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Jipeng Fei, Di Han, Xuan Zhang, Ke Li, Nicolas Lavielle, et al.. Ultrahigh Passive Cooling Power in Hydrogel with Rationally Designed Optofluidic Properties. *Nano Letters*, 2023, 24 (2), pp.623-631. 10.1021/acs.nanolett.3c03694 . hal-04468285

**HAL Id: hal-04468285**

**<https://hal.science/hal-04468285v1>**

Submitted on 20 Feb 2024

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

3 Jipeng Fei,<sup>△</sup> Di Han,<sup>△</sup> Xuan Zhang,<sup>△</sup> Ke Li,<sup>△</sup> Nicolas Lavielle, Kai Zhou, Xingli Wang, Jun Yan Tan,  
4 Jianwei Zhong, Man Pun Wan, Elyes Nefzaoui, Tarik Bourouina, Shuzhou Li, Bing Feng Ng, Lili Cai,  
5 and Hong Li\*



Cite This: <https://doi.org/10.1021/acs.nanolett.3c03694>



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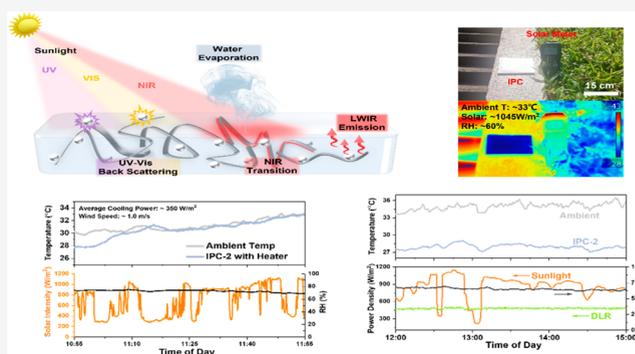
Article Recommendations



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/m<sup>2</sup> 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/m<sup>2</sup>, 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.<sup>1–4</sup> 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.<sup>5</sup> 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.<sup>6</sup> 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).<sup>7–12</sup> Daytime subambient  
43 cooling in many temperate regions is achievable when the PRC  
44 has an ultrahigh solar reflectivity ( $R_{\text{solar}}$ ) of more than 94% to  
45 greatly reduce solar heat gain.<sup>7</sup> However, effective thermal  
46 radiation to outer space in the tropical region is significantly

