

Butterfly-inspired, nano-cement-engineered wood aerogel for superhydrophobic and flame-retardant passive daytime cooling

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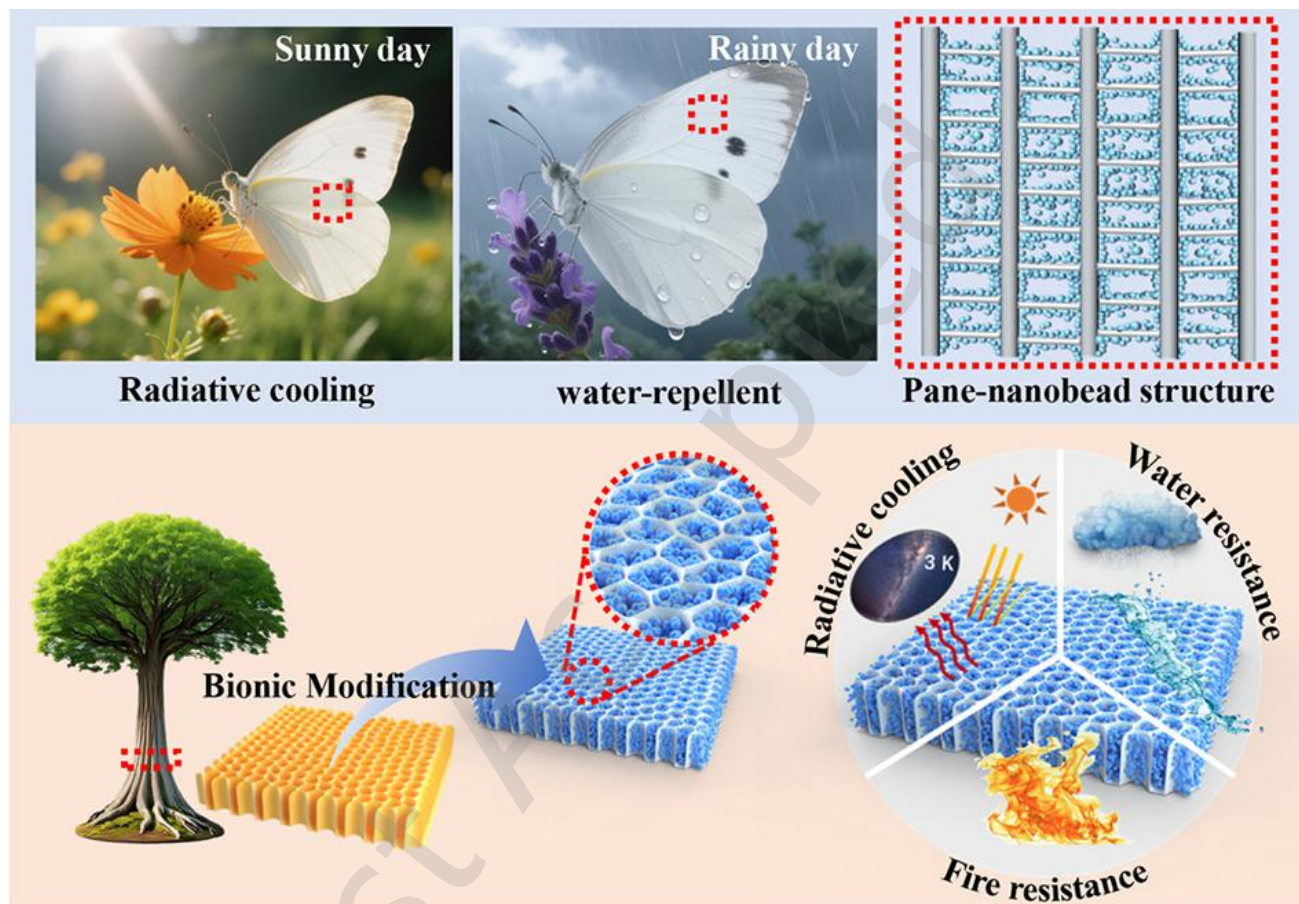
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Butterfly-Inspired, Nano-Cement-Engineered Wood Aerogel for Superhydrophobic and Flame-Retardant Passive Daytime Cooling

TABLE OF CONTENTS (TOC)



Inspired by the “pane-nanobead” structure of butterfly wings, we report a robust wood aerogel engineered with nano-cement. This biomimetic material achieves efficient passive daytime radiative cooling while overcoming wood's flammability and hydrophilicity.

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ABSTRACT: Bio-based radiative cooling materials offer a promising pathway toward sustainable and carbon-free thermal management. However, their practical application is often constrained by limited cooling efficiency, inherent hydrophilicity, and flammability. Inspired by the photonic architecture of *Pieris rapae* butterfly wings, a biomimetic wood cement aerogel featuring a "pane-nanobeads" structure was developed. This was achieved through in-situ anchoring of cement-based nanoparticles followed by fluorination modification. Benefiting from its unique bioinspired design, the resulting aerogel exhibited a high solar reflectance of 91.3% and an infrared emissivity of 99.1%, enabling a cooling temperature of 11.6 °C and a net cooling power of 84.8 W/m² under solar irradiance of 800 W/m². In hot regions of China, this performance translates to annual cooling energy savings of approximately 43.6 MJ/m². Furthermore, the material demonstrated exceptional superhydrophobicity, flame resistance, and remarkable environmental durability. Even after severe accelerated weathering involving extreme thermal cycling and intense UV irradiation, the aerogel maintained its structural integrity, superhydrophobicity, and a robust cooling drop of 9.4 °C. This work presents a scalable and effective strategy for designing high-performance, fire-safe radiative cooling materials, advancing the development of eco-friendly passive cooling technologies.

KEYWORDS: wood cement aerogel, bionic butterfly wing, radiative cooling, superhydrophobic, flame resistance

1 Instruction

The building sector accounts for nearly 50% of global energy consumption [1-3], with conventional air-conditioning cooling systems being a primary contributor [4, 5], exacerbating global warming and the increasingly frequent occurrence of extreme hot weather events [6]. Cooling-related electricity demand is projected to double or triple by 2050, further intensifying greenhouse gas emissions [7-11]. Radiative cooling, which dissipates heat through the atmospheric transparency window (8–13 μm) into cold outer space, offers a promising zero-energy pathway to reduce energy consumption [12-14]. However, achieving daytime sub-ambient radiative cooling remains profoundly challenging as intense solar absorption and downward atmospheric heat introduce significant parasitic heat transfer. Thus, effective cooling requires optimizing solar reflectance and mid-infrared emissivity synergistically [15-22].

With the development of micro-nano processing

technology, a wide variety of radiative cooling materials has been explored, including photonic/nanophotonic structures [23-28], hierarchical structures [29, 30], hybrid metamaterials [31, 32], polymer composites [33, 34], etc. Although multilayer photonic designs can achieve sub-ambient cooling through spectral selectivity, they typically rely on nanofabrication techniques such as electron-beam evaporation, vacuum deposition, or nanoimprinting, which hinder scalable production [25]. Polymeric radiative cooling materials still rely on high-reflectivity metal layers to ensure daytime cooling performance. Nonetheless, such metal layers are susceptible to corrosion and delamination, compromising long-term durability [34]. Moreover, many polymers, such as polydimethylsiloxane (PDMS), are derived from non-biodegradable precursors, raising concerns about environmental sustainability [35]. Ceramic-based cooler offered mechanical robustness but depend on high-temperature sintering [36]. Given these limitations, the development of radiative cooling materials that combine high performance, facile fabrication, green sustainability,

and low cost has emerged as an inescapable priority.

Wood, as an abundant, renewable, and lightweight biomaterial with a low-carbon lifecycle [37-40], possesses a naturally hierarchical porous structure that drives broadband solar reflection via multiple Mie scattering, while molecular vibrations of cellulose enable near-ideal mid-infrared emission [41, 45]. This convergence of structural superiority and chemical functionality establishes wood as an ideal candidate for bridging the principles of green and sustainable design with advanced cooling technologies. To date, several studies have explored wood-based radiative cooling materials [19, 43-46]. However, the existing wood-based radiative cooling materials only exhibit moderate cooling performance. Moreover, they encounter problems of performance degradation caused by rain washing and dust accumulation during practical applications. Furthermore, the complex processing, high cost, and poor fire resistance pose significant barriers to their industrial-scale implementation.

Nature's evolutionary solutions offer a blueprint for this challenge, as butterfly wings of *Pieris Rapae* have evolved a "pane-nanobeads" architecture where venation-like macrostructures and pterin pigment nanobeads synergistically optimize broadband optical scattering, band-specific absorption/emission, and superhydrophobicity to ensure long-term thermal regulation [47-50]. Coincidentally, wood stands out as an ideal biomimetic scaffold, whose cross-sectional cellular morphology exhibits a typical "pane-like" microstructure. Translating this biological design to wood by embedding functional optical nanobeads into its innate pane-like structure could overcome existing limitations, yielding a green photonic system with enhanced radiative cooling performance and long-term stability. Calcium Aluminum Silicate Hydration (C-A-S-H), a major component of cement that can be synthesized at ambient temperature, accounts for about 70 % of its volume and reduces associated carbon emissions by over 50 % [51-55]. With its abundant feedstocks, facile fabrication, and intrinsic thermal stability, C-A-S-H is an ideal candidate for constructing such nanobeads. Embedding cement-derived nanobeads within wood's cellular panes can thus mimic the butterfly wing's pane-nanobead structure. These nanobeads enhance broadband Mie scattering and boost mid-infrared emission through Si-O/Ca-O bond vibrations [56-58]. The resulting surface roughness also facilitates superhydrophobicity via straightforward fluorination. Moreover, the durable inorganic network formed by cement nanoparticles can protect wood from fire, imparting robust flame resistance. The integration of multifunctionality facilitates the practical application of radiative cooling wood.

Herein, inspired by the pane-nanobead photonic structure of butterfly wings, the multifunctional wood cement aerogels (WCAF) with "pane-nanobeads" microstructure were developed by delignification, in-situ anchoring of cement-based nanoparticles, and fluorine modification (Figure 1a, Figure S1). Benefiting from the optimized structural design and molecular vibrations of cellulose (C-O-C bonds), C-A-S-H (Si-O/Ca-O bonds), as well as the fluoroalkyl silane (C-F bonds), WCAF exhibited promising applications of radiative cooling in building sectors (Figure 1b). Figure 1c illustrates a lightweight WCAF

sample (100 mm×100 mm×10 mm), which can rest on a petal without causing damage. Notably, WCAF achieved a daytime temperature drop of 11.6 °C (Figure 1d), attributable to its excellent optical performance (solar reflectance of 91.3% and mid-infrared emissivity of 99.1%). Consequently, energy-consumption simulations confirm its superior performance, with maximum cooling energy savings reaching 43.6 MJ/m² in hot regions of China (Figure 1e). In addition, WCAF exhibited outstanding superhydrophobicity (contact angle of 154.0°±1.5°, rolling angle of 4.1°±0.6°) and flame resistance, ensuring durability and safety in practical applications. Thus, WCAF exhibited comprehensive advantages including high cooling efficiency, superhydrophobicity, flame resistance, low cost, and environmental friendliness (Figure 1f, evaluation indices are detailed in Supplementary Text 1 and Table S1). This bioinspired green photonic structure design bridges the gap between renewable wood-based systems and real-world building requirements, offering a scalable, zero-energy solution that addresses both climate mitigation and material sustainability.

2 Results and discussion

2.1 Morphology analysis

Inspired by the photonic structure of Butterfly (Figure 2a-c), the "pane-nanobead" structure of WCAF was systematically characterized via optical imaging and SEM (Figure 2d-f). Furthermore, the macroscopic and microscopic morphological evolutions of materials throughout the entire preparation process were shown in Figure S2. Specifically, optical images revealed that NW whitened distinctly after delignification, a change driven by the removal of light-absorbing lignin (Figure S2a, d). In contrast, WCA and WCAF maintained a uniform white appearance following cement incorporation and superhydrophobic modification (Figure S2g, Figure 2d), indicating negligible macroscopic morphological alterations after functionalization. Meanwhile, the hierarchical structural transitions from NW to WCAF were also highlighted by SEM micrographs. NW exhibited a well-preserved porous cellular architecture with intact cell walls (2-4 μm thickness) and interconnected lumen channels (20-30 μm, Figure S2b, c). DW retained the cellular framework but showed slight interlayer delamination and increased microporosity according to BET test (Figures. S2e, f, and S3-S4). For WCA, cement nanoparticles formed a nanostructured layer via self-assembly [59]. Intriguingly, this mimicked the photonic periodicity of butterfly wing scales (Figure S2h, i), where longitudinal ridges and transverse ribs interconnect to form a "pane-like" structure, and numerous randomly arranged pterin pigment nanoparticles were embedded within. After superhydrophobic modification, WCAF retained most cement nanoparticles and high-magnification imaging confirmed persistent anchoring of cement microspheres to cell walls (Figure 2e, f), preserving the butterfly-inspired photonic architecture intrinsic to WCA. Statistical analysis of the microstructure reveals that the pterin pigment nanoparticles on the butterfly wing scales exhibit diameters predominantly distributed between 80 and 140 nm (Figure S5a). Similarly, C-A-S-H nanobeads uniformly anchored onto the cellular wood lumen 'panes'

display a dominant diameter distribution of 0.4–0.6 μm (Figure S5b). EDS elemental mapping (Figure S6) confirmed the homogeneous distribution of Al (1.0%), Si (4.6%), and Ca (1.7%) across the cellular matrix, which verified the successful incorporation of cement nanoparticles. The other

relative element content can be observed in the bar chart.

