## Supporting Information for "An inorganic water-based paint for highdurability passive radiative cooling"

## 1. Optical properties of coating

Measurements were taken across the wavelength range of  $0.25 - 2.5 \mu m$  using an integrating sphere and a calibrated Spectralon® reference board (Labsphere). Equation 1 displays the calculation formula for the average solar reflectance across the entire solar spectrum (250-2500nm).

$$R_{\text{solar}} = \frac{\int_{0.25}^{2.5} I_{\text{solar}}(\lambda) R(\lambda) d\lambda}{\int_{0.25}^{2.5} I_{\text{solar}}(\lambda) d\lambda}$$
(1)

Here,  $I_{solar}(\lambda)$  represents the solar intensity spectrum at air mass (AM) 1.5, and R( $\lambda$ ) denotes the spectral reflectance across the entire solar spectrum. Equations (2) and (3) present the calculation formula for the average infrared radiance within the atmospheric window (8-13µm).

$$\varepsilon_{8-13\mu\mathrm{m}} = \frac{\int_{8}^{13} I_{\mathrm{BB}}(T,\lambda)\varepsilon(T,\lambda)d\lambda}{\int_{8}^{13} I_{\mathrm{BB}}(T,\lambda)d\lambda}$$
(2)  
$$I_{\mathrm{BB}}(T,\lambda) = \frac{2hc^2}{\lambda^5} \frac{1}{e^{\lambda Tk_B} - 1}$$
(3)

Here,  $I_{BB}(T, \lambda)$  represents the spectral intensity emitted by the standard blackbody as temperature varies, with T denoting the spectral radiation temperature of the blackbody, and h being Planck's constant. Additionally,  $k_B$  represents Boltzmann's constant, c is the speed of light, and  $\lambda$  represents the wavelength. Furthermore,  $\varepsilon(T, \lambda)$  stands for the spectral emissivity of the cooler. Kirchhoff's formula defines  $\varepsilon(T, \lambda)$  as presented in Equation (4).

$$\varepsilon = 1 - T - R$$
 (4)

where T is the radiance and R is the reflectance in infrared spectroscopy.

1.2 Radiative Cooling Performance of the Coating

Calculation of the theoretical daytime radiant cooling power: the net cooling power  $P_{\text{net}}$  depends on the following formula:

$$P_{\text{net}}(T) = P_{\text{rad}}(T) - P_{\text{atm}}(T_{\text{amb}}) - P_{\text{sol}} - P_{(\text{cond+conv})}$$
(c)

Among these variables,  $P_{rad}$  represents the heat radiated power of the radiant cooler.  $P_{atm}$  denotes the absorbed energy from atmospheric thermal radiation,  $P_{sol}$  indicates the absorbed solar radiation power,  $P_{cond+conv}$  represents the combined heat transfer through conduction, convection, and radiation from the surroundings. Thermal radiation power ( $P_{rad}$ ) is defined as follows:

$$P_{\rm rad}(T) = \int d\Omega \cos\theta \int_0^\infty d\lambda I_{\rm BB} (T,\lambda) \varepsilon(\lambda,\theta)$$
$$\int d\Omega = 2\pi \int_0^{\frac{\pi}{2}} \sin\theta d\theta$$

Within this context,  $\theta$  represents the local zenith angle, while  $I_{BB}(T, \lambda)$  denotes the real-time intensity of wavelength radiation emitted by a black body at temperature T.  $\varepsilon(\lambda, \theta)$  signifies the emissivity of the material at a given wavelength  $\lambda$  and local zenith angle  $\theta$ .

Energy absorbed from atmospheric thermal radiation, denoted as  $P_{\text{atm}}$ , is calculated as follows:

$$P_{\rm atm}(T_{\rm amb}) = \int d\Omega \cos\theta \int_0^\infty d\lambda \, I_{\rm BB} \, (T_{\rm amb}, \lambda) \varepsilon(\lambda, \theta) \varepsilon_{\rm atm}(\lambda, \theta) \tag{7}$$

Here,  $T_{amb}$  represents the ambient temperature, while  $\varepsilon_{atm}(\lambda,\theta)$  signifies the atmospheric radiance at a given zenith angle  $\theta$  and wavelength  $\lambda$ .