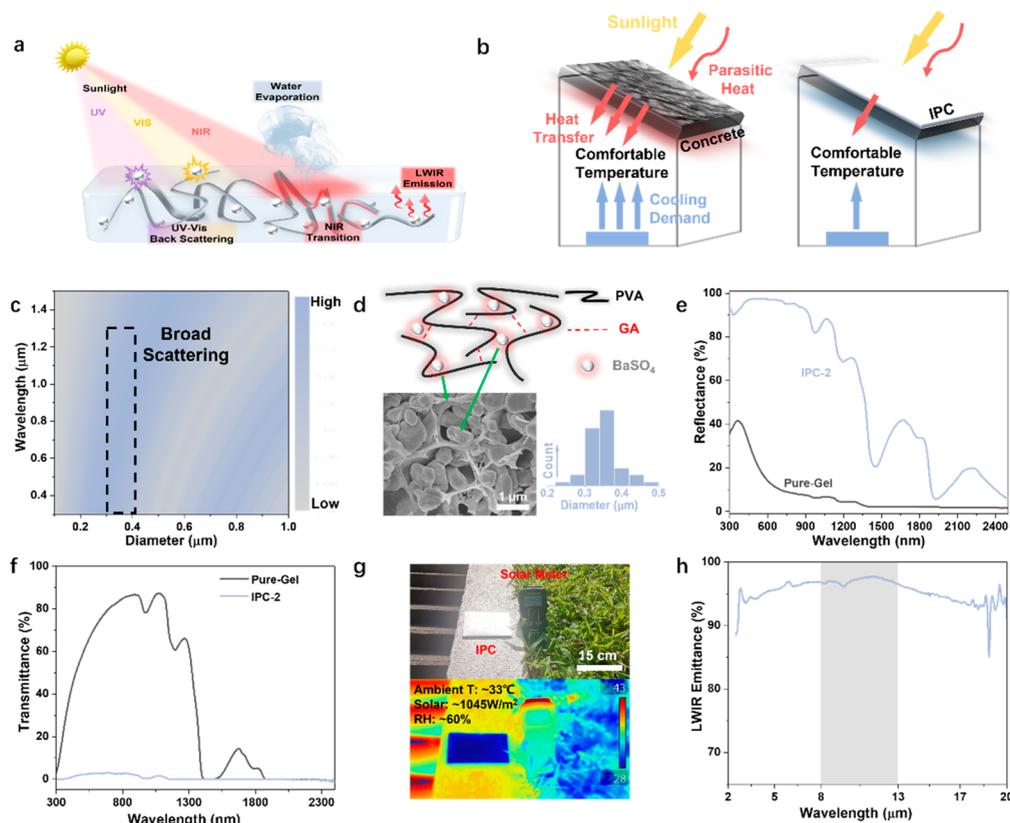
impeded by the high relative humidity (RH) and abundant  
47 thick clouds, leading to more than halved cooling power,<sup>13</sup> as  
48 evidenced by the exceptionally high downwelling radiation  
49 from the sky (up to 500 W/m<sup>2</sup>). As a result, effective cooling  
50 power through the atmospheric window is significantly lower  
51 (down to <15 W/m<sup>2</sup>) than its theoretical upper limit of ~150  
52 W/m<sup>2</sup>.<sup>14</sup> Thus far, subambient radiative cooling has hardly  
53 been achieved in a tropical climate with >1000 W/m<sup>2</sup> solar  
54 irradiance even using the best reported PRC ( $R_{\text{solar}} \approx 97\%$  and  
55 long-wavelength infrared emissivity  $E_{\text{LWIR}} \approx 96\%$ ).<sup>15</sup> 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

**Received:** September 26, 2023

**Revised:** November 20, 2023

**Accepted:** November 20, 2023



**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  $\mu\text{m}$ ) 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.

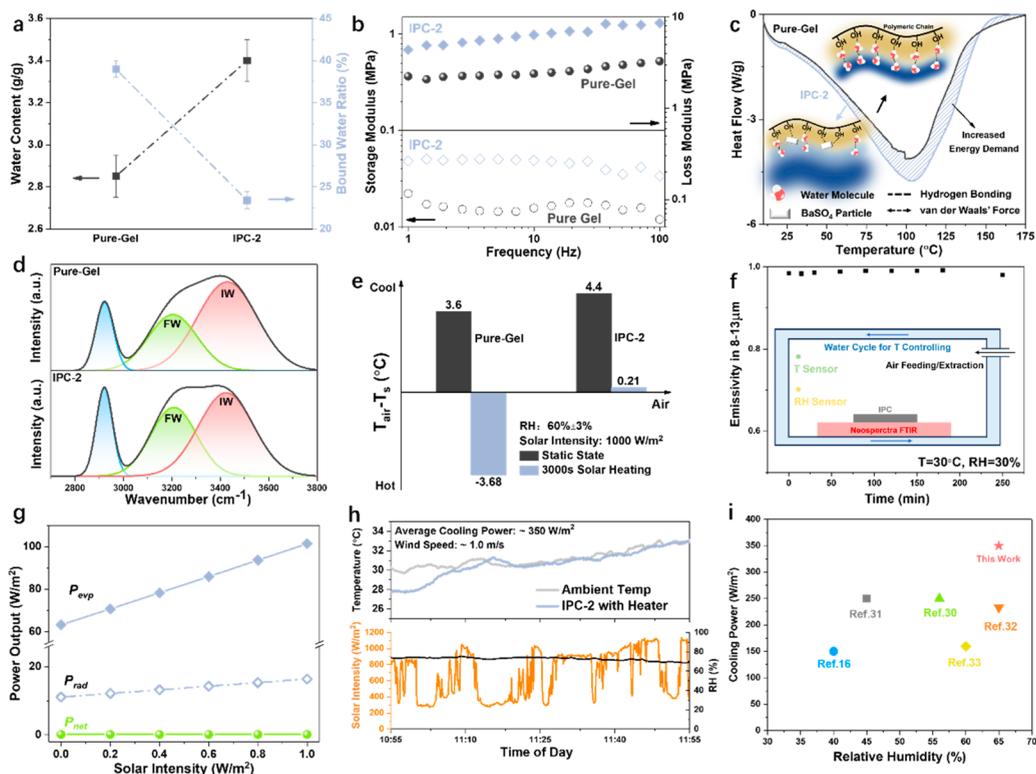
63 double-layer hydrogel–hydrophobic PRC structure.<sup>16</sup> A cooling  
64 power of  $\sim 250 \text{ W/m}^2$  has been achieved, which is much  
65 higher than that of an ideal radiative cooler. However, such a  
66 design has a few outstanding issues to address. Most  
67 importantly, the cooling 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  $\text{m}^2$  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 ( $P_{evp}$ ) 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.<sup>17</sup> 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.<sup>18</sup> 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.<sup>19,20</sup>