2.2 Chemical analysis

To validate the efficacy of each sample preparation step, Fourier-transform infrared spectroscopy (FT-IR), X-ray

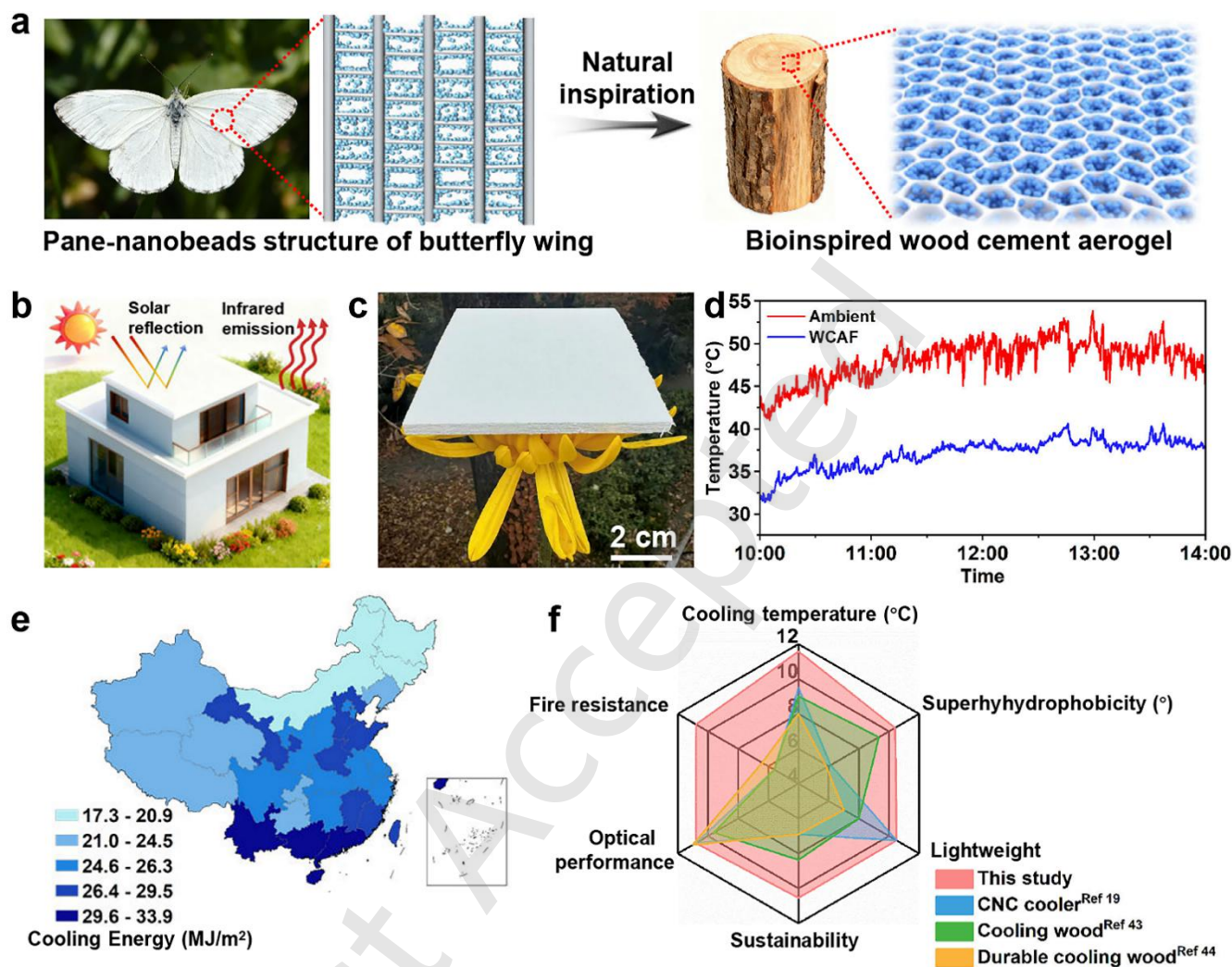


Figure 1. Design concept, comprehensive advantages of wood cement aerogel (WCAF). (a) Bioinspired design concept: inspired by pane-nanobeads photonic structure of butterfly to construct biomimetic wood cement aerogel. (b) Schematic diagram of WCAF's radiative cooling application. (c) Photograph of large-size WCAF standing on a petal. (d) Real-time temperature of ambient and WCAF when directly under solar irradiation. (e) Predicted cooling map of China when WCAF was used in simulation. (f) Advantage comparison of WCAF.

diffraction (XRD), and X-ray photoelectron spectroscopy (XPS) were employed to characterize chemical signatures and structural evolutions. FT-IR spectra revealed distinct functional group changes across samples (Figure 2a). Compared with NW, DW lacked absorption peaks at 1594 cm^{-1} and 1505 cm^{-1} , which can be attributed to the aromatic ring stretching vibrations of lignin [60]. The absorption peak bands at 1725 cm^{-1} in NW corresponded to carbonyl groups of acetyl ester in the hemicellulose and carbonyl aldehyde in the lignin, while the bands weakened significantly in DW, indicating that most of the hemicellulose and lignin had been successfully removed after delignification [61]. Furthermore, WCA and WCAF exhibited peaks corresponding to Si-O-Si (1330 cm^{-1} , 917 cm^{-1}) and Si-O-C (1150 cm^{-1}) bonds,

suggesting successful chemical interaction between the cement and the cellulose matrix [48]. After superhydrophobic treatment, WCAF showed characteristic C-F stretching vibrations at 1242 cm^{-1} and 1204 cm^{-1} , verifying the effective grafting of fluoroalkyl silane [62, 63]. A broad absorption band at 3200–3500 cm^{-1} , corresponding to O-H stretching vibrations, was observed in all samples. Notably, the intensity of this band in WCAF was significantly weaker than that in WCA, reflecting fluorinated groups partially masked surface hydroxyls while preserving the intrinsic cellulose framework. XRD analysis further supported structural transformations (Figure 2b). WCA and WCAF displayed a prominent diffraction peak at 31.2°, which matched the (110) plane of calcium silicate hydration [52].

The sharp peak observed at 45.7° can be attributed to the unreacted alite (C_3S) phase since the C-A-S-H was synthesized at room temperature. As the primary hydration product of cement, this phase confirmed the formation of C-A-S-H nanoparticles. XPS spectra (Figure 3c-i, Figure S7) complemented the chemical composition analysis. Full-scan XPS (Figure 3c) showed that NW and DW were dominated

by C and O elements. In contrast, WCA and WCAF exhibited additional peaks for Na, Ca, Si, and Al elements derived from cement. The presence of these elements corroborated the incorporation of inorganic phases, consistent with EDS results in Figure S6. WCAF uniquely showed an F 1s peak, confirming the presence of fluorinated moieties from superhydrophobic modification.

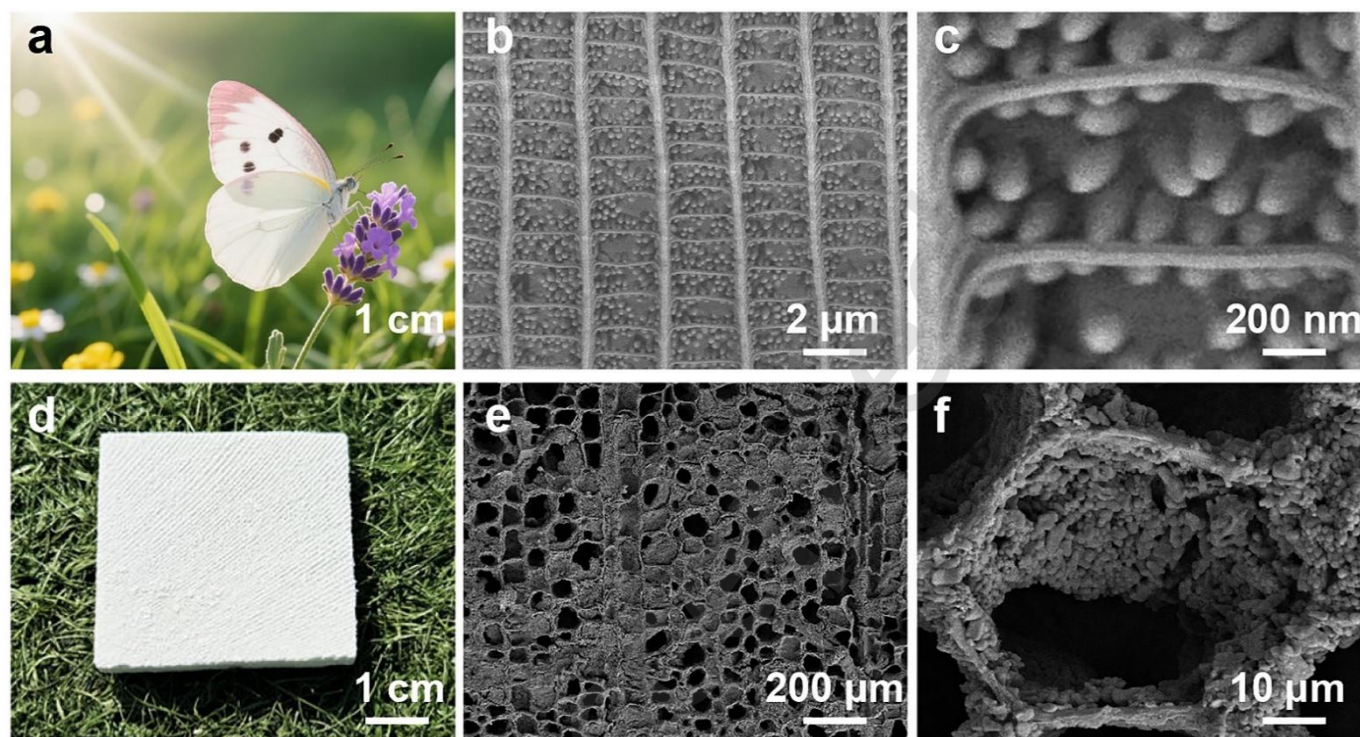


Figure 2. Optical images and microscopic features of Butterfly and WCAF. (a, d) Optical images of Butterfly and WCAF. (b-c) SEM images of butterfly wing scales at different magnifications. (e-f) SEM images of WCAF at different magnifications.

High-resolution XPS deconvolution provided insights into chemical state evolutions. In contrast, DW showed a 40.2% reduction in C1 intensity (to 47.2%) accompanied by a 212% increase in the C2 peak (C-O, 12.8% to 39.9%), reflecting lignin removal and relative enrichment of cellulose [64]. Equally, the composition analysis of NW and DW (Table S2) revealed consistent results. In WCA's high-resolution spectra (Figure 3f), the C-C/C-Si peak intensity rose to 50%, presumably due to the rich Si-O bonds in cement. Additionally, the novel detection of C-Si in WCAF's spectra (Figure 3g) validated the successful anchoring of cement nanoparticles on DW. The emergence of peaks associated with C-F groups further evidenced the successful superhydrophobization of surface. Al 2p and Ca 2p spectra (Figure 3h i, Figure S7) revealed Al-O and Ca^{2+} associated with the formation of Calcium Aluminum Silicate Hydration.

To elucidate the atomistic origins of the material's optical response, first-principles density functional theory (DFT) calculations were performed. The crystal structure used in calculations can be observed in Figure 3j. For the cellulose crystal in our aerogel (Figure 3k), the spin-up and spin-down channels are observed to be fully degenerate. This confirmed the non-magnetic nature of the material. A detailed analysis revealed an indirect band gap of 2.3 eV. The valence band maximum (VBM) is located at the A high-

symmetry point, while the conduction band minimum (CBM) is situated at the M point Brillouin zone. It was well-established that the standard PBE functional tended to underestimate the band gap of insulators. Thus, the actual quasiparticle band gap was expected to be significantly wider. This implied that the intrinsic absorption edge lied well into the ultraviolet region (<400 nm). For the cementitious component, an Al-substituted tobermorite model was constructed to simulate the C-A-S-H phase. In contrast to pure silicate chains, the incorporation of Al dopants induces distinct electronic behaviors characterized by pronounced spin-splitting and half-metallic properties within the modeled local structure. As shown in Figure 3l, the spin-up channel exhibited localized states near the Fermi level with a narrow gap of 0.0015 eV, which can be attributed to the hole states introduced by the Al substitution. Conversely, the spin-down channel retained a wide insulating gap of 4.9 eV, corresponding to deep UV absorption (approximately 253 nm). This electronic configuration suggested that the C-A-S-H phase possessed a dual optical functionality, where the wide intrinsic gap ensured broadband transparency and the doping-induced states in the conductive channel may enhanced the complex refractive index and scattering cross-section. Collectively, the distinct electronic landscapes of the insulating cellulose

matrix and the C-A-S-H network formed the fundamental basis for the superior optical scattering and radiative cooling performance of WCAF. Although the standard PBE functional tends to underestimate the absolute band gap of wide-gap insulators, it is computationally efficient and fully sufficient to qualitatively reveal the band structure dispersion trends, density of states, and intrinsic optical response mechanisms

of the complex composite matrix. Given the large supercell size and complex atomic configurations of the C-A-S-H and cellulose models, computationally demanding hybrid functionals (e.g., HSE06) were not employed, as PBE provides adequate qualitative physical insights for understanding the broad optical transparency and scattering properties.

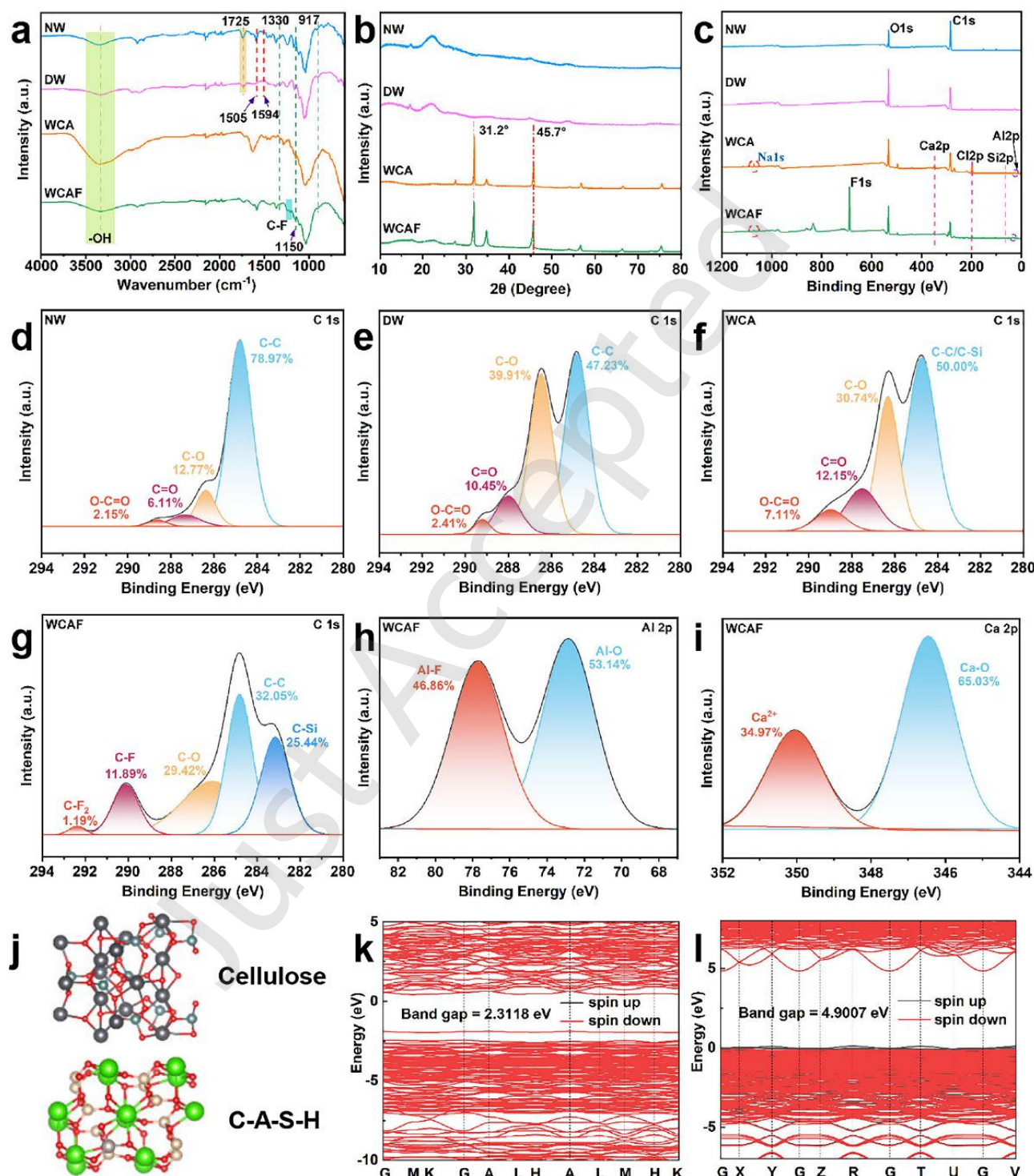


Figure 3. Chemical composition analysis of NW, DW, WCA and WCAF. (a) FT-IR spectra of NW, DW, WCA, and WCAF. (b) XRD of NW, DW, WCA, and WCAF. (c) XPS total spectrum of NW, DW, WCA, and WCAF. (d-g) C 1s high-resolution spectrum of NW, DW, WCA, and WCAF. (h) Al 2p high-resolution spectrum of WCAF. (i) Ca 2p high-resolution spectrum of NW, DW, WCA, and WCAF. (j) Crystal structure of cellulose and C-A-S-H. (k, l) Calculated band structures of cellulose and C-A-S-H.