The absorbed incident solar radiation, denoted as  $P_{sol}$ , is defined as follows:

$$P_{\rm sol} = \int_0^\infty d\lambda \,\varepsilon(\lambda, \theta_{\rm sol}) \,I_{\rm AM1.5}(\lambda) \tag{8}$$

 $I_{AM1.5}$  ( $\lambda$ ) represents the received AM1.5 standard solar spectrum irradiation intensity, while  $\theta_{sol}$  indicates the inclination angle of the sample surface facing the sun.

The heat conduction-convective radiation power of ambient radiation, denoted as  $P_{\text{cond+conv}}$ , is defined as follows:

$$P_{\text{cond+conv}}(\mathbf{T}, \mathbf{T}_{\text{amb}}) = h_{\text{cc}}(\mathbf{T}_{\text{amb}} - \mathbf{T})$$
(9)

The non-radiative heat transfer coefficient, denoted as  $h_{cc}$ , typically ranges from 0 to 12 W m<sup>-2</sup> K<sup>-1</sup> in low-humidity climates. Cooling power is significantly influenced by factors such as atmospheric transmittance, humidity, wind speed, thermal mass, and solar irradiance ( $h_{cc}$ =12 Wm<sup>-2</sup>,  $T_{amb}$ =303 K).

## 1.3 Durability test

Here, we calculate the conversion of test time between the ultraviolet aging test

chamber and outdoor lighting conditions, utilizing indicators provided by the equipment company. Outdoor lighting conditions vary not only with season and region. The time equivalent to one day of outdoor UV testing is estimated using the following formula:

$$\frac{Q_{\text{year}}}{365 \times 24} = Q_{\text{hour}}$$
$$\frac{T_{\text{set}}}{T_{\text{amb}}} \times \frac{Q_{\text{a}}}{Q_{\text{hour}}} \times 1.5 = D_{\text{outdoor}}$$

 $Q_{\text{year}}$  represents the total annual solar radiation.  $Q_{\text{hour}}$  denotes the annual average hourly solar irradiance.  $Q_{\text{a}}$  signifies the total radiation intensity set for testing purposes.  $T_{\text{set}}$  indicates the temperature set for the test.  $T_{\text{amb}}$  refers to the local average annual temperature.  $D_{\text{outdoor}}$  represents the duration of outdoor irradiation.

In our experiments, Heilongjiang Province experienced a  $Q_{\text{year}}$  of 1200 kW h<sup>-1</sup> m<sup>-2</sup>, with  $T_{\text{amb}}$  at 12 °C.  $Q_{\text{a}}$  was set to 0.7 kW m<sup>-2</sup>, and  $T_{\text{set}}$  was set to 25 °C. Upon calculation,  $D_{\text{outdoor}}$  duration was determined as 16 days. Consequently, the 72h ultraviolet irradiation test conducted in the experiment is equivalent to 48 days of outdoor irradiation.

## 2. Supplementary Figures



Figure S1 SEM and Particle size distribution of ZY-1, ZY-3 and ZY-4



Figure S2 Cross-sectional SEM picture of PRC-2-4:1



Figure S4 Theoretical radiative cooling power of  $PRC-Al_2O_3$  at different ambient temperature

 $(h_{\rm cc} = 12 \text{ W m}^{-2})$ 



Figure S5 The IR emittance of a) PRC-Al<sub>2</sub>O<sub>3</sub>, b) CIC before and after 72 h UV irradiation presented against the atmospheric transparency window (blue). c) Calculated values of emittance under different UV irradiation times.



Figure S6 a) Solar irradiance, relative humidity, and wind speed data of the outdoor test environment (Harbin, 45°43'49"N, 126°38'11"E, China, July 24th 2024, 09:00–18:00). b) Real-time temperature curves of the outdoor experiment with the pristine (i.e., no hydrophobic treatment), PRC-Al<sub>2</sub>O<sub>3</sub>, and UV-aged PRC-Al<sub>2</sub>O<sub>3</sub>.



