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## ▶ To cite this version:

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-04468285

Submitted on 20 Feb 2024

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

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



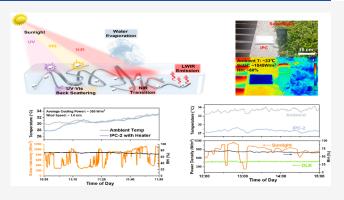
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6 **ABSTRACT:** The cooling power of a radiative cooler is more 7 than halved in the tropics, e.g., Singapore, because of its harsh 8 weather conditions including high humidity (84% on average), 9 strong downward atmospheric radiation ( $\sim$ 40% higher than 10 elsewhere), abundant rainfall, and intense solar radiation (up to 11 1200 W/m² with  $\sim$ 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) cooling 16 even under the strongest solar radiation in Singapore. The 17 integrated passive cooler achieves an ultrahigh cooling power of 18  $\sim$ 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 various climates.

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

To achieve a sustainable development, COP26 set the ambitious target of net-zero carbon emission by the middle of the century, which urgently calls for low-carbon technologies. Energy-related carbon emissions totaled 31.5 billion metric tons in 2020. Active cooling consumes a considerable amount of energy, especially in the summer season of most countries and in all seasons of tropical zones such as Singapore. For instance, cooling is responsible for over 60% of the electricity consumption in nonresidential buildings in Singapore, where 95% of the electricity comes from burning natural gas. Thus, developing an energy-saving cooling method is crucial for achieving the COP26 goal of carbon neutrality and sustainability.

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

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

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



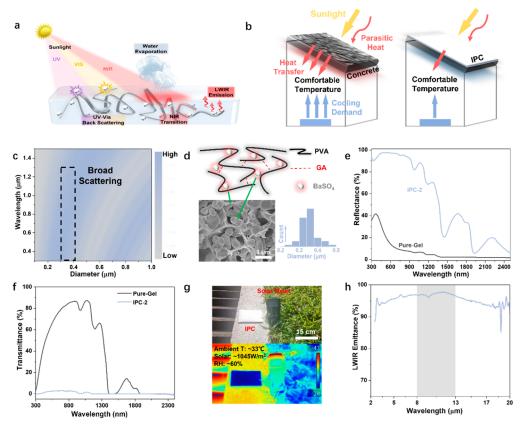


Figure 1. IPC design and characterizations. (a) Schematic illustration of cooling mechanisms. (b) Illustration of active cooling demand (ACD) difference with bare (left) and IPC-covered (right) concretes. (c) Simulated scattering efficiency (300–1300 nm) with varying BSP sizes. (d) Structural illustration (upper) and scanning electron microscopy image (lower left, scale bar is 1  $\mu$ 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. 16 A cool64 ing power of ~250 W/m² 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 problem70 atic interfacial contact between the hydrated polymer (hydro71 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.

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

Herein, we achieved an ultrahigh cooling power of  $\sim$ 350 W/ m<sup>2</sup> 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 °C below 93 ambient temperature) is achieved in tropical Singapore, while a 94 better cooling performance (up to 10 °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

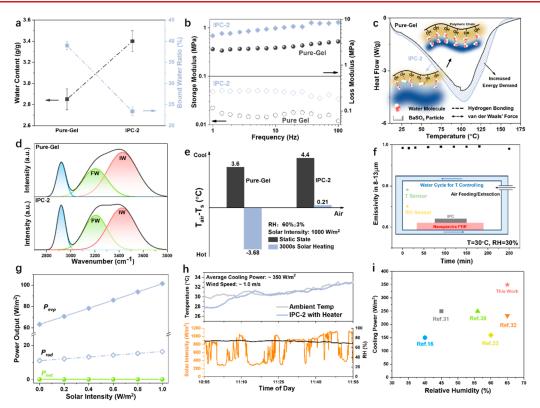


Figure 2. Passive cooling mechanisms and performance evaluation of IPC. (a) Comparison of water content and bound water ratio within Pure-Gel and IPC-2. (b) Storage and loss moduli of Pure-Gel and IPC-2. (c) DSC thermogram of water evaporation within Pure-Gel and IPC-2. Insets: schematic structures. (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  $(P_{\text{evp}})$  and radiative  $(P_{\text{rad}})$  cooling and the power difference  $(P_{\text{net}})$  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.