2.3 Daytime Radiative Cooling Performance

To evaluate the cooling performance of NW, DW, WCA, and WCAF, a cooling experiment was conducted on a sunny day (25th March 2025) in Nanjing (118.8°E, 32°N). As depicted in Figure S8, the polystyrene foam was completely wrapped by aluminum foils to prevent it from being heated by direct solar irradiance. Samples were placed in the cut-out cavities, where thermocouples were installed to monitor ambient and sample temperatures. The environmental parameters of this cooling test, including an average solar irradiance of 779.6 W/m², wind speed of 0.8 m/s, and relative humidity of 26.5% (Figure S9). The radiative cooling capacity of each sample was demonstrated based on the cooling temperature difference (T_{sample} , T_{ambient} , and $T_{\text{sample}} - T_{\text{ambient}}$). For all cooling measurements, the thermocouple recording T_{ambient} was placed inside the PE-covered insulated cavity at the edge above the sample, so the reference temperature reflects the local microenvironment within the test chamber rather than the external meteorological ambient temperature. As can be seen from Figure 4a, b, the temperature of NW gradually rose with increasing solar irradiance and eventually exceeding the ambient temperature. While benefiting from the removal of lignin, DW exhibited remarkably enhanced cooling performance, with the maximum cooling temperature reaching 7.1 °C. Following the incorporation of cement nanoparticles and fluorination, WCA and WCAF demonstrated further improvements, with the maximum cooling temperature reaching 10.2 °C. This is primarily attributed to the enhancement of Mie scattering and mid-infrared emissivity by cement nanoparticles [66]. Notably, after superhydrophobic treatment, the cooling curves of the two samples are almost identical. To quantify cooling performance under peak solar irradiance (10:00-13:00), the average temperature difference was calculated (Figure 4c). Results demonstrated that NW had no cooling effect and was even 2.3 °C above the ambient temperature, mainly due to the absorption of sunlight by lignin and its low solar reflectivity (65.1%). WCA demonstrated the highest radiative cooling capacity with an average cooling temperature difference of 8.9 °C, followed closely by WCAF (8.5 °C) and DW (3.4 °C). The cooling effect of WCAF was marginally inferior to WCA, presumably due to the loss of cement nanoparticles during superhydrophobic treatment. From spectral data, WCAF showed 91.3% solar reflectivity and 99.1% atmospheric window emissivity, slightly lower than WCA's 91.6% and 99.2% (Figure 4d, e). This suggested that the C-F bond emissivity did not fully compensate for the optical losses caused by nanoparticle reduction. In addition, car models covered with samples were positioned outdoors under direct solar irradiance to evaluate their cooling capability (Figure S10). The superior optical performance of WCAF was quantitatively governed by this biomimetic hierarchical architecture, as corroborated by numerical Mie scattering simulations (Figure S11). Theoretical calculations revealed that spherical C-A-S-H nanobeads with diameters of 0.4-0.6 μm achieved maximum scattering efficiencies within the visible regime, directly underpinning the ultrahigh visible reflectance of 97.1% in WCAF. Concurrently, optical

simulations on the cellular wood lumens (20-30 μm, Figure S11a) demonstrated high scattering efficiencies across the near-infrared band, functioning as microscale reflection cavities that prolong optical propagation paths via multiple internal reflections. This multi-scale optical coupling synergistically enlarges the backward scattering cross-section across the full solar spectrum, boosting the integrated solar reflectance from 76.4% in DW to 91.3% in WCAF. Thermal imaging and temperature curve analysis confirmed that the cooling effect of samples matched the previous results, achieving up to 7.4 °C interior temperature reduction. For further quantitative characterization of the cooling capacity, the cooling power was calculated under 800 W/m² solar irradiance (Figure 4f). The cooling power of WCAF was calculated to be 84.8 W/m² and it was estimated that WCAF could achieve a temperature drop of 10.1 °C with nonradiative heat coefficient of 6 W·m⁻²·K⁻¹, indicating efficient thermal management under convective conditions. To simulate real-world operating conditions, the PE shield was removed to expose samples to natural convection. Environmental parameters were also continuously recorded (Figure S12), yielding an average solar irradiance of 816.4 W/m², wind speed of 0.8 m/s, and relative humidity of 31.8%. The temperature curves (Figure 4g) revealed that NW in this experiment surprisingly achieved daytime cooling effect, primarily attributed to the high mid-infrared emissivity of cellulose and lignin. DW, WCA, and WCAF maintained superior efficiency, with maximum temperature drops exceeding 13 °C (Figure 4h). During the peak solar period (11:00-13:00), WCA and WCAF achieved average temperature drops of 11.9 °C and 11.6 °C, respectively (Figure 4i). This confirmed that the superhydrophobic modification did not compromise the material's radiative cooling efficiency, as the cooling performance of both samples remained nearly identical.

The outstanding cooling performance of WCAF stems from its rational biomimetic hierarchical design of structure (Figure 4j, k, l), which synergistically optimizes optical scattering and mid-infrared emissivity. Achieving daytime cooling required that solar absorption be outweighed by radiative heat dissipation, which means reflecting most of the sunlight. In our designed "pane-nanobeads" structure, the macro-scale wood cellular panes and constructed nano-scale cement nanoparticles significantly amplified the broadband Mie scattering. Chemically, the 2.4 eV indirect band gap of WCAF ensured minimal absorption of solar energy above 509 nm (Visible-NIR range), which is crucial for maximizing solar reflectance and passive radiative cooling performance. Simultaneously, the rich chemical bonding network including Si-O, Ca-O, C-O-C, C-C and C-F stretching vibrations collectively contributed to the 99.1% mid-IR emissivity of WCAF within the atmospheric window (8-13 μm). Interestingly, the low thermal conductivity (0.112 W/m·K ± 0.005 W/m·K) may also play a role, considering the environmental thermal convection and thermal conduction. Beyond surface optical properties, the bulk thermal transport characteristics play an equally vital role in determining practical radiative cooling efficiency. Benefiting from the oriented cellular channels and the

porous inorganic nanobead network, WCAF possesses an intrinsically low thermal conductivity of $0.112 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. At a thickness of 5 mm, the aerogel functions as a robust thermal barrier that effectively suppresses parasitic conductive and convective heat flux from hot underlying substrates and ambient air. This decoupling prevents upward thermal leakage from counteracting the surface radiative dissipation, thereby sustaining a pronounced temperature gradient and substantially lowering the bottom-side temperature for enhanced building thermal

management. In theory, as shown in Equation (7) of the Supporting Information, the low thermal conductivity of the material notably reduced the non-radiative heat transfer coefficient hc . Theoretically, a lower thermal conductivity minimizes non-radiative cooling power, which in turn enhances the net cooling power. Accordingly, materials with low thermal conductivity delivered more prominent radiative cooling efficacy.

A comparison between the two testing configurations revealed that all samples exhibited superior cooling

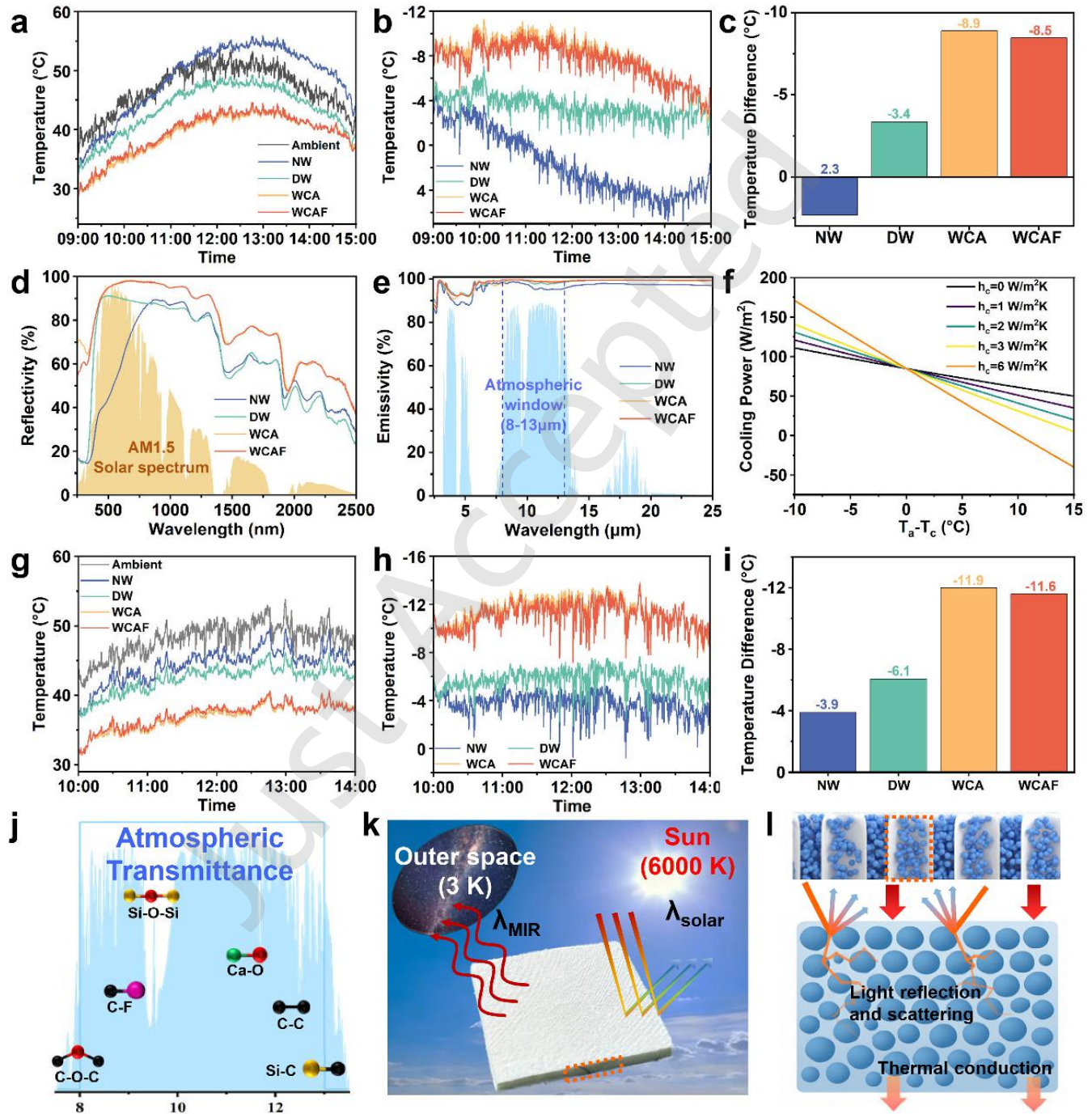


Figure 4. The schematic illustration and outdoor cooling performance of WCAF. (a) Real-time temperature of samples. (b) Temperature differences between samples and ambient. (c) Average temperature difference between sample and ambient from 10:00 to 13:00. (d) Solar reflectance of NW, DW, WCA, and WCAF. (e) Infrared emissivity of NW, DW, WCA, and WCAF. (f) Cooling power calculations during daytime under solar irradiance of 800 W/m^2 . (g) Real-time temperature of samples under the test without PE film. (h) Temperature differences between samples and ambient under the test without PE film. (i) Average temperature differences between samples and ambient from 11:00 to 13:00 under the test without PE film. (j) Vibrations of bonds in WCAF within atmospheric window (8-13 μm). (k) Schematic illustration of radiative cooling mechanism. (l) Multiple

light scattering and low thermal conductivity properties of WCAF.

performance without the PE film, highlighting the complexity of real-world thermal dynamics. The setup covered with PE film exacerbated the greenhouse effect, drastically reducing heat exchange with the external environment, even the temperature inside was significantly higher than the outside and forcing cooling to rely entirely on the material's radiative properties. The PE film's high transmittance in the atmospheric transparency window (82.3–90%) minimized interference with the test material's infrared emission, making the cooling effect nearly equivalent to pure radiative cooling [65]. However, even the high-transmittance PE film may partially influence the material's actual radiative capacity through the atmospheric window, which implied that the radiative capacity of samples was constrained. By contrast, in the cooling test without PE film, the temperature inside the cavities still rose rapidly and exceeded the temperature outside because of the direct solar irradiation. This temperature difference would amplify wind-driven convection, where the wind removes heat from the sample surface to achieve a cooling effect. Specifically, convection provided an additional heat dissipation pathway alongside radiative cooling, allowing the material to shed heat through both infrared emission to the atmosphere and wind-driven thermal exchange. This dual-channel heat dissipation significantly enhanced the overall cooling performance, demonstrating that real-world thermal dynamics leverage both radiative and convective mechanisms for optimal efficiency.