Figure S7 a) UV–vis–NIR reflectance and b) IR emittance of PRC-Al<sub>2</sub>O<sub>3</sub> before and after hydrophobic treatment



Figure S8 Cross-cut Testing PRC-Al<sub>2</sub>O<sub>3</sub> on glass



Figure S9 Bond strength of the PRC-Al<sub>2</sub>O<sub>3</sub> and CIC



Figure S10 PRC-Al<sub>2</sub>O<sub>3</sub> acid and alkali resistance test. a) Acidic solutions treatment (pH = 3). b) Alkaline solutions treatment (pH = 10). c) Cross-cut Testing a), b) after soaking

Sample	Reflectivity (%)	Emissivity (%)	Radiative coo power (W m <sup>-2</sup> )	oling UV durability	Reference
PTFE-coated cellulose-fiber- based composites	>90	89	104	~1% reflectivity reduction after 30 days under 3 mW/cm <sup>2</sup>	[1]
AACP	92,5	97	84.9 ± 14.8	0.4% reflectivity decrease after 6 months of outdoor exposure	[2]
SiO <sub>2</sub> coating	>97	>94	/	/	[3]
TiO <sub>2</sub> +SiO <sub>2</sub>	90.7	90.11	~150	/	[4]
TiO2/ Y <sub>2</sub> O <sub>3</sub> /PDMS	92.2	94.9	/	/	[5]
PS/PDMS/PECA	96	95	/	slight degradation after 10 cycles of 8h under a UV lamp (0.89 W/m <sup>2</sup> )	[6]
FEP+K-Al <sub>2</sub> O <sub>3</sub>	96	96.3	>95.2	simple visual inspection after outdoor exposure for 7 days	[7]
PDMS/PECA/PS	>86.6	>95.3	43.2	1 month UV lamp exposure of a yellow sample	[8]
DRCC	>93	>94	134	/	[9]
HGM-FHA coating	93	94	81.76	/	[10]
SiO <sub>2</sub> -GB	96	98	104.26	significant yellowing upon UV irradiation for >10 days	[11]
FHPCM	97.2	93.5	100	/	[12]
SAC coating	92.3	95.8	/	slight degradation after 20 cycles of 8h under a UV lamp (0.89 W/m <sup>2</sup> )	[13]
PTFE/PDMS composite coating	92	93	/	>1% reflectivity decrease after 10 cycles under a UV lamp	[14]
PGEO coating	$\approx 90$	95	57.8	/	[15]
PRC-Al <sub>2</sub> O <sub>3</sub>	96.1	92.1	109	UV accelerated aging for 48 days	This work

Table S1. Comparison of the solar reflectance, IR emissivity, radiative cooling power and UV durability of PRC-Al<sub>2</sub>O<sub>3</sub> with other paint-like sprayable cooling materials reported in the literature.

[1] Y. Tian, H. Shao, X. Liu, F. Chen, Y. Li, C. Tang, Y. Zheng, Super-hydrophobic and Recyclable Cellulose-Fiber-Based Composites for High-Efficiency Passive Radiative Cooling, ACS Applied Materials & Interfaces. 13 (2021) 22521-22530.

[2] J. Song, W. Zhang, Z. Sun, M. Pan, F. Tian, X. Li, M. Ye, X. Deng, Durable radiative cooling against environmental aging, Nature Communications. 13 (2022) 4805.

[3] S. Atiganyanun, J.B. Plumley, S.J. Han, K. Hsu, J. Cytrynbaum, T.L. Peng, S.M. Han, S.E. Han, Effective Radiative Cooling by Paint-Format Microsphere-Based Photonic Random Media, ACS Photonics. 5 (2018) 1181-1187.

[4] H. Bao, C. Yan, B. Wang, X. Fang, C.Y. Zhao, X. Ruan, Double-layer nanoparticlebased coatings for efficient terrestrial radiative cooling, Solar Energy Materials and Solar Cells. 168 (2017) 78-84.

[5] T. Du, J. Niu, L. Wang, J. Bai, S. Wang, S. Li, Y. Fan, Daytime Radiative Cooling Coating Based on the Y<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> Microparticle-Embedded PDMS Polymer on Energy-Saving Buildings, ACS Applied Materials & Interfaces. 14 (2022) 51351-51360.

[6] H.-D. Wang, C.-H. Xue, Z.-Y. Ji, M.-C. Huang, Z.-H. Jiang, B.-Y. Liu, F.-Q. Deng, Q.-F. An, X.-J. Guo, Super-hydrophobic Porous Coating of Polymer Composite for Scalable and Durable Daytime Radiative Cooling, ACS Applied Materials & Interfaces. 14 (2022) 51307-51317.

[7] W. Zhou, X. Ma, M. Liu, J. Niu, S. Wang, S. Li, W. Wang, Y. Fan, Super-hydrophobic Composite Coatings Can Achieve Durability and Efficient Radiative Cooling of Energy-Saving Buildings, ACS Applied Materials & Interfaces. 16 (2024) 46703-46718.

[8] H.-D. Wang, C.-H. Xue, C.-Q. Ma, X.-X. Jin, M.-C. Huang, Y.-G. Wu, S.-Q. Lv, A.J. Chang, J. Li, X.-J. Guo, Durable and Scalable Super-hydrophobic Colored Composite Coating for Sub-ambient Daytime Radiative Cooling, ACS Sustainable Chemistry & Engineering. 12 (2024) 1681-1693.

[9] Y. Liu, X. Bu, T. Yu, X. Wang, M. He, Z. Zhang, M. Feng, Y. Zhou, Design and scalable fabrication of core-shell nanospheres embedded spectrally selective single-layer coatings for durable daytime radiative cooling, Solar Energy Materials and Solar Cells. 260 (2023) 112493.

[10] Q. Wu, Y. Cui, G. Xia, J. Yang, S. Du, X. Xiong, L. Yang, D. Xu, X. Deng, J. Cui,

Passive daytime radiative cooling coatings with renewable self-cleaning functions, Chinese Chemical Letters. 35 (2024) 108687.

[11] Y. Sun, H. He, X. Huang, Z. Guo, Super-hydrophobic SiO<sub>2</sub>–Glass Bubbles Composite Coating for Stable and Highly Efficient Daytime Radiative Cooling, ACS Applied Materials & Interfaces. 15 (2023) 4799-4813.

[12] J. Wang, J. Sun, T. Guo, H. Zhang, M. Xie, J. Yang, X. Jiang, Z. Chu, D. Liu, S. Bai, High-Strength Flexible Membrane with Rational Pore Architecture as a Selective Radiator for High-Efficiency Daytime Radiative Cooling, Advanced Materials Technologies. 7 (2022) 2100528.

[13] B.-Y. Liu, J. Wu, C.-H. Xue, Y. Zeng, J. Liang, S. Zhang, M. Liu, C.-Q. Ma, Z. Wang,G. Tao, Bioinspired Super-hydrophobic All-In-One Coating for AdaptiveThermoregulation, Advanced Materials. 36 (2024) 2400745.

[14] T. Jiang, S. Lei, F. Wang, J. Ou, W. Li, R. Dai, F. Dai, Q. Gu, W. Ni, All-Polymer Super-hydrophobic Radiative Cooling Coating Based on Polytetrafluoroethylene/Polydimethylsiloxane Composites, Industrial & Engineering Chemistry Research. 62 (2023) 5024-5034.

[15] G. Chen, Y. Wang, J. Qiu, J. Cao, Y. Zou, S. Wang, D. Jia, Y. Zhou, Robust Inorganic Daytime Radiative Cooling Coating Based on a Phosphate Geopolymer, ACS Applied Materials & Interfaces. 12 (2020) 54963-54971.