, Z.;
569 Wang, R.; Tunuhe, A.; Sun, X.; Xia, Z.; Tian, M.; Chen, M.; Ma, X.;
570 Yang, L.; Zhou, J.; Zhou, H.; Yang, Q.; Li, X.; Ma, Y.; Tao, G.
571 Hierarchical-morphology metafabric for scalable passive daytime
572 radiative cooling. *Science* **2021**, *373* (6555), 692–696.
573 (9) Mandal, J.; Fu, Y.; Overvig, A. C.; Jia, M.; Sun, K.; Shi, N. N.;
574 Zhou, H.; Xiao, X.; Yu, N.; Yang, Y. Hierarchically porous polymer
575 coatings for highly efficient passive daytime radiative cooling. *Science*
576 **2018**, *362* (6412), 315–319.
577 (10) Zhan, Z.; Elkabbash, M.; Li, Z.; Li, X.; Zhang, J.; Rutledge, J.;
578 Singh, S.; Guo, C. Enhancing thermoelectric output power via
579 radiative cooling with nanoporous alumina. *Nano Energy* **2019**, *65*,
580 104060.
581 (11) Zhai, Y.; Ma, Y.; David, S. N.; Zhao, D.; Lou, R.; Tan, G.; Yang,
582 R.; Yin, X. Scalable-manufactured randomized glass-polymer hybrid
583 metamaterial for daytime radiative cooling. *Science* **2017**, *355* (6329),
584 1062–1066.
585 (12) Li, D.; Liu, X.; Li, W.; Lin, Z.; Zhu, B.; Li, Z.; Li, J.; Li, B.; Fan,
586 S.; Xie, J.; Zhu, J. Scalable and hierarchically designed polymer film as
587 a selective thermal emitter for high-performance all-day radiative
588 cooling. *Nat. Nanotechnol.* **2021**, *16* (2), 153–158.
589 (13) Han, D.; Ng, B. F.; Wan, M. P. Preliminary study of passive
590 radiative cooling under Singapore’s tropical climate. *Sol. Energy Mater.*
591 *Sol. Cells* **2020**, *206*, 110270.
592 (14) Kim, S.; Shang, W.; Moon, S.; Pastega, T.; Lee, E.; Luo, T.
593 High-Performance Transparent Radiative Cooler Designed by
594 Quantum Computing. *ACS Energy Lett.* **2022**, *7* (12), 4134–4141.
595 (15) Han, D.; Fei, J.; Mandal, J.; Liu, Z.; Wan, M. P.; Li, H.; Raman,
596 A. P.; Ng, B. F. Highly Reflective Polymeric Coating for Passive
597 Radiative Cooling Under Tropical Climate. *Sol. Energy Mater. Sol.*
598 *Cells* **2022**, *240*, 111723.

(16) Feng, C.; Yang, P.; Liu, H.; Mao, M.; Liu, Y.; Xue, T.; Fu, J.;
Cheng, T.; Hu, X.; Fan, H. J.; Liu, K. Bilayer porous polymer for
600 efficient passive building cooling. *Nano Energy* **2021**, *85*, 105971.
601 (17) Ustin, S. L.; Riaño, D.; Hunt, E. R. Estimating canopy water
602 content from spectroscopy. *Isr. J. Plant Sci.* **2012**, *60* (1), 9–23.
603 (18) Mandal, J.; Jia, M.; Overvig, A.; Fu, Y.; Che, E.; Yu, N.; Yang, Y.
604 Porous Polymers with Switchable Optical Transmittance for Optical
605 and Thermal Regulation. *Joule* **2019**, *3* (12), 3088–3099.
606 (19) Zhou, X.; Guo, Y.; Zhao, F.; Shi, W.; Yu, G. Topology-
607 Controlled Hydration of Polymer Network in Hydrogels for Solar-
608 Driven Wastewater Treatment. *Adv. Mater.* **2020**, *32* (52),
609 No. e2007012.
610 (20) Zhou, X.; Zhao, F.; Guo, Y.; Zhang, Y.; Yu, G. A hydrogel-
611 based antifouling solar evaporator for highly efficient water
612 desalination. *Energy Environ. Sci.* **2018**, *11* (8), 1985–1992.
613 (21) Fei, J.; Ding, B.; Koh, S. W.; Ge, J.; Wang, X.; Lee, L.; Sun, Z.;
614 Yao, M.; Chen, Y.; Gao, H.; et al. Mechanistic Investigation of
615 Electrostatic Field-Enhanced Water Evaporation. *Adv. Sci.* **2021**, *8*
616 (18), 2100875.
617 (22) Ramaswamy, V.; Vimalathithan, R.; Ponnusamy, V. Synthesis
618 and characterization of BaSO₄ nano particles using micro emulsion
619 technique. *Adv. Appl. Sci. Res.* **2010**, *1* (3), 197–204.
620 (23) El Khoury, Y.; Hellwig, P. Far infrared spectroscopy of
621 hydrogen bonding collective motions in complex molecular systems.