2.4 Flame resistance performance and superhydrophobic behavior

Cone calorimetry (CONE) stands as a pivotal technique for evaluating material flame retardancy in laboratory settings [67], as it recapitulates real-world fire scenarios by simultaneously yielding a suite of combustion parameters. The NW exhibited an immediate peak in heat release rate (HRR) after ignition (Figure 5a), with the highest peak heat release rate (PHRR) reaching 135.3 kW/m². DW showed the second PHRR of 121.6 kW/m², followed by WCAF with PHRR of 73.4 kW/m² and WCA with PHRR of 32.3 kW/m² (Figure 5b), which was reduced by 76.1% than that of NW. Despite the occurrence of an HRR peak, the total heat release (THR) of WCAF was only 6.8 MJ/m², which was remarkably close to that of WCA at 6.6 MJ/m². This suggested that although WCAF exhibited an initial spike in heat release rate, the cumulative heat emission over the combustion process remained nearly equivalent to WCA, indicating similar overall fire energy release characteristics between the two materials. Additionally, the smoke generation during combustion emerges as a critical metric in evaluating the fire resistance of materials, given that smoke inhalation accounts for most of fire-related fatalities (Figure 5c, d) [68–69]. The smoke production rate of NW correlated well with its HRR, featuring a maximum SPR of 0.01 m²/s and a total smoke production of 3.8 m². However, DW exhibited a discrepancy between SPR and HRR. This phenomenon arises because delignification process rendered the material highly porous and flammable, leading to rapid burnout that curtailed the duration of smoke generation. Interestingly, although WCA

remained unignited throughout the test and maintained a low smoke production rate (SPR), its total smoke production (TSP) surprisingly surpassed that of WCAF (0.5 m²>0.3 m²). This counterintuitive result can be explained that even without ignition, WCA underwent smoldering that accumulated smoke over time, as its intact inorganic network suppressed

combustion but allowed gradual release of volatile components. Besides, the structure of residues after cone calorimetry testing revealed distinct flame retardancy mechanisms (Figure S13). NW showed sparse and loosely structured residues, indicating rapid combustion, which aligned with its high HRR and consistent SPR. In contrast, DW exhibited powdery residues with a thin, discontinuous char layer, attributed to lignin removal that enhanced ignitability and accelerated combustion, thus limiting smoke accumulation despite inconsistent SPR and HRR. The residuals of WCA after cone calorimetry formed a dense char layer on its surface, a critical indicator of its flame retardancy mechanism. The inorganic network formed by cement nanoparticles constructed a thermal protective layer, which acted as an oxygen barrier to suppress combustion. By contrast, WCAF exhibited even more intact structural residues, presumably because the superhydrophobic layer is rapidly ignited, prompting the inorganic network to activate and protect the internal wood architecture.

To further observe their combustion behaviors, flame exposure tests were conducted on NW, DW, WCA, and WCAF (Figure S14). When subjected to the outer flame of an alcohol lamp, NW exhibited instantaneous ignition, with the flame propagating rapidly across the entire sample. Notably, the combustion persisted unabated even after removing the heat source, culminating in complete carbonization. Similarly, DW demonstrated immediate ignition, characterized by even faster flame front progression and more thorough fuel consumption. This intensified combustion was attributed to its lignin-depleted structure, which possessed higher porosity and specific surface area (Figure S3, S4). WCA and WCAF showed no ignition after 30 s of flame exposure in stark

contrast to NW and DW, with charring confined to the flame front. Although minor charring was observed strictly at the flame contact zone, any residual surface flame self-extinguished immediately upon removal of the external heat source, preserving the structural integrity of the samples. The inorganic network formed by cement nanoparticles played a critical role here because it inhibited ignition and flame propagation while acting as a protective layer to maintain the oriented cellulose macromolecular chains. This network effectively blocked heat transfer and oxygen diffusion, preventing the underlying material from reaching its ignition point.

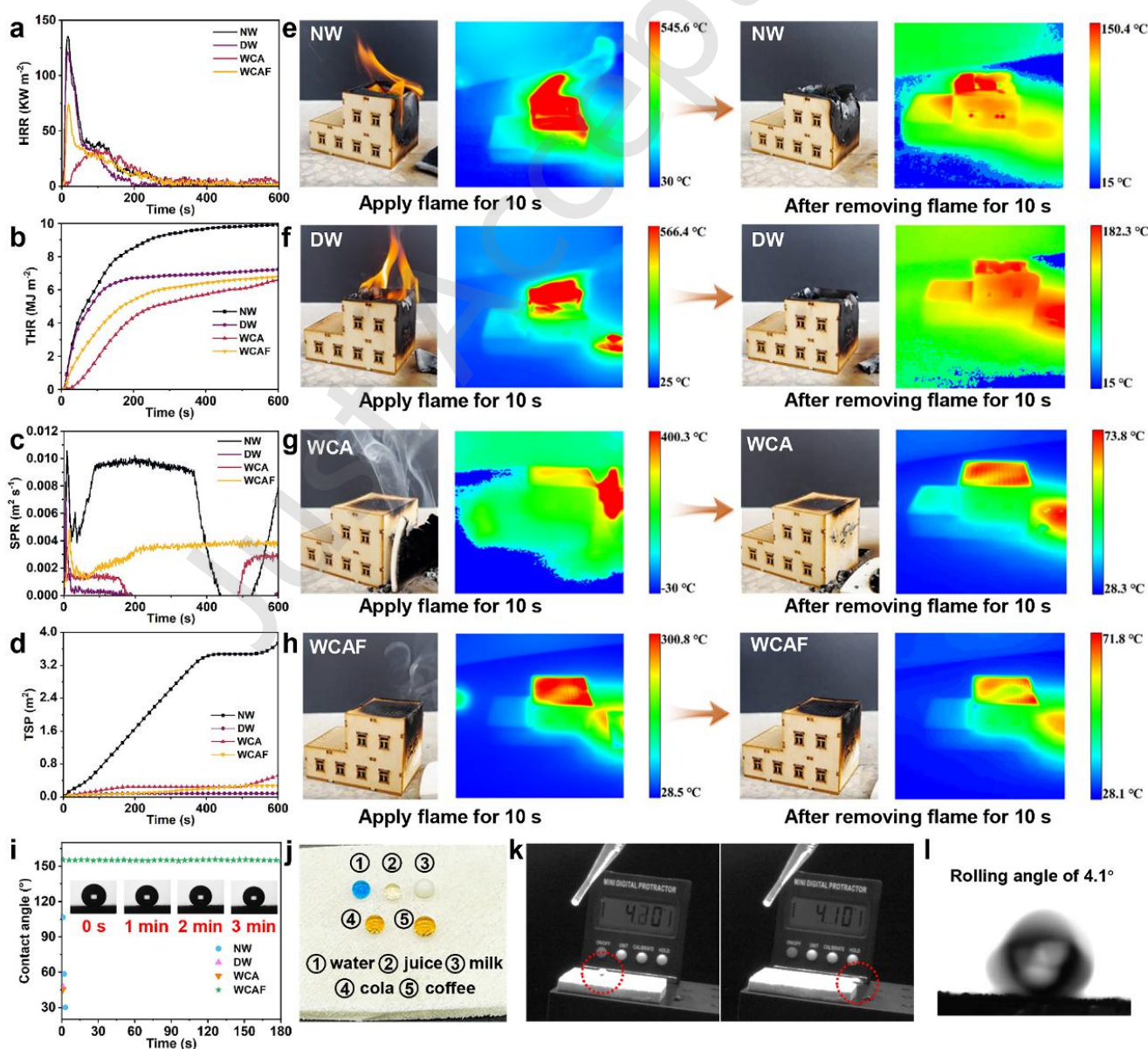
A custom-made fire testing apparatus was additionally constructed to visually evaluate fire resistance in a more practical scenario, complementing the aforementioned small-scale flame tests. A camera and infrared thermal imager were used to observe the ignition behavior of wooden cabins, capturing both visible flame propagation

and thermal radiation dynamics (Figure 5e-h, Figure S15). Upon flame contact, NW ignited rapidly, with its porous structure causing immediate volume shrinkage. In contrast, DW exhibited more intense and rapid combustion and volumetric contraction, attributed to lignin removal that enhanced combustibility and heat transfer efficiency. Although WCA and WCAF exhibited moderate structural shrinkage and surface damage, they remained unignited, forming a dense char layer that further validated their ignition resistance.

Beyond fire resistance, superhydrophobicity was crucial for material's practical applications. As shown in Figure S16, the initial contact angle of NW is $77.1^\circ \pm 1.1^\circ$, indicating the natural hydrophilicity of wood. In contrast, Movie S1 and Movie S2 demonstrated the strong hydrophilicity of DW and WCA, where water was absorbed in less than a second, making it impossible to measure the contact angle. Notably, WCAF demonstrated exceptional water repellency and

stability. The contact angle remained essentially unchanged at 154° over a three-minute observation period (Figure 5i), confirming the durability of the wetting state. This outstanding superhydrophobic performance was attributed to hierarchical micro-nano roughness constructed by cement nanoparticles combined with low-surface-energy modifications, which effectively traps air to form a stable Cassie state [70]. Consequently, various common liquid droplets, such as water, milk, coffee, juice, and cola, retained spherical shapes on the surface (Figure 5j). Furthermore, WCAF exhibited a sliding angle of merely 4.1° (Figure 5k, l), well below the 10° threshold for superhydrophobicity. The low adhesion allowed water droplets to rolled off the WCAF surface, carrying away deposited dust bestowing WCAF with self-cleaning functionality (Figure S17). This property was primarily ascribed to the minimal solid-liquid contact area provided by the re-entrant texture of the nanoparticle-coated surface.

Figure 5. Cone calorimeter and thermal insulation measurement together with contact angles and sliding angles of NW, DW, WCA, and WCAF. (a) Average heat release rate (HRR). (b) Total heat release. (c) Average smoke production rate (SPR). (d) Total smoke production (TSP) of samples. (e-h) Digital images for the burning behavior of the wooden building model protected by NW, DW, WCA, and WCAF along with their side temperature variation with time captured by an IR camera. (i) Contact angle test for 3 min and the image of contact angle when it was 0 s, 1 min, 2 min, 3 min. (j) Non-wettability of various



liquids on WCAF. (k) Image of water droplet rolling down at 4.1° tilt board. (l) Roll-off behavior on WCAF at 4.1°.

2.5 Accelerated weathering and outdoor daytime radiative cooling performance of WCAF

To assess the environmental durability of WCAF, accelerated weathering experiments were conducted, including high temperature exposure (60 °C, 6 h), low temperature treatment (-20 °C, 6 h) and ultraviolet irradiation (10 h). As shown in Figure 6a, the WCAF-Aged maintained its overall morphology without visible cracking or deformation, indicating excellent structural stability. Contact angle measurements confirmed that the superhydrophobic state was largely preserved. The pristine WCAF exhibited a water

contact angle of $154.0^{\circ} \pm 1.5^{\circ}$, which remained above 150° after weathering test ($151.8^{\circ} \pm 1.2^{\circ}$). Such negligible variations demonstrate that the fluorinated surface chemistry and cement-induced hierarchical roughness were highly resistant to thermal and photochemical degradation.

The intrinsic optical stability of the materials was further verified (Figure 6b). After the aging process, the solar reflectance exhibited a minor decrease from 91.3% to 88.0%, while the mid-infrared emissivity within the atmospheric window (8-13 μm) remained virtually uncompromised. These results indicated that the bioinspired pane-nanobeads

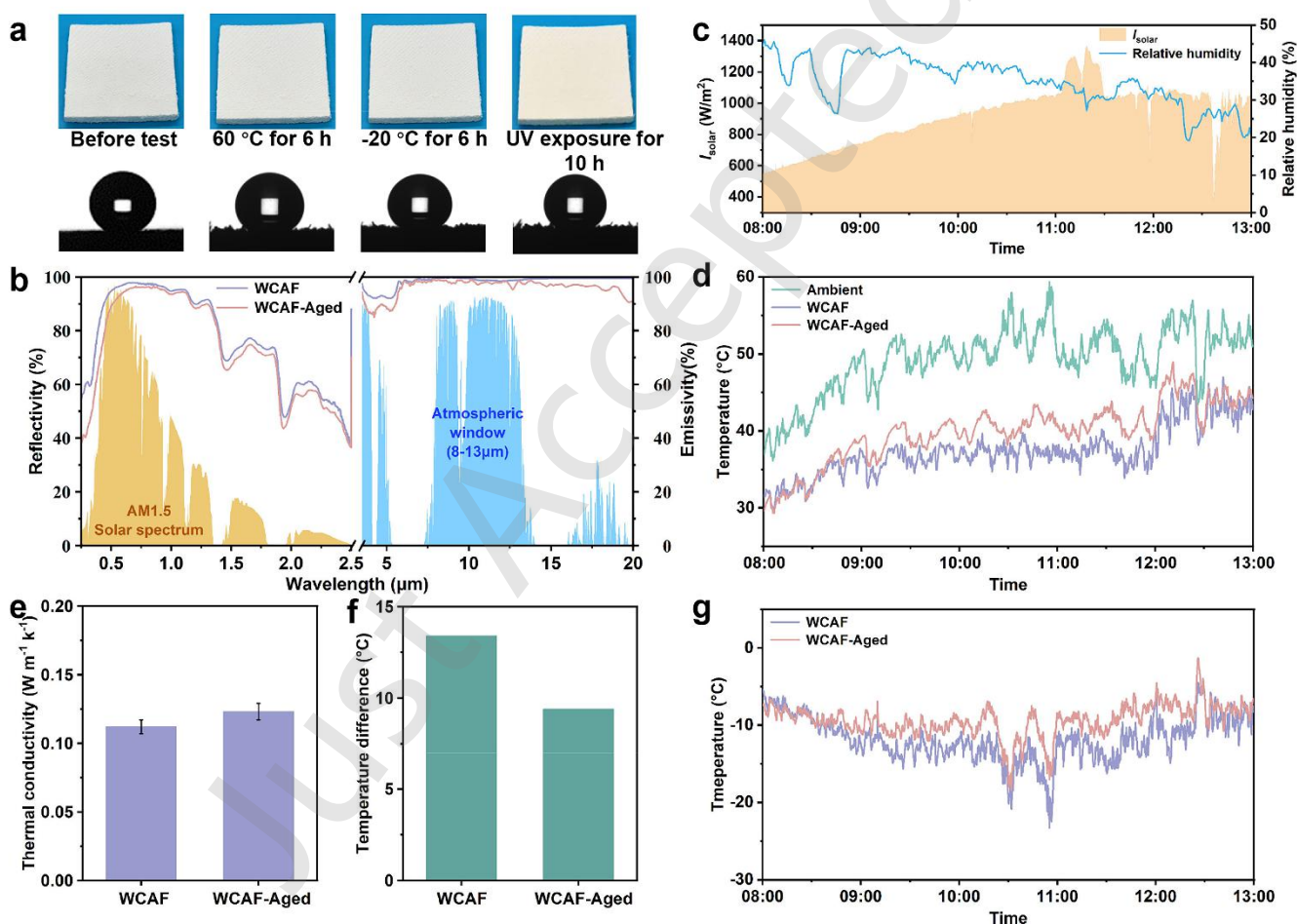


Figure 6. Accelerated weathering and outdoor daytime radiative cooling performance of WCAF. (a) Optical photographs and water contact angles of the WCAF and WCAF-Aged samples. (b) Spectral solar reflectance and mid-infrared emissivity curves of the WCAF before and after the accelerated weathering test. (c) Solar intensity fluctuation and relative humidity change during the outdoor test. (d) Real-time temperature of samples and ambient during the outdoor test. (e) Thermal conductivities of WCAF and before and after the accelerated weathering test. (f) Average temperature differences between samples and ambient during the outdoor test. (g) Real-time temperature differences between samples and ambient during the outdoor test.

architecture largely sustained its broadband scattering and radiative capabilities under aggressive environmental stresses. Subsequently, the outdoor daytime radiative cooling performance of aged sample was then evaluated, with meteorological parameters continuously recorded to account for ambient variability (Figure 6c). As illustrated in

Figure 6d, WCAF maintained its excellent cooling capability while the temperature of WCAF-Aged was 3.96 °C higher than that of the pristine WCAF. This modest performance degradation is primarily ascribed to the slight loss in solar reflectance, leading to marginally increased solar heat gain under intense irradiation.