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.<sup>154</sup>

**Optical and Structural Characterizations of IPC.** An  
155 IPC (Experimental Section in the Supporting Information)  
156 was synthesized by dispersing BaSO<sub>4</sub> 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).<sup>17,21</sup> OH bending (water and  
162 PVA) and SO<sub>4</sub> 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),<sup>168</sup>

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

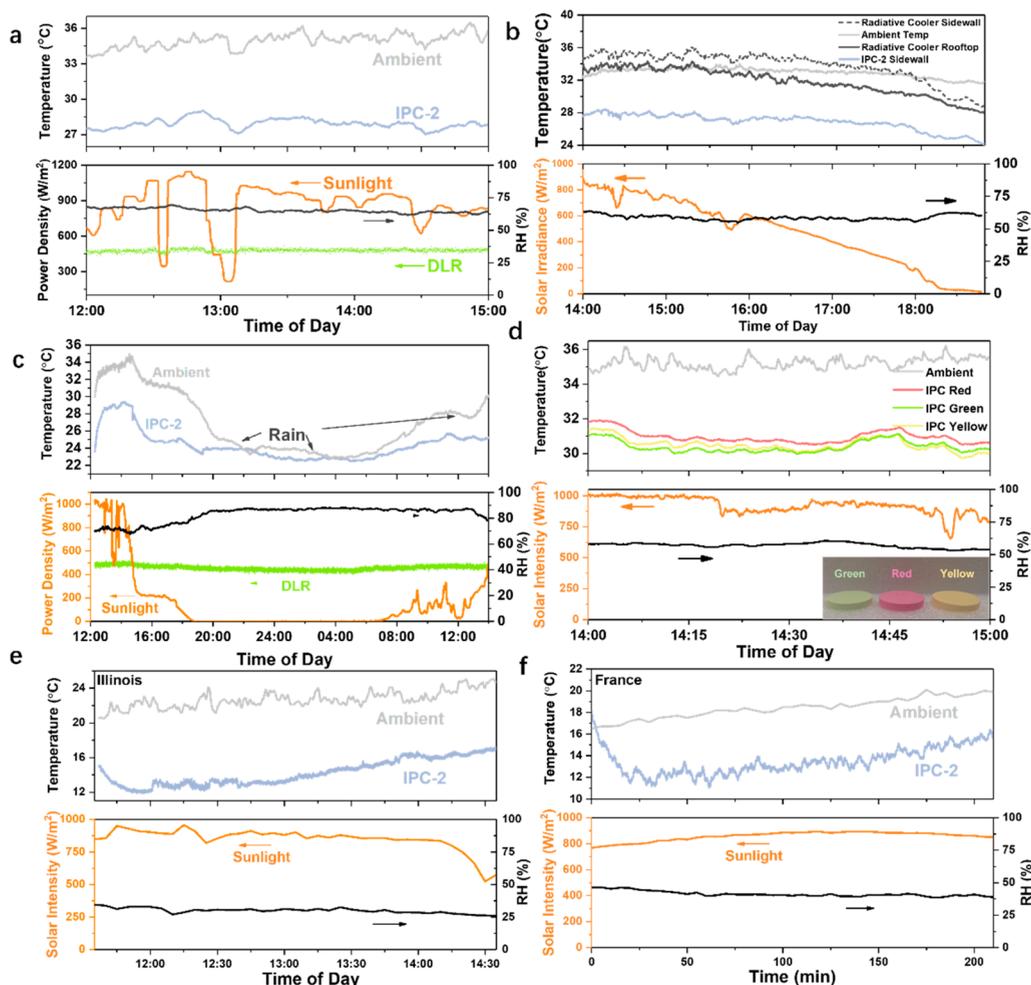
171 A theoretical light scattering analysis suggests that UV–vis–  
172 NIR light is strongly scattered by the added BSPs with  
173 diameters of 0.3–0.4  $\mu\text{m}$ , resulting in efficient optical  
174 regulation (Figure 1c and Supplementary Note 1). Gluta-  
175 raldehyde cross-links polymer chains 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).<sup>22,23</sup> 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  $\text{SO}_4$  bonds.  
189 Notably, 97% LWIR emittance benchmarks that of the state-  
190 of-the-art PRC.<sup>24–27</sup> 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 ( $\sim 1045$   
194  $\text{W}/\text{m}^2$ , 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).<sup>19,28</sup> 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.<sup>26</sup> (Figure S6  
217 and Supplementary Note 6).<sup>29</sup> 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.<sup>19</sup> The presence of  $\text{BaSO}_4$  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,  $\text{BaSO}_4$ –water  
226 hydrogen bonds have an  $\sim 1.5$  times longer lifetime and  $\sim 5$   
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 ( $800\text{--}1000$   
 $\text{W}/\text{m}^2$ ). 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).<sup>24,25</sup> 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