622 *Chem. Commun. (Camb)* **2017**, *53* (60), 8389–8399.
623 (24) Li, X.; Peoples, J.; Huang, Z.; Zhao, Z.; Qiu, J.; Ruan, X. Full
624 Daytime Sub-ambient Radiative Cooling in Commercial-like Paints
625 with High Figure of Merit. *Cell Rep. Phys. Sci.* **2020**, *1* (10), 100221.
626 (25) Wang, T.; Wu, Y.; Shi, L.; Hu, X.; Chen, M.; Wu, L. A
627 structural polymer for highly efficient all-day passive radiative cooling.
628 *Nat. Commun.* **2021**, *12* (1), 365.
629 (26) Zhong, H.; Li, Y.; Zhang, P.; Gao, S.; Liu, B.; Wang, Y.; Meng,
630 T.; Zhou, Y.; Hou, H.; Xue, C.; Zhao, Y.; Wang, Z. Hierarchically
631 Hollow Microfibers as a Scalable and Effective Thermal Insulating
632 Cooler for Buildings. *ACS Nano* **2021**, *15* (6), 10076–10083.
633 (27) Zhao, H.; Sun, Q.; Zhou, J.; Deng, X.; Cui, J. Switchable
634 Cavitation in Silicone Coatings for Energy-Saving Cooling and
635 Heating. *Adv. Mater.* **2020**, *32* (29), No. e2000870.
636 (28) Zhou, X.; Zhao, F.; Guo, Y.; Rosenberger, B.; Yu, G.
637 Architecting highly hydratable polymer networks to tune the water
638 state for solar water purification. *Sci. Adv.* **2019**, *5* (6), eaaw5484.
639 (29) Zhou, J.; Lin, S.; Zeng, H.; Liu, J.; Li, B.; Xu, Y.; Zhao, X.;
640 Chen, G. Dynamic intermolecular interactions through hydrogen
641 bonding of water promote heat conduction in hydrogels. *Mater. Horiz.*
642 **2020**, *7* (11), 2936–2943.
643 (30) Li, J.; Wang, X.; Liang, D.; Xu, N.; Zhu, B.; Li, W.; Yao, P.;
644 Jiang, Y.; Min, X.; Huang, Z.; et al. A tandem radiative/evaporative
645 cooler for weather-insensitive and high-performance daytime passive
646 cooling. *Sci. Adv.* **2022**, *8* (32), eabq0411.
647 (31) Yao, H.; Cheng, H.; Liao, Q.; Hao, X.; Zhu, K.; Hu, Y.; Qu, L.
648 Integrated radiative and evaporative cooling beyond daytime passive
649 cooling power limit. *Nano Res. Energy* **2023**, *2*, No. e9120060.
650 (32) Xu, L.; Sun, D.-W.; Tian, Y.; Fan, T.; Zhu, Z. Nanocomposite
651 hydrogel for daytime passive cooling enabled by combined effects of
652 radiative and evaporative cooling. *Chem. Eng. J.* **2023**, *457*, 141231.
653 (33) Yang, M.; Zhong, H.; Li, T.; Wu, B.; Wang, Z.; Sun, D. Phase
654 Change Material Enhanced Radiative Cooler for Temperature-
655 Adaptive Thermal Regulation. *ACS Nano* **2023**, *17* (2), 1693–1700.
656 (34) Xu, J.; Mandal, J.; Raman, A. P. Broadband directional control
657 of thermal emission. *Science* **2021**, *372* (6540), 393–397.
658 (35) Yang, J.; Wang, Z.-H.; Kaloush, K. E. Environmental impacts of
659 reflective materials: Is high albedo a ‘silver bullet’ for mitigating urban
660 heat island? *Renew. Sustain. Energy Rev.* **2015**, *47*, 830–843.
661 (36) Chen, Y.; Mandal, J.; Li, W.; Smith-Washington, A.; Tsai, C.-
662 C.; Huang, W.; Shrestha, S.; Yu, N.; Han, R. P.; Cao, A.; et al. Colored
663 and paintable bilayer coatings with high solar-infrared reflectance for
664 efficient cooling. *Sci. Adv.* **2020**, *6* (17), eaaz5413.
665 (37) Sheng, C.; An, Y.; Du, J.; Li, X. Colored Radiative Cooler under
666 Optical Tamm Resonance. *ACS Photonics* **2019**, *6* (10), 2545–2552.

- 668 (38) Lee, G. J.; Kim, Y. J.; Kim, H. M.; Yoo, Y. J.; Song, Y. M.
669 Colored, Daytime Radiative Coolers with Thin-Film Resonators for
670 Aesthetic Purposes. *Adv. Opt. Mater.* **2018**, *6* (22), 1800707.
- 671 (39) Xue, Z.; Zhang, W.; Yan, M.; Liu, J.; Wang, B.; Xia, Y. Pyrolysis
672 products and thermal degradation mechanism of intrinsically flame-
673 retardant carrageenan fiber. *RSC Adv.* **2017**, *7* (41), 25253–25264.