Despite this, quantitative thermal analysis underscored the sustained cooling durability of the aged material. The thermal conductivity exhibited a minimal increment from $0.112 \text{ W m}^{-1} \text{ K}^{-1} \pm 0.005 \text{ W m}^{-1} \text{ K}^{-1}$ to $0.123 \text{ W m}^{-1} \text{ K}^{-1} \pm 0.006 \text{ W m}^{-1} \text{ K}^{-1}$ (Figure 6e), verifying that the porous cellular framework and the inorganic nanobead network remained structurally intact to effectively suppress parasitic convective heat transfer. Consequently, during the continuous daytime cooling test, the pristine WCAF achieved an average temperature drop of 13.4°C , and impressively, the WCAF-Aged sample still delivered a substantial cooling difference of 9.4°C (Figure 6f). The dynamic temperature tracking (Figure 6g) further corroborated that both samples consistently operated well below the ambient temperature. Collectively, these results indicate that WCAF retained its overall morphology and cooling performance after the tested thermal and UV aging protocol, supporting its promising environmental stability under the tested conditions. However, these results do not establish long-term outdoor service life.

To further assess the resistance of WCAF to mechanical and water-related stresses, separate specimens were subjected to extended thermal cycling, UV irradiation, mechanical abrasion, and rain rinsing (Figure S18). Thermal cycling caused almost no visible color or morphological change, with the solar reflectance decreasing from 91.3% to 89.1%, the mid-infrared emissivity remaining at 97.3%, and the water contact angle remaining at 151.2° . UV irradiation caused noticeable yellowing and reduced the solar reflectance to 85.07%, mainly owing to a decrease in the visible-light region, while the emissivity and water contact angle remained 94.78% and 150.3° , respectively. After mechanical abrasion, the solar reflectance and emissivity were 87.06% and 94.9%, respectively, and the contact angle remained 145.9° , possibly because partial damage to the surface C-A-S-H nanoparticle structure affected the optical and wetting properties. Rain rinsing resulted in a solar reflectance of 85.88%, an emissivity of 94.81%, and a contact angle of 135.7° . This decrease in wettability may be related to water infiltration into the wood cellular network, whose cell lumens are approximately 20–30 μm in diameter, thereby disturbing the surface architecture. These supplementary tests further support the environmental stability of WCAF under the tested conditions, while indicating that mechanical wear and water exposure remain potential limitations for long-term outdoor applications.

2.6 Energy consumption simulation analysis

Given the excellent weathering stability and sustained radiative cooling performance after accelerated aging, the practical energy-saving potential of WCAF was further evaluated through whole-building energy simulations. The high solar reflectivity (91.3%) and infrared emissivity (99.1%) of WCAF, coupled with its superhydrophobicity and fire resistance, positioned it as an highly efficient outdoor radiative cooling material. Energy Plus version 22.0 was employed to simulate its impact on cooling energy demand and building energy balance. Table S3 detailed the parameters for applying WCAF to building exterior walls and roofs, while parameters of all other materials used in simulation were listed in Table S4. Additionally, the

simulation adopted an internal boundary condition with a cooling setpoint of 26°C , utilizing typical meteorological year (TMY) weather data for external conditions. Furthermore, the environmental benefits were quantified by calculating the reduction in building-related carbon emissions based on specific grid carbon emission factors (Table S5).

The schematic of the energy-saving building model is presented in Figure S19, illustrating how radiative cooling enhances indoor thermal comfort for occupants. A typical three-story office building model (Figure S20) was constructed based on the properties of baseline wall and roof materials, as well as the performance of WCAF, to predict energy consumption. In this study, capital cities of each province and municipality were selected as simulation targets, including Beijing, Jinan, Taipei, Hefei, Urumqi, etc. These cities represent all climatic zones in China, enabling the research findings to be generalized nationwide. Figure 7a–b showed excellent cooling energy savings related to the baseline buildings. Xishuangbanna (43.6 MJ/m^2), Baise (32 MJ/m^2), Haikou (30.8 MJ/m^2), and Guangzhou (30.2 MJ/m^2) exhibited the highest cooling energy savings among these 20 cities. These cities are all located in southern China, characterized by hot climates, making them optimal candidates for installing this material to reduce cooling energy consumption. Regarding the energy saving rate, Lanzhou possessed the highest cooling energy reductions of 21.5%. Substantial CO_2 emission reductions were also revealed in Figure 7c, d, showing similar results with that of energy savings. In hot region like Taipei and Haikou, where baseline CO_2 emissions reached 88.7 TCO_2 and 75.8 TCO_2 , the WCAF brought 7.4 TCO_2 and 6.2 TCO_2 reduction, respectively. However, Taiyuan and Shijiazhuang exhibited the greatest emission reductions of 8.6 TCO_2 and 9.8 TCO_2 , which can be ascribed to their energy structure dominated by non-renewable resources. To elucidate the energy performance of buildings integrated with a selective emitter WCAF relative to the baseline building, Figure 7e–g present the energy consumption distribution and cooling cost savings across representative cities. A heat map (Figure S21) vividly demonstrated the remarkable climatic diversity in China, mirroring the spatial distribution of multiple climate types spanning from tropical to temperate and from humid to arid regimes. Five representative cities were chosen to conduct the following energy analysis. Figure 7e, f depicted the energy consumption share (%) of four end uses—cooling, lighting, equipment and heating according to Table S6, S7. In the baseline building, cooling dominated energy use in tropical and subtropical cities (e.g., 59.7% in Haikou), reflecting intense cooling demand in warm climates. For the building with WCAF, the share of cooling energy is reduced (e.g., 57.6% in Haikou), while the shares of lighting and equipment energy increase moderately, indicating that WCAF alleviated the cooling load without compromising other functional energy needs. Since all energy uses in the simulation are electricity-driven, the reduction in energy consumption translated directly to a lower carbon footprint, verifying that buildings with WCAF operate as low-carbon buildings. Figure 7g quantified economic benefits, which showed that cities with high cooling demand (e.g.,

Xishuangbanna) achieved substantial absolute savings and most cost savings ranged from 15%-20%. Similar to the simulation results of energy savings, Lanzhou again demonstrated the highest cost savings. Overall, the simulation indicated that WCAF reduced cooling energy consumption by an average of 27.2 MJ/m², corresponding to a mean energy saving rate of 12.9% (Figure S22).

At the same time, the potential energy-saving effects of WCA on building exterior walls and roofing were also simulated under the same conditions as before, with the exception of material parameters as shown in Table S8. Xishuangbanna (43.7 MJ/m²), Baise (32.1 MJ/m²), Haikou (30.9 MJ/m²), and Guangzhou (30.3 MJ/m²) exhibited the highest cooling energy savings among these 20 cities (Figure S23, S24). The results of cooling energy savings were almost the same as WCAF's. Although these values slightly outperform WCAF, the rapid water absorption of comparative materials restricted their outdoor applicability. Similarly, an energy-saving map for typical years in China

was further calculated (Figure S25). The energy-saving map was completely consistent with that of WCAF, further illustrating the significant potential of WCAF in practical applications for energy conservation.

A comprehensive comparison between WCAF and other radiative cooling aerogel materials underscore WCAF's distinctive advantage in integrating high performance and high practical utility (Table S9). WCAF achieved a robust cooling temperature of 11.6 °C, standing out as a top performer among radiative cooling aerogels while avoiding the trade-off that arise from prioritizing cooling alone. For instance, P4-G1 exhibited exceptionally high cooling performance yet lacked critical mechanical properties and hydrophobicity, leaving its practical applicability uncertain. Notably, WCAF's mechanical strength stood out with a compression strength of 2.8 MPa (Figure S26), far surpassing that of comparative materials. Water and fire resistance are

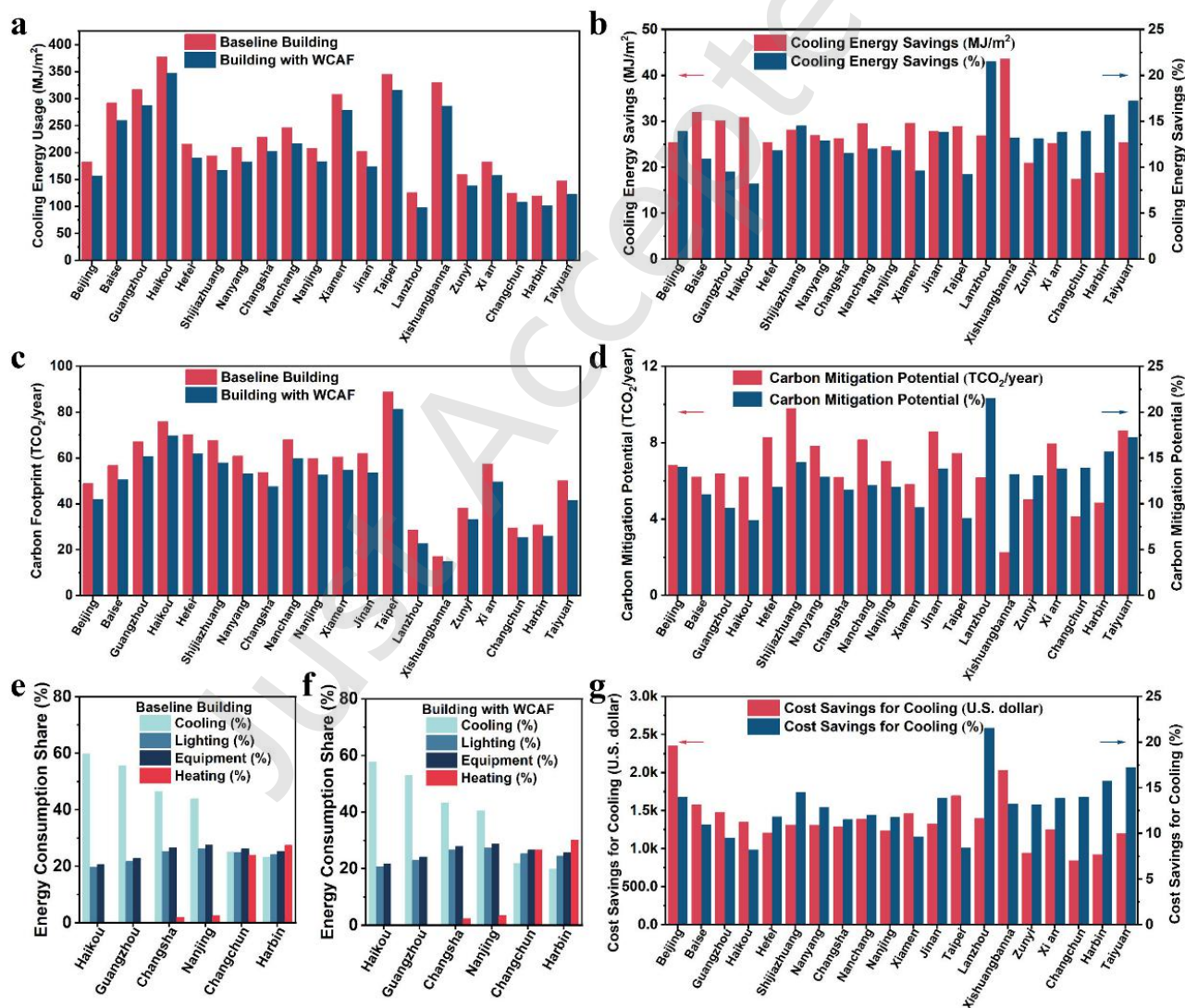


Figure 7. (a, b) Cooling energy usage (MJ/m²) and energy savings for the baseline building and the building covered by WCAF. (c, d) Carbon footprint (TCO₂/year) of cooling energy used in the baseline building and the building covered by WCAF. (e) Energy consumption share of baseline building in representative climate zones. (f) Energy consumption share of building with WCAF in representative climate zones. (g) Average cost savings (U.S. dollars) for cooling when Baseline Building applied with WCAF.

often concurrent requirements for outdoor applications. However, most materials in the table struggled to balance

these two attributes, typically excelling in one while falling short in the other. Even GMS0.6, which managed to integrate both fire resistance and hydrophobicity, only achieved a modest cooling effect of 4.8 °C. Thus, taken together, WCAF demonstrated a highly balanced performance profile, making it uniquely positioned to bridge lab-scale innovation and real-world demands by simultaneously delivering high cooling efficiency, superior mechanical strength, exceptional fire resistance, and stable superhydrophobicity.

3 Conclusions

In summary, we have fabricated butterfly-inspired WCAF material by in-situ anchoring of cement-based nanoparticles and fluorination modification. Leveraging the unique pane-nanobeads photonic structure, WCAF exhibited excellent solar reflectivity (91.3%) and infrared emissivity (99.1%). Under solar irradiance (>800 W/m²), the material achieved a remarkable cooling temperature of 11.6 °C, demonstrating exceptional energy-saving potential by reducing cooling energy consumption by up to 43.6 MJ/m². Beyond its cooling capabilities, the WCAF exhibited outstanding superhydrophobicity and flame resistance. Crucially, the WCAF demonstrated exceptional environmental durability against rigorous accelerated weathering. After enduring severe thermal cycling and continuous UV irradiation, the material preserved its biomimetic architecture and optical stability, sustaining a robust cooling drop of 9.4 °C and a water contact angle over 150°, thereby demonstrating promising environmental stability and weather resistance under the tested accelerated conditions. This multifunctional WCAF, combining energy-saving, water repellency, fire resistance, easy-to-process, and green sustainable characteristics, significantly contributes to improving material efficiency and achieving carbon neutrality.

Electronic Supplementary Material: Supplementary material, including materials, methods, notes, and characterization data, spectra, results is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909199>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Chen Lu: Data curation, Formal analysis, Investigation,

Methodology, Writing-original draft. Kaili Wang: Conceptualization, Funding acquisition, Project administration, Supervision, Writing-review and editing. Yue Li: Data curation, Formal analysis. Wenjuan Huang: Formal analysis, Investigation. Youming Dong: Data curation, Methodology. Yahui Cai: Data curation, Formal analysis. Dan Tian: Data curation, Formal analysis. Jin Wang: Data curation, Investigation. Jianwei Zhao: Methodology, Formal analysis. Xiaorong Liu: Writing-review and editing, Supervision, Funding acquisition. Jianzhang Li: Writing-review and editing, Supervision. Wenshuai Chen: Writing-review & editing.

Use of AI statement

None.

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Electronic Supplementary Material

Butterfly-inspired, nano-cement-engineered wood aerogel for superhydrophobic and flame-retardant passive daytime cooling

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Experimental section

Materials

Balsa wood was purchased from Zhuhai, Guangdong Province, then cut into 50 mm × 50 mm × 5 mm, 10 mm × 100 mm × 100 mm (radial × tangential × longitudinal). All chemicals used were as followed: Sodium chlorite (NaClO₂, 80%), acetic acid glacial (CH₃COOH), 1H,1H,2H,2H-perfluorodecyltrimethoxysilane (PFDTs, 97%), Calcium chloride dihydrate (CaCl₂·2H₂O, powder, 99.0%), aluminum chloride hexahydrate (AlCl₃·6H₂O, powder, ≥97.0%), sodium metasilicate nonahydrate (Na₂SiO₃·9H₂O, powder, 100%) and ethanol (C₂H₅OH, ≥99.7%).