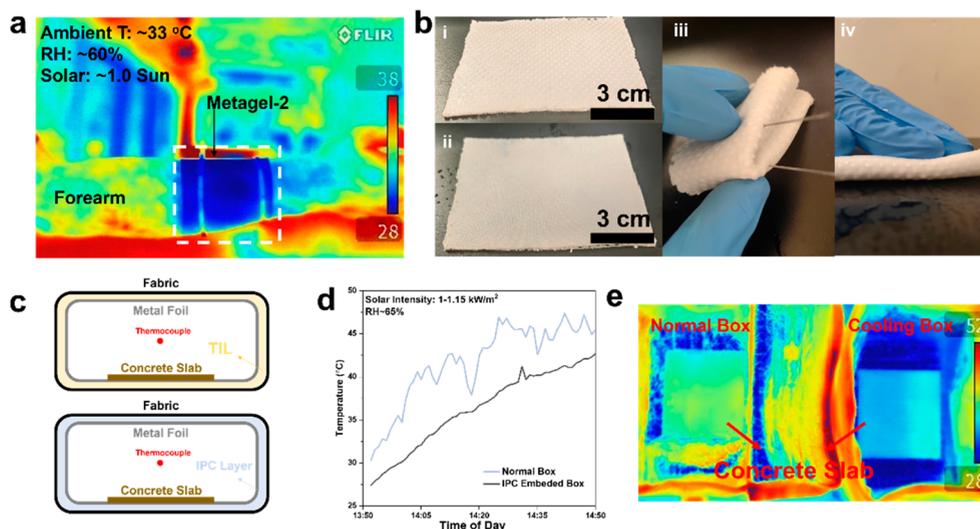
**Figure 3.** Cooling performances of IPC-2 under various weather conditions. (a) Outdoor cooling performance of IPC-2 in a typical sunny daytime in Singapore. (b) Outdoor cooling performance of IPC-2 compared with the state-of-the-art PRC at different faces. (c) Day-long temperature recording of ambient 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  $\text{m}^2$ . 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).<sup>16,30–33</sup> 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).<sup>15</sup> This  
 344 directly reveals the cooling contribution from evaporative  
 345 behavior. Moreover, the temperature of the cooled substrate  
 346 remained very stable at  $28 \pm 0.5$  °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.<sup>34</sup> 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.<sup>35</sup> 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  
 376 gain that raises surface temperatures to above ambient.<sup>36–38</sup> In  
 377 contrast, colored IPC-2 still shows a subambient (~3.5 °C,  
 378 Figure 3d and Figure S22) cooling performance under ~1000  
 379 W/m<sup>2</sup> thanks to the rationally integrated cooling strategies.  
 380