Water has negligible ultraviolet-visible-near-infrared (UVvis-NIR) light (300-1300 nm, which accounts for ~87.5% of total solar energy) absorption but partial short-wave IR (1300-2500 nm) absorption in the solar spectrum.<sup>17</sup> Thus, the optical regulation in an IPC should reject most solar energy similarly to a conventional PRC. To cover a tropical climate, UV reflection should be enhanced compared to that of a 127 conventional PRC. The total reflection of NIR absorbance is hallenging; it only accounts for ~3% solar energy. With omplementary simultaneous evaporation cooling, NIR bsorption-induced heating can be well compensated. Besides, the structural design should balance the water maintenance and light scattering, because the stored water leads to 132 weakened light scattering due to a reduced refractive index difference between water and the polymeric matrix, while additive solar scattering also affects water maintanence. 18 To this end, a rationally designed hydrogel consisting of a porous polymeric matrix that effectively regulates water storage, transportation, and evaporation through multistate water bonding is suitable for an IPC framework. 19,20

Water evaporation, leading to evaporative cooling, can be triggered by several factors including solar irradiance, ambient 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.

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 f1 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), 168

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

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  $\mu$ 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  $-SO_4$  and -OH177 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 (~94%, Figure 1h) in a dry state (freeze-drying), 188 attributed to the intrinsic vibration of OH and SO<sub>4</sub> 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<sup>2</sup>, RH ~60%), as revealed by the IR image in Figure 1g 195 (lower panel).

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.<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 BaSO<sub>4</sub> 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<sub>4</sub>-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 232 it can also prolong the cooling time. It is worth noting that 233 both mass and thermal transfer are considered in the 234 theoretical model, which is further validated by experimental 235 results, proving its accuracy. The Raman OH stretching mode 236 shows a lower intermediate/free water (IW/FW) ratio in IPC- 237 2 than in Pure-Gel (Figure 2d and Figure S11 and 238 Supplementary Note 9), suggesting less intermediate water 239 content that leads to a higher evaporative enthalpy in the IPC 240 (Figure 2c). The distinct water states fundamentally differ- 241 entiate the evaporation behavior of water in IPC from that of 242 normal water, which leads to fine control of evaporation 243 behavior dynamically, resulting in a drastically increased 244 evaporative cooling efficiency. Specifically, with the same 245 amount of evaporated water, IPC-2 saves 92% ACD on a 246 sunny day (solar intensity, 1000 W/m²; RH, 60%; wind speed, 247 ~1.5 m/s). Moreover, IPC-2 shows a 50% longer cooling cycle 248 than normal water, indicating a much more sustainable cooling 249 effect (Figure S12). Cooling performances are compared 250 between IPC and conventional spray cooling in Figure \$13 251 (Supplementary Note 10). Though the surficial temperature of 252 bare aluminum (Al) decreased by 25 °C under a continuous 253 water spray (13.8 mL/( $m^2$  s)), its temperature is still ~2 °C 254 above that of IPC-2 under high solar irradiance (800-1000 255 W/m<sup>2</sup>). Moreover, the fluctuating Al temperature is in obvious 256 contrast to the stable temperature of the IPC. Importantly, the 257 water consumption is >200 times higher than that of IPC-2 258 ( $\sim 0.05 \text{ mL/(m}^2 \text{ s})$ ). It is worth noting that the surface 259 temperature rose immediately after most of the water 260 evaporated, suggesting nonsustainable cooling by spraying 261

IPC Cooling Capability Evaluation. The lower indoor 263 (to exclude radiative cooling) temperature of IPC-2 than that 264 of Pure-Gel (Figure 2e and Figure S14 and Supplementary 265 Note 11) proves the effectiveness of enhanced fluidic 266 properties on evaporative cooling. Apparently, 3000 sun solar 267 illumination (1000 W/m²) heated Pure-Gel to 3.68 °C above 268 ambient temperature. In contrast, IPC-2 remained at a 269 subambient temperature. Notably, the thickness of IPC is 270 negligible in affecting surface temperatures (Figure S15) but is 271 responsible for increasing the length of each cooling cycle 272 (Figures S16 and S17).