Preparation of Delignified Wood (DW)

The acid delignification method was used in this work. Put the balsa wood into 1 wt% NaClO₂ solution (PH was adjusted to 4.6 with CH₃COOH) at 80 °C for 18 h. The solution was changed every 6 h. After the treatment, the samples were washed with hot deionized water to until the acid solution were completely removed. Lastly, the DW was obtained by freeze-drying for 24 h.

Preparation of Wood Cement Aerogel (WCA)

The cement particle was produced by three precursors: CaCl₂·2H₂O, AlCl₃·6H₂O, Na₂SiO₃·9H₂O. To begin with, 100 g CaCl₂ solution (1M), 80 g Na₂SiO₃ solution (1M), 20 g AlCl₃ solution (1M) were mixed in sequence. Then DW was immersed into the mixture promptly for 1 h. The process above was performed at ambient temperature under stirring. After synthesis, the sample was freeze-dried for 24 h to get WCA.

Preparation of Fluorinated Wood Cement Aerogel (WCAF)

The WCA sample was immersed into 2 wt% PFDTs solution for 24 h, which was dissolved in ethanol. Then the samples were placed in an oven at 60 °C to get WCAF.

Materials Characterization

The environmental scanning electron microscopy (ESEM, FEI Quanta 200) was used to observe surface morphologies of

NW, DW, WCA and WCAF samples. The elemental maps were obtained by energy dispersive spectroscopy (EDS) on the attachment to SEM. X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha, America), Fourier transform infrared spectroscopy (FT-IR, Thermo Scientific Nicolet iS 50), X-ray Diffraction (XRD, Rigaku SmartLab SE, Japan) were used to analysis chemical composition. Also, FT-IR equipped with an integrating sphere was used to measure the reflectance of samples from 400 to 4000 cm⁻¹. Thermal conductivity was measured by Thermal conductivity meter (Hot Disk TPS 2500S, Sweden). The combustion behavior of samples was evaluated by Cone calorimeter (VOUCH 6810, China) at a heat flux of 35 KW·m⁻² according to ISO5660. Water contact angles (Cas) were measured with a Contact Angle System OCA20 (Dataphysics, Germany). The multipath channel temperature recorder (HuiPu TCP-400X) was used to monitor the ambient temperature and samples' temperature. The solar power was monitored by solar power meter (TES-1333R, China). The wind speed and humidity were measure by anemometer (BiaoZhi GT8913) and hydrometer (MiaoXin TH40G4-EX). A thermal infrared camera (UNI-T UTI160S) was employed to record the thermal infrared images.

Outdoor daytime radiative cooling performance test

The cooling performance of NW, DW, WCA, and WCAF was conducted on a sunny day at Nanjing Forestry University. Six cavities were cut on one side of a 30×30×30 cm polypropylene foam, each with a length and width of 5 cm. All sides except the one with cavities were wrapped with aluminum foil. To investigate the influence of thermal convection, the cavity-side surface will be covered with a polyethylene film on one day of the test, whereas no coverage will be applied on another day. Temperatures of samples and cavities were recorded using a multipath channel temperature recorder.

Accelerated Weathering and Environmental Stability Testing

To evaluate the environmental durability of the materials under extreme outdoor conditions, an accelerated weathering test was designed and conducted on the WCAF samples. Specifically, the pristine WCAF samples were first heated in an oven at 60 °C for 6 h, and subsequently transferred to a freezer at -20 °C for 6 h to simulate severe thermal cycling. Following this, the samples were placed at a distance of 10 cm from a 365 nm ultraviolet (UV) lamp and exposed to continuous irradiation for 10 h. After this accelerated aging process, the treated samples (denoted as WCAF-Aged) were collected for optical characterization and Outdoor daytime radiative cooling performance test. Besides, we performed additional environmental-stability tests, including mechanical abrasion and rain rinsing, alongside the thermal-cycling and UV-irradiation tests. The thermal-cycling test comprised 10 cycles, with each cycle consisting of exposure at 60 °C for 6 h followed by treatment at -20 °C for 6 h. The UV-irradiation test was conducted for 48 h. Mechanical abrasion was performed by rubbing the sample on sandpaper under a 100 g load, while rain rinsing was simulated by exposing the sample to tap water from a faucet for 1 min.

Note S1. Calculations of solar reflectivity and infrared emissivity.

The solar reflectivity R_{solar} of NW, DW, WCA, and WCAF were calculated as follows:

$$R_{solar} = \frac{\int_{0.3}^{2.5} I_{solar}(\lambda) \cdot R(\lambda) d\lambda}{\int_{0.3}^{2.5} I_{solar}(\lambda) d\lambda} \quad (1)$$

where λ is the wavelength, $\bar{\epsilon}_{8-13}$ is AM 1.5 global solar irradiance, $R(\lambda)$ is spectral reflectance of sample's surface at wavelength λ .

The average infrared emissivity with atmospheric window $\bar{\epsilon}_{8-13}$ can be calculated as follows:

$$\bar{\epsilon}_{8-13} = \frac{\int_8^{13} \epsilon(\lambda) \cdot \mu(\lambda, T) d\lambda}{\int_8^{13} \mu(\lambda, T) d\lambda} \quad (2)$$

All samples are not transparent, so according to Kirchhoff's law, emissivity is equal to absorptivity, which means $\epsilon(\lambda)=1-R(\lambda)$ is samples' spectral infrared emissivity. $\mu(\lambda, T)$ is calculated by Planck's law of blackbody radiation at a

defined temperature T:

$$\mu(\lambda, T) = \frac{2\pi hc^2}{\lambda^5} \cdot \frac{1}{e^{\frac{hc}{\lambda k_B T}} - 1} \quad (3)$$

where h is Planck's constant, c is the speed of light, k_B is the Boltzmann constant.

Note S2. Calculation of daytime cooling power of WCAF.

Considering the complexity of thermal environment, the net cooling power (P_{net}) during daytime can be calculated by the equations followed:

$$P_{net} = P_{rad} - P_{amb} - P_{sun} - P_{non-rad}$$

where P_{rad} is the power that WCAF radiate out through infrared emission, P_{amb} is the power that atmosphere radiate heatdownward, P_{sun} is the heating energy from solar irradiance absorbed by coolers, $P_{non-rad}$ is the heat lost caused by non-radiative path-conduction and convection. All of these can be calculated as follows:

$$P_{rad} = \int \cos \theta d\Omega \int_0^\infty \mu(\lambda, T) \varepsilon(\lambda, \theta) d\lambda \quad (4)$$

$$P_{amb} = \int \cos \theta d\Omega \int_0^\infty \mu(\lambda, T_{amb}) \varepsilon(\lambda, \theta) \varepsilon_{atm}(\lambda, \theta) d\lambda \quad (5)$$

$$P_{sun} = \int_0^\infty I_{solar}(\lambda) \varepsilon(\lambda, \theta_{solar}) d\lambda \quad (6)$$

$$P_{non-rad} = h_c(T_{amb} - T) \quad (7)$$

Extraterrestrial AM1.5 and Global Tilt AM1.5 are two commonly used solar spectral distribution curves. Since the radiative cooling investigated in this study is performed at the Earth's surface, Global Tilt AM1.5 is selected as demonstrated in ASTM G173-23. Besides, what needs to be explained is that $\int d\Omega$ can be expressed as $2\pi \int_0^{\frac{\pi}{2}} \sin \theta d\theta$, which means the angular integral over a hemisphere. In equation (5) $\varepsilon_{atm}(\lambda, \theta) = 1 - \tau(\lambda) \frac{1}{\cos \theta}$ is the emissivity of the atmosphere, and the $\tau(\lambda)$ is the atmospheric transmittance in the zenith direction. The sample surface is designed to face the sun, so there is no angular integral. That is to say: $\theta=0$, only the value in the zenith direction is adopted in the calculation of the emissivity of the sample. $\varepsilon_{atm}(\lambda, \theta)$ represents the spectral transmittance of the atmosphere, which can be obtained from the professional website in the field of atmospheric radiation: <https://www.gemini.edu/observing/telescopes-and-sites>. The non-radiative heat transfer coefficients (h_c) in equation (7) were chosen as 0,1,2,3,6 W/m² K. T_{amb} and T are temperatures of ambient and coolers, where T_{amb} is fixed at 298 K.

Note S3. DFT Calculation.

Spin-polarized density functional theory (DFT) calculations were carried out with the Vienna Ab initio Simulation Package (VASP)¹. Exchange–correlation effects were described using the Perdew-Burke-Ernzerhof (PBE) functional within the generalized gradient approximation (GGA)². Van der Waals interactions were included via the DFT-D3 scheme with zero damping³. A plane-wave kinetic-energy cutoff of 450 eV was adopted. The γ -centered Monkhorst-Pack scheme with a K-point resolved value of 0.08 $\pi/\text{\AA}$ was used in all DFT computations⁴. The electronic self-consistent field (SCF) convergence was set to 10⁻⁵ eV, and ionic relaxations proceeded until residual forces were below 0.05 eV/ $\text{\AA}$. Post-processing was performed using VASPKIT⁵.

Note S4. Calculation of WCAF's band gap.

The reflectance spectrum $R(\lambda)$ was converted to photon energy E firstly. To estimate the optical band gap, the diffuse reflectance data were transformed using the Kubelka–Munk function called $F(R)$, which relates reflectance to an absorption-like term. According to the Tauc relation for indirect allowed transitions, the optical absorption near the band edge follows a power-law dependence on photon energy. Substituting α with $F(R)$ gives $(F(R)E)^n \propto (E - E_g)$. A linear

fit was applied to the rising portion using an indirect transition ($n=1/2$). All of these can be calculated as follows:

$$E(eV) = \frac{1.2398}{\lambda(\mu m)} \quad (8)$$

$$F(R) = \frac{(1-R)^2}{2R} \quad (9)$$

$$(\alpha E)^n = A(E - E_g) \quad (10)$$

Extrapolation of this linear region to the energy axis yielded an indirect optical band gap of 2.433 eV, with a correlation coefficient $R^2 \approx 0.95$. All data processing and fitting were performed in a Python environment, with full parameter ranges and procedures explicitly stated for reproducibility.

Note S5. Energy saving simulation in building sectors and carbon footprint (TCO₂/year) calculation.

Energy Plus version 22.0 was employed to evaluate the energy-saving potential. A three-story office building model with dimensions of 40×18×10.8 m was established, where each room measures 8×8 m and the corridor width is 2 m. All envelopes in the building adopt the baseline properties as listed in Table S3. For the building model with our radiative coolers, the samples are employed to replace both the exterior walls and the roof. Specific parameters of the samples can be found in Table S2 and Table S4. In our simulation, the cooling systems would cool down the room when the indoor temperature was above 26 °C. For the internal heat loads and ventilation boundary conditions in EnergyPlus, standard office schedules were defined as follows: an occupant density of 0.10 person/m² with a sensible heat gain of 120 W/person, an interior lighting power density of 9.0 W/m², and an electric equipment power density of 15.0 W/m² during active working hours (08:00-18:00 on weekdays). The cooling energy consumption of the building model will be calculated using the Typical Meteorological Year (TMY) weather files for different cities, and the TMY weather files can be obtained from "<https://climate.onebuilding.org/>".

To quantify the environmental benefits of WCAF in reducing building-related carbon emissions, CO₂ emission reduction data were derived from a two-step calculation process, integrating simulated cooling energy savings by Energy Plus and region-specific grid carbon emission factors. The calculation in the previous step has been mentioned in the preceding paragraph. Subsequently, the annual energy savings (kWh) simulated for each city are multiplied by region-specific grid carbon emission factors to obtain the carbon dioxide emissions. The data for the specific grid carbon emission factors can be accessed from the report of the Ministry of Ecology and Environment of China.

Note S6. Explanation of performance radar chart.

The parameters and evaluation indicators used to draw the performance radar chart were shown in Table S1. It can be seen that the cooling temperature and superhydrophobicity are evaluated by the tested temperature reduction and contact angle. For fire resistance, scoring was carried out according to the PHRR value of cone calorimetry, with PHRR=5 kW/m² as the optimal value of 100 points and PHRR=200 kW/m² as the worst value of 0 points. That is, the scoring formula for the flame resistance of the sample is $y = \frac{200 - PHRR_{sample}}{200 - 5} \times 100$. Since CNC cooler and Cooling wood were pure cellulose materials and flammable, they were rated 0 points. While Durable cooling wood had a certain flame resistance due to the incorporation of Mica@TiO₂. For optical performance, this work only considered the two representative indicators of solar reflectance and infrared emissivity. Due to the absorption of sunlight during the daytime, the calculation formula was $y = 0.6R_{solar} + 0.4E_{atmosphere_atmosphere}$. For sustainability, it was mainly evaluated from the perspectives of resource consumption and carbon dioxide emissions. Since this work was synthesized at room temperature, this score was set at 85 points. CNC cooler was evaluated as 60 because its bottom-up preparation method consumes a huge amount of energy in preliminary work; Cooling wood and Durable cooling wood adopt long-time hot press, so their scores in sustainability were slightly lower, 70 and 60 respectively. The evaluation of lightweight mainly

referred to the density. Taking a density of 0.001 as the optimal value of 100 points and a density of 1.2 as the worst value of 0 points. That is, the calculation formula is $y = (1.2 - \rho_{\text{sample}}) / (1.2 - 0.001) \times 100$.

Note S7. Porosity of samples.

Figures S3-S4 present the adsorption-desorption isotherms, pore size distribution curves, and specific surface area data of NW, DW, WCA, and WCAF, providing crucial insights into the pore structure evolution. NW had a dense stacking of cellulose-lignin, resulting in scarce and closed pores. With a specific surface area of only $0.007 \text{ m}^2/\text{g}$, the adsorption of NW was gentle in the low relative pressure region. The DW disrupted the rigid cross-linking of the cell wall, exposing mesopores between cellulose/hemicellulose units. The specific surface area jumps to $2.3 \text{ m}^2/\text{g}$, and the pore connectivity was enhanced, reflecting an increase in the openness of the pore network. The cement nanoparticles' construction (WCA) enabled "precise regulation" of the pore structure. The "occupancy-etching" effect of nanoparticles, on one hand, constructed a nanoscale rough structure on the cell wall surface; on the other hand, forms secondary mesopores within the pores. Cooperatively, the specific surface area reached a peak of $2.6 \text{ m}^2/\text{g}$. At this time, the steep rise of the adsorption-desorption curve in the medium-high relative pressure region ($P/P_0 < 0.6$) confirmed the enhancement of mesopore capillary condensation and the synergistic adsorption mechanism of multiscale pores induced by nanoparticles. Hydrophobic treatment triggered reconstruction of the pore structure. The interfacial deposition and pore infiltration of hydrophobic molecules led to the blockage of some mesopores. Meanwhile, the agglomeration and loss of nanoparticles due to hydrophobic interactions disrupted the multiscale pores constructed in WCA, causing the WCAF's specific surface area to drop to $1.7 \text{ m}^2/\text{g}$. Notably, WCAF maintained a relatively stable adsorption amount in the low relative pressure region ($P/P_0 < 0.4$), suggesting the ordered arrangement of hydrophobic agents in the micropore region, providing new clues for the study of interfacial interactions.