Since the tropical climate is the most challenging for passive  
 381 cooling, and our IPC cooler works well in tropical Singapore,  
 382 we anticipate it would work equally well or better in other  
 383 climates. To verify this, the cooling performance of the IPC  
 384 was measured on two different continents (America and  
 385 Europe). Recorded temperature profiles in Illinois, USA  
 386 (Figure 3e) and Paris, France (Figure 3f) indicate a  
 387 subambient cooling performance up to 10 °C with the IPC.  
 388 These results suggest a generic cooling performance and  
 389 excellent adaptivity.  
 390

Next, we further evaluated the applicable potential of the  
 391 IPC design. IPC holds promise as cooling pads due to its  
 392 nonhazardous nature (Figure S23a) and excellent mechanical  
 393 flexibility and strength. 2 mm thick IPC-2 can be conformally  
 394 attached to the skin (Figure 4a and Figure S22b–d, resulting in  
 395 an ~28 °C skin temperature despite the high ambient  
 396 temperature (~33 °C) under 1 sun irradiation. Moreover,  
 397 IPC could be easily composited into a commercial fabric  
 398 through a facile one-step cross-linking process (Figure 4b-i,ii).  
 399 The soaked fabric filled with an IPC precursor spontaneously  
 400 forms a porous network within the framework, holding its  
 401 original appearance but gaining a cooling capability as an IPC.  
 402 As shown, the composited fabric-IPC exhibits great flexibility  
 403 and antipuncture properties (Figure 4b-iii). Thanks to the  
 404 abundant hydrogen bonding interactions between IPC and  
 405 fabric bones, strong adhesion is achieved with supreme  
 406 mechanical performance, which is further confirmed by the  
 407 lack of falling off under strong bending and pressing (Figure  
 408 4b-iv). The demonstrated composition integrated with a  
 409 commercial fabric provides better human comfort. Beyond  
 410 this, we demonstrated an effective cooling performance in a  
 411 commercial food box, where the IPC serves as an interlayer  
 412 (replacing a thermal isolative layer) to provide a cooling  
 413 function (Figure 4c). An obvious temperature reduction of up  
 414

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.<sup>39</sup> 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/m<sup>2</sup> 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)

## 455 ■ AUTHOR INFORMATION

### 456 Corresponding Author

457 **Hong Li** – School of Mechanical and Aerospace Engineering  
 458 and School of Electric and Electronic Engineering, Nanyang  
 459 Technological University, Singapore 639798, Singapore;  
 460 CINTRA CNRS/NTU/THALES, UMI 3288, Singapore  
 461 637553, Singapore; [orcid.org/0000-0002-6975-7787](https://orcid.org/0000-0002-6975-7787);  
 462 Email: [ehongli@ntu.edu.sg](mailto:ehongli@ntu.edu.sg)

### 463 Authors

464 **Jipeng Fei** – School of Mechanical and Aerospace Engineering,  
 465 Nanyang Technological University, Singapore 639798,  
 466 Singapore

467 **Di Han** – School of Mechanical and Aerospace Engineering,  
 468 Nanyang Technological University, Singapore 639798,  
 469 Singapore

470 **Xuan Zhang** – Department of Energy and Power Engineering,  
 471 School of Mechanical Engineering, Beijing Institute of  
 472 Technology, Beijing 100081, People's Republic of China

**Ke Li** – Institute of Materials Research and Engineering,  
 Agency for Science, Technology and Research, Singapore  
 138634, Singapore; School of Materials Science and  
 Engineering, Nanyang Technological University, Singapore  
 639798, Singapore; [orcid.org/0000-0002-3140-3983](https://orcid.org/0000-0002-3140-3983)

**Nicolas Lavielle** – Univ Gustave Eiffel, CNRS, ESYCOM,  
 Marne la Vallée F77454, France; [orcid.org/0000-0001-5842-2992](https://orcid.org/0000-0001-5842-2992)

**Kai Zhou** – Department of Mechanical Science and  
 Engineering, University of Illinois at Urbana–Champaign,  
 Urbana, Illinois 61801, United States; [orcid.org/0000-0003-4653-6436](https://orcid.org/0000-0003-4653-6436)

**Xingli Wang** – CINTRA CNRS/NTU/THALES, UMI 3288,  
 Singapore 637553, Singapore

**Jun Yan Tan** – School of Mechanical and Aerospace  
 Engineering, Nanyang Technological University, Singapore  
 639798, Singapore

**Jianwei Zhong** – Pillar of Engineering Product Development,  
 Singapore University of Technology and Design, Singapore  
 487372, Singapore; [orcid.org/0000-0001-6198-868X](https://orcid.org/0000-0001-6198-868X)

**Man Pun Wan** – School of Mechanical and Aerospace  
 Engineering, Nanyang Technological University, Singapore  
 639798, Singapore

**Elyes Nefzaoui** – Univ Gustave Eiffel, CNRS, ESYCOM,  
 Marne la Vallée F77454, France

**Tarik Bourouina** – Univ Gustave Eiffel, CNRS, ESYCOM,  
 Marne la Vallée F77454, France; CINTRA CNRS/NTU/  
 THALES, UMI 3288, Singapore 637553, Singapore;  
[orcid.org/0000-0003-2342-7149](https://orcid.org/0000-0003-2342-7149)

**Shuzhou Li** – School of Materials Science and Engineering,  
 Nanyang Technological University, Singapore 639798,  
 Singapore; [orcid.org/0000-0002-2159-2602](https://orcid.org/0000-0002-2159-2602)

**Bing Feng Ng** – School of Mechanical and Aerospace  
 Engineering, Nanyang Technological University, Singapore  
 639798, Singapore; [orcid.org/0000-0001-8112-1151](https://orcid.org/0000-0001-8112-1151)

**Lili Cai** – Department of Mechanical Science and Engineering,  
 University of Illinois at Urbana–Champaign, Urbana,  
 Illinois 61801, United States; [orcid.org/0000-0003-1222-248X](https://orcid.org/0000-0003-1222-248X)

Complete contact information is available at:

<https://pubs.acs.org/doi/10.1021/acs.nanolett.3c03694>

### Author Contributions

△J.L. and D.H. contributed equally to this work.

### Author Contributions

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.,  
 and J.Z. helped conduct material characterizations. M.P.W.,  
 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  
 related to this work, filed by Nanyang Technological  
 University Singapore (application no. 10202113624U). The  
 authors declare no other competing interests.

## 533 ■ ACKNOWLEDGMENTS

534 This work was supported by Nanyang Technological  
535 University under an NAP award (M408050000) and the  
536 Singapore Ministry of Education Tier 1 program (RG58/21 &  
537 RG97/18 (S)). The authors acknowledge the Facility for  
538 Analysis, Characterization, Testing and Simulation (FACTS),  
539 Nanyang Technological University Singapore, for use of  
540 electron microscopy and X-ray facilities.

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