An in situ study was conducted to investigate the interaction 274 between radiation and evaporation, which helps to understand 275 the rationality of the multifunction surface design in the IPC. It 276 is clear that continuous water evaporation negligibly affects 277 surficial emissivity within the atmospheric window, indicating 278 the coexistence of excellent radiative and evaporative features 279 (Figure 2f). Correspondingly, a near-steady-state thermal 280 analysis derived net power density (sunlight and environment) 281 shows that the IPC-2 coating significantly reduces the ACD of 282 rooftop by a >81.5% to maintain a comfortable indoor 283 temperature (25 °C). Compared to the ideal PRC, IPC-2 leads 284 to a lower ACD when it is thicker than 6 mm and possesses the 285 potential to fully minimize the ACD when it gets thicker over a 286 long run (Figure S18a).<sup>24,25</sup> Heat isolation is enhanced when 287 IPC thickness increases, leading to a more effective rejection of 288 incoming atmospheric thermal energy. Though the ACD is 289 reduced effectively, the cooling efficiency (the ratio of cooling 290 power density to thickness) starts to decrease because 291 thickness plays a negligible role in decreasing solar heat gain. 292 The intersection of cooling efficiency and reciprocal of ACD 293 indicates 10 mm to be an appropriate thickness for outdoor 294

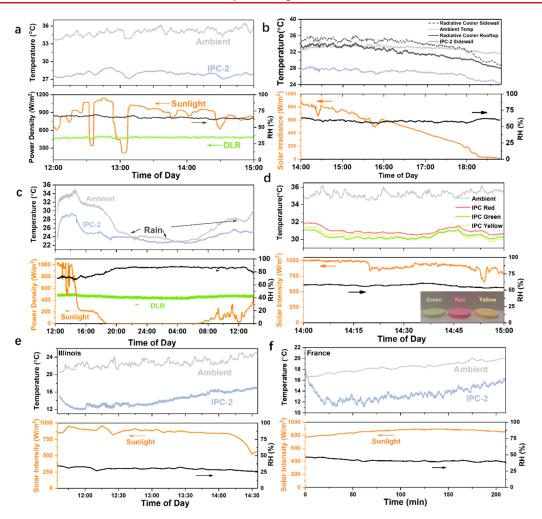


Figure 3. Cooling performances of IPC-2 under various weather conditions. (a) Outdoor cooling performance of IPC-2 in a typical sunny daytime in Singapore. (b) Outdoor cooling performance of IPC-2 compared with the state-of-the-art PRC at different faces. (c) Day-long temperature recording of ambient 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).

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 consumption varies significantly with solar irradiance, while the radiative energy output slightly changes and remains at a low level due to the unfavorable atmospheric hindrance (high 304 RH). Notably,  $\sim 2-5\%$  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 W/ 307 m<sup>2</sup>. 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 W/m² 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 a 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 °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 °C below ambient temperature in the early afternoon of a 334 sunny day (solar irradiance up to 1140 W/m² 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  $\times$  7 cmW  $\times$  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 Solareflect 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). 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. 34 In contrast, IPC-2 shows 4–6 °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.  $^{36-38}$  In  $^{377}$  contrast, colored IPC-2 still shows a subambient ( $\sim 3.5$  °C,  $^{378}$  Figure 3d and Figure S22) cooling performance under  $\sim 1000$   $^{379}$  W/m² thanks to the rationally integrated cooling strategies.

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.

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 f4 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. 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).

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 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.

## ASSOCIATED CONTENT

## **SI Supporting Information**

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

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

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

Details of sample preparation, theoretical simulations,

and supporting experimental results (PDF)

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J.F characterization. K.L. and S.L. conducted DFT and MD 519 simulation work. D.H. and X.Z. conducted computational work 520 for theoretical analysis. S.-W.K., X.W., N.L., E.N., K.Z., J.Y.T., 521 and J.Z. helped conduct material characterizations. M.P.W., 522 B.F.N., L.C., T.B., and H.L. provided financial and equipment 523 support on the conduction of experiments. J.F. and X.Z. 524 drafted the manuscript, which was reviewed by all coauthors. 525 The whole work was supervised by H.L. 526

**Notes** 527

The authors declare the following competing financial 528 interest(s): J.F. and H.L. are inventors on a patent application 529 related to this work, filed by Nanyang Technological 530 University Singapore (application no. 10202113624U). The 531 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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