Note S8. Anisotropic mechanical compressive property.

Owing to the anisotropic structure of wood, three-directional compression performance tests were conducted on the four samples. Figure S26a-d showed the compressive stress-strain curves of NW, DW, WCA, and WCAF in the longitudinal, radial, and tangential directions, respectively. It could be observed from the stress-strain curves that all wood samples underwent the following deformation processes during compression. At low strains, the cell walls underwent lateral bending, exhibiting initial linear elastic deformation; they subsequently entered the yield stage, where nonlinear plastic deformation occurred with increasing strain; the cell walls gradually collapsed; and finally, due to the elimination of air voids, the cell walls fractured and densified. In addition, the longitudinal compressive strength of each sample was much higher than that in the radial and tangential directions, which can be attributed to the oriented arrangement of cellulose nanofibers in wood. The partially amplified compressive stress-strain curves were shown in Figure. S30e-h. It was evident that the compressive performance of DW was reduced due to the removal of lignin. The incorporation of cement nanoparticles enabled the longitudinal yield strength of WCA to reach 3.18 MPa. In contrast, the longitudinal yield strength of WCAF decreased to 2.85 MPa, which might be attributed to the loss of some nanoparticles caused by hydrophobic treatment, resulting in the loss of structural integrity. However, WCAF exhibited a longer elastic deformation period and was more comparable to the compression behavior of NW; that is, although WCAF had lost some rigidity, it had gained some plasticity. Notably, our WCAF, which was fabricated via top-down strategy, showed remarkably elevated mechanical performance compared with bottom-up aerogels. To date, most bottom-up aerogels had low compressive strength^{8, 12,18-20}, ranging from 0.1-0.7 MPa. Bottom-up aerogels relied on weak interparticle bonding and random structural packing, easily contributing to stress concentration induced by disordered aggregation. Conversely, our top-down WCAF inherited the intrinsic hierarchical structure of wood, particularly the oriented arrangement of wood cellulose nanofibers. Such oriented structure enabled efficient stress transfer across the matrix.

Figure S26i-l showed the appearance of $1 \times 1 \times 1$ cm³ samples before and after compression testing. As expected, NW and DW exhibited collapse in the longitudinal direction, cracking in the radial direction, while only densification was observed in the tangential direction. This anisotropic compression behavior was precisely determined by their hierarchical anisotropic structure. Similar results were also observed in WCA and WCAF, indicating that the incorporation of cement nanoparticles did not alter the intrinsic anisotropic nature of the material. Interestingly, almost no collapse was observed in the longitudinal compression of WCA, which was likely attributed to the dense "pane-nanobead" structure that effectively preserved the structural integrity, thus preventing significant collapse.



Figure S1. The preparation process of WCAF.

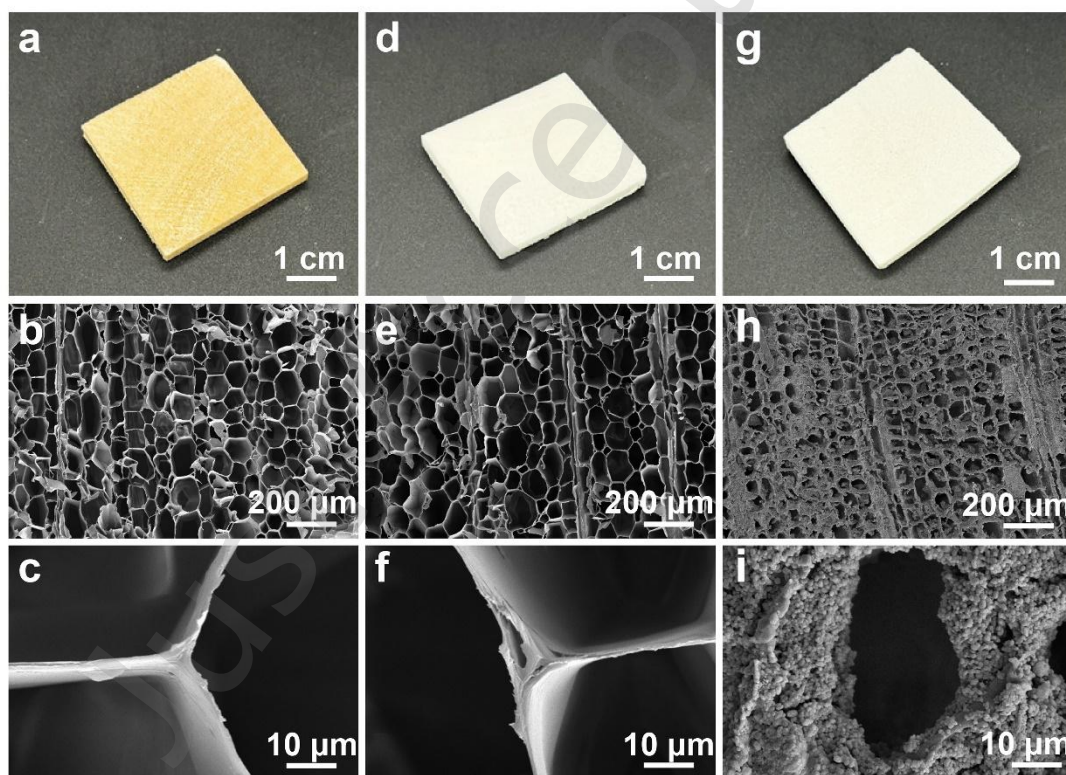


Figure S2. Optical images and microscopic features. (a-c) NW, (d-e) DW, (g-i) WCA.

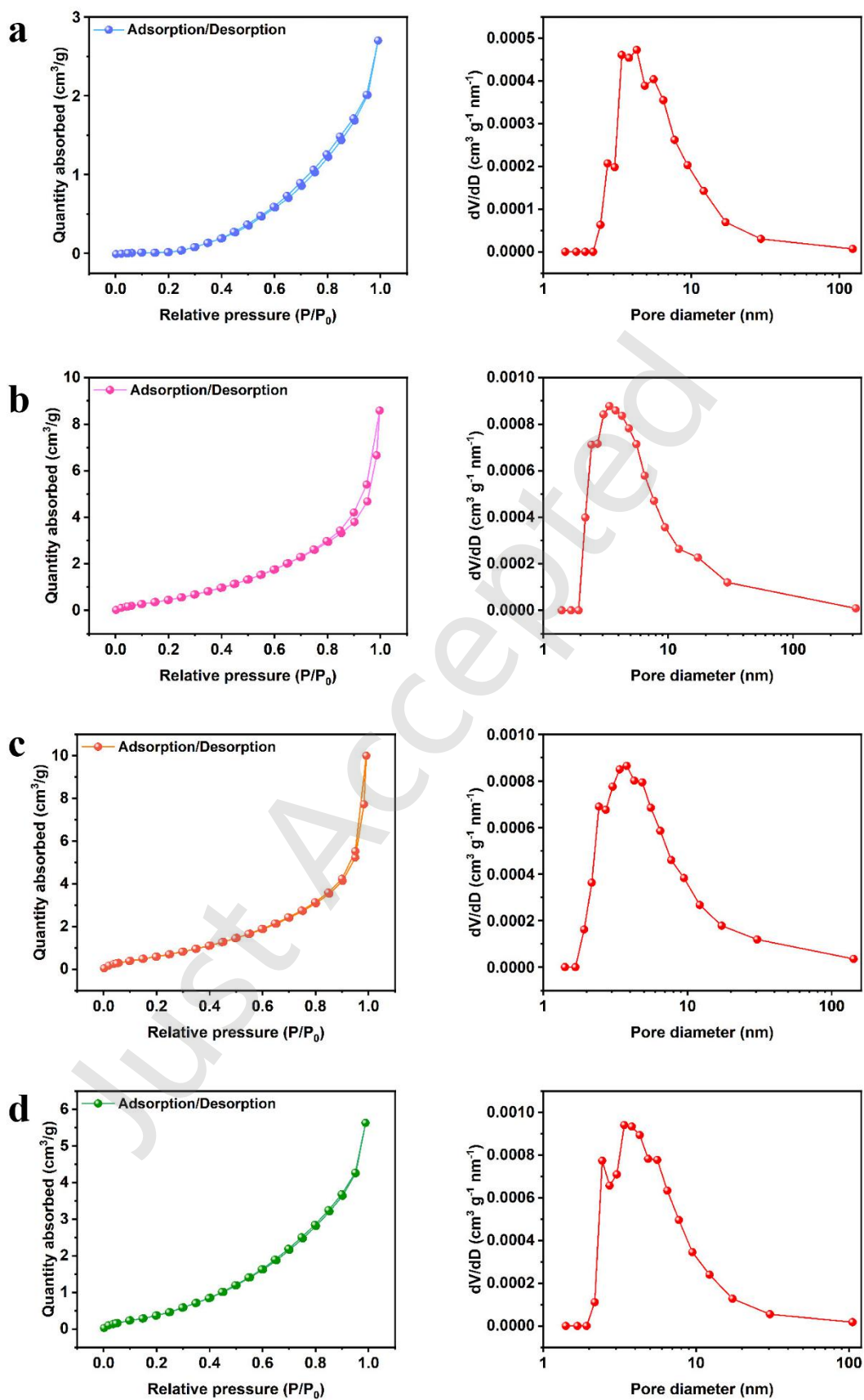


Figure S3. The BET test analysis. (a) NW, (b) DW, (c) WCA and (d) WCAF.

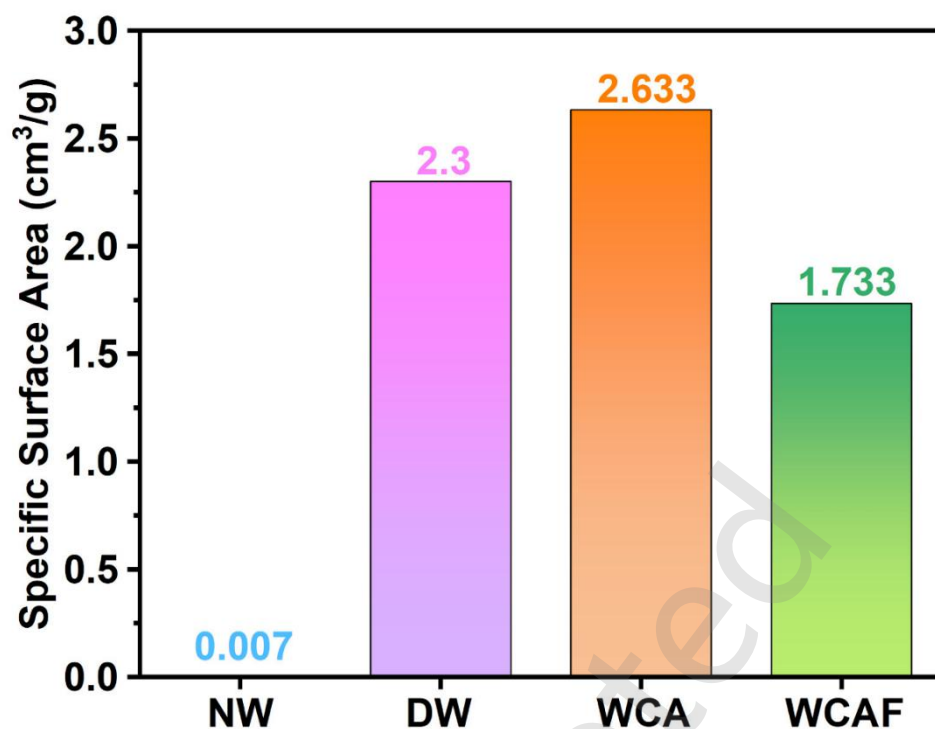


Figure S4. The specific area of NW, DW, WCA and WCAF.

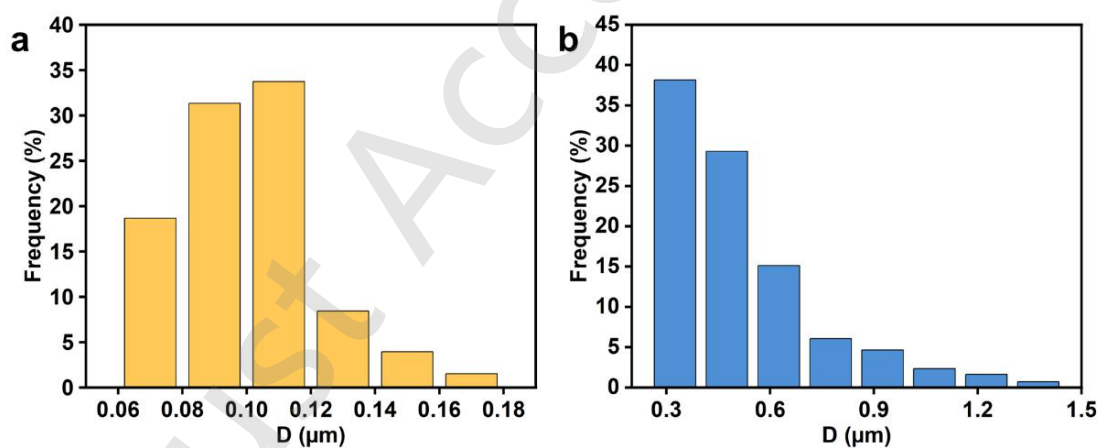


Figure S5. (a) Size distribution of pterin pigment nanoparticles on the butterfly wing scales. (b) Size distribution of C-A-S-H nanobeads anchored onto WCAF.

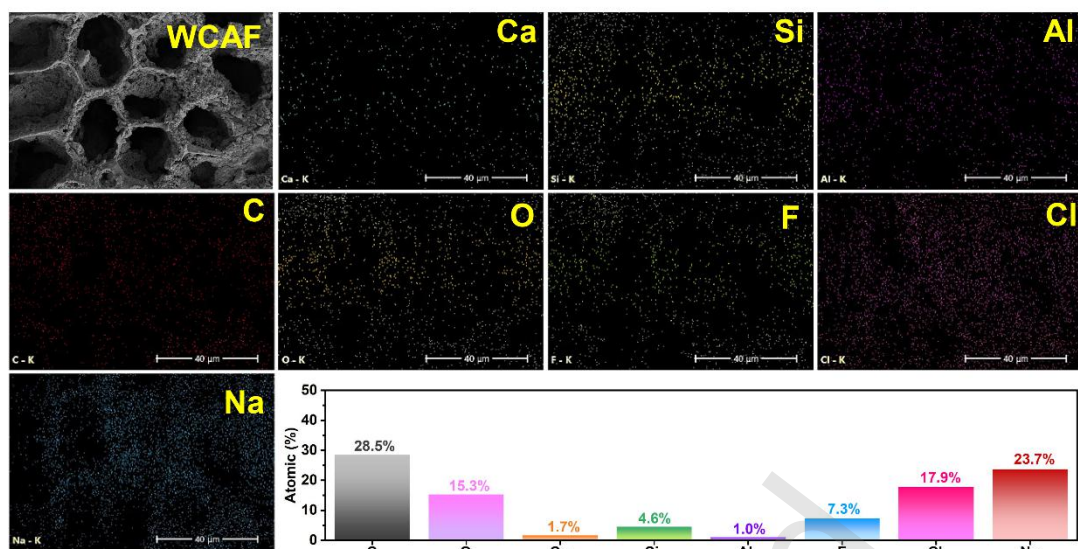


Figure S6. The EDS map of WCAF and the content of each element.

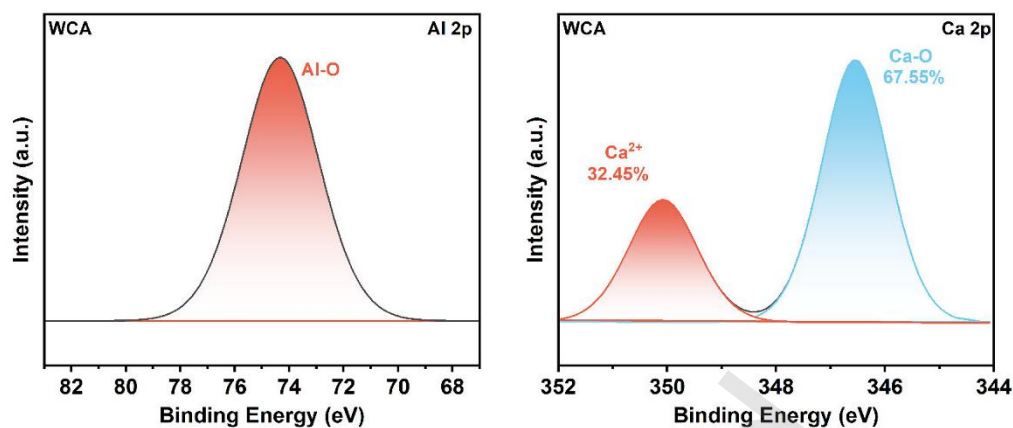


Figure S7. The Al 2p high-resolution and Ca 2p high-resolution spectrum of WCA.

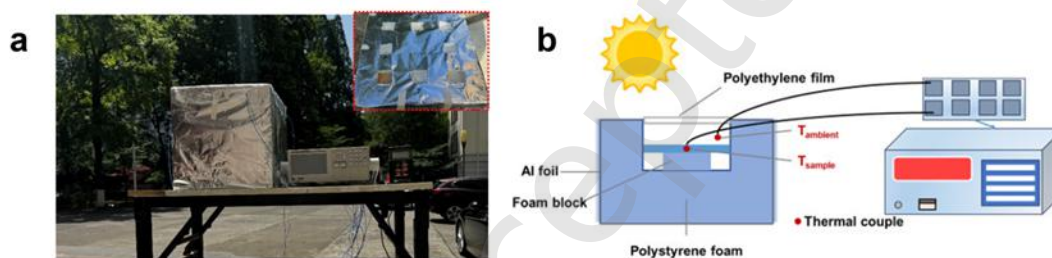


Figure S8. (a) Optical image of test devices and the inset was from top view. (b) Schematic diagram of test device.

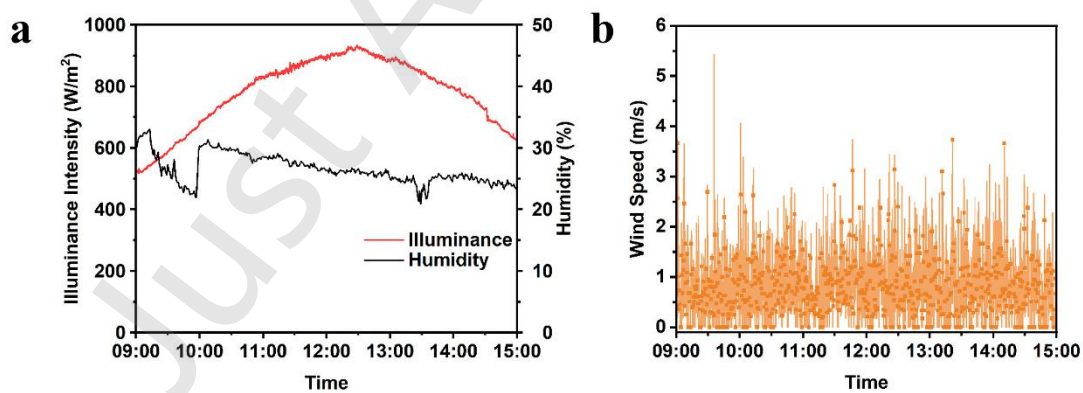


Figure S9. (a) The curves of solar irradiance and relative humidity during the test. (b) The wind speed data.

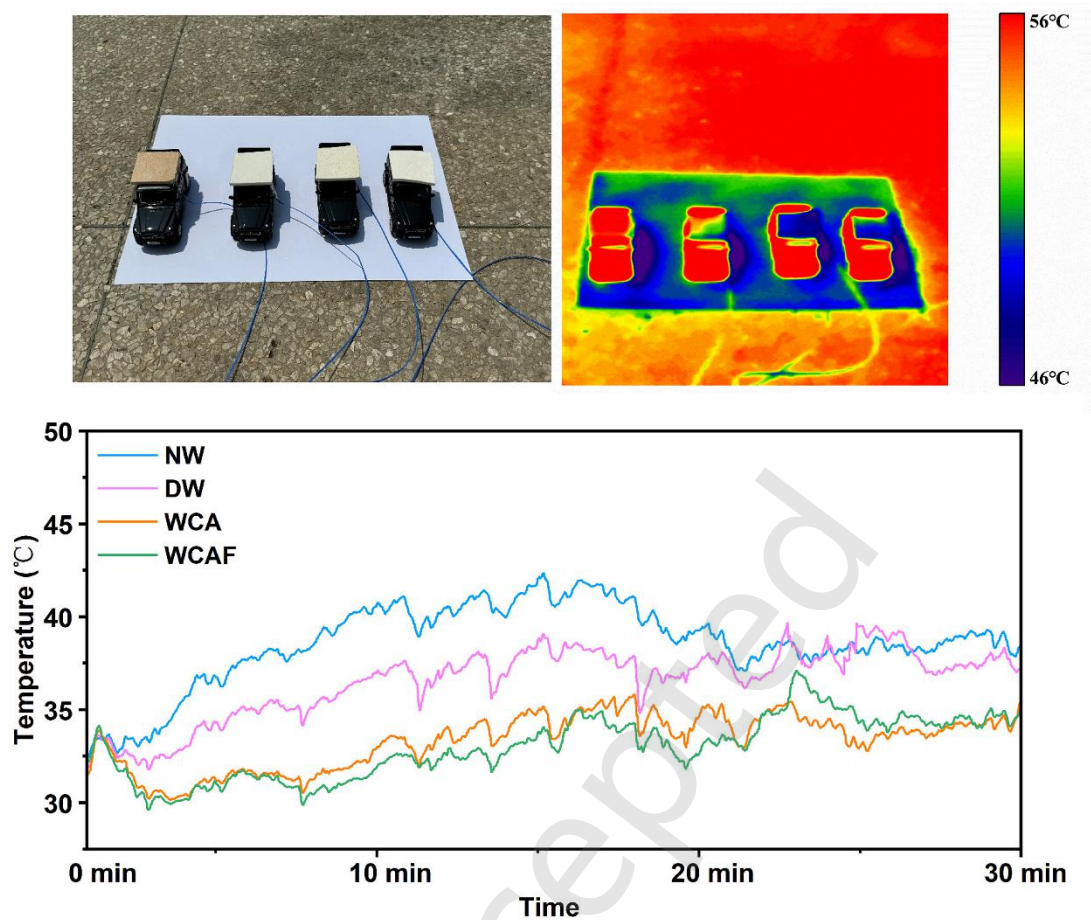


Figure S10. The optical image, thermal image and real time temperature curves of the car models.

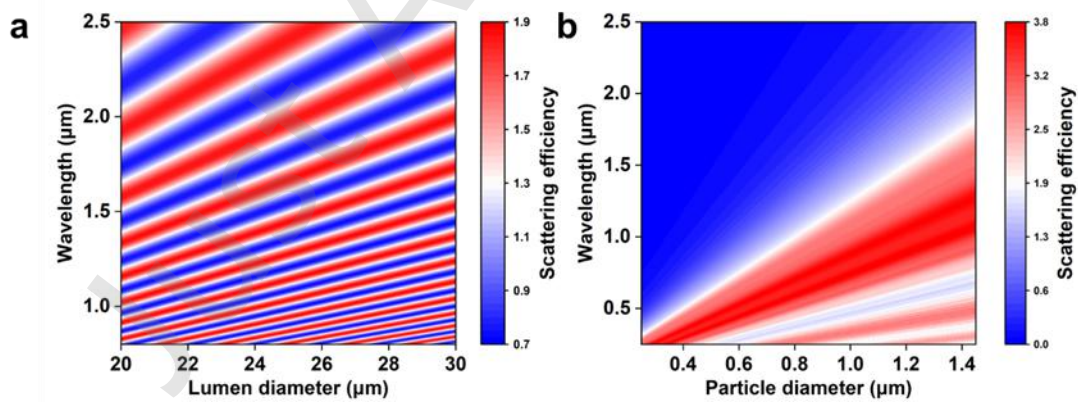


Figure S11. Theoretical optical scattering efficiency of (a) wood lumen and (b) C-A-S-H particle.

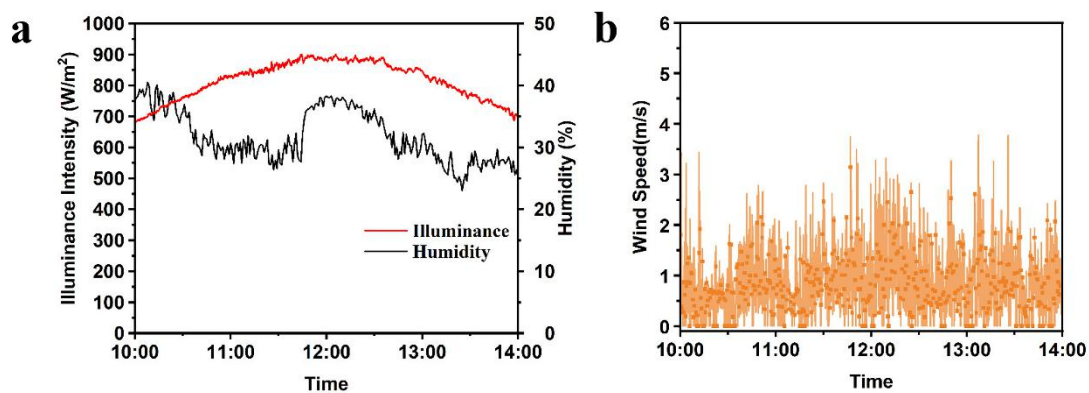


Figure S12. (a) The curves of solar irradiance and relative humidity during the test. (b) The wind speed data.

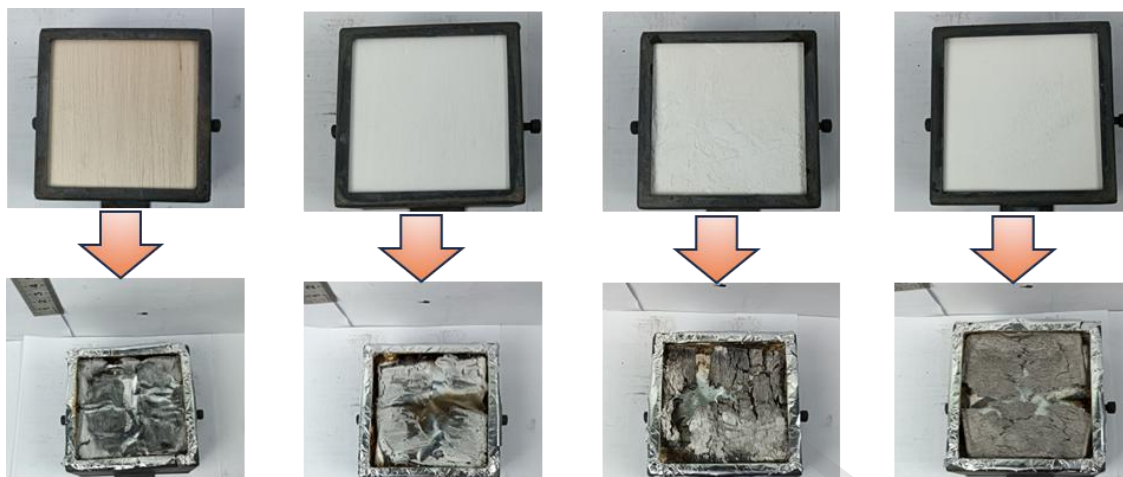


Figure S13. Morphology of char residue after Cone calorimeter measurement of NW, DW, WCA and WCAF.

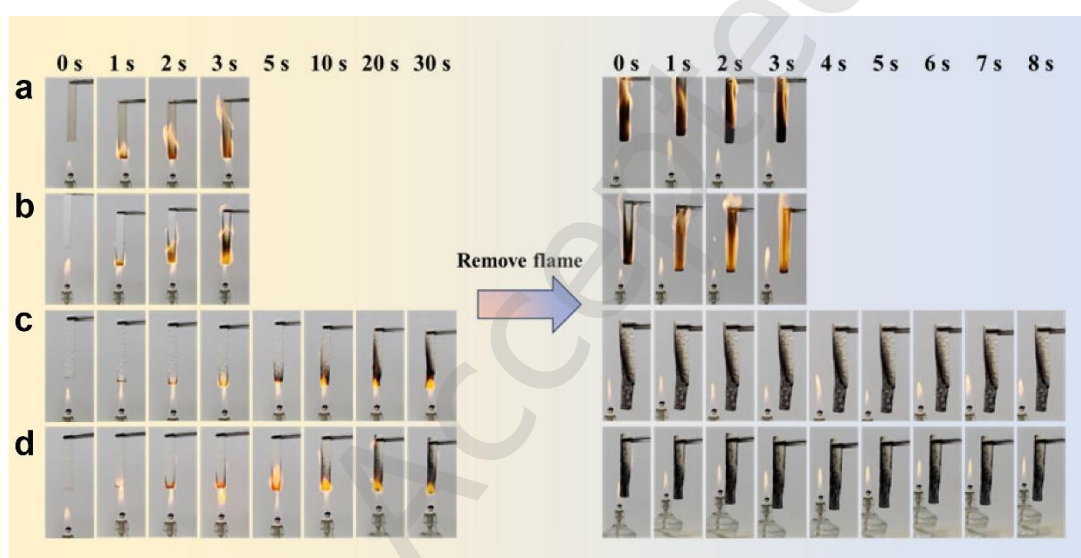


Figure S14. Flame exposure tests. (a) NW, (b) DW, (c) WCA, and (d) WCAF.



Figure S15. Digital images of NW, DW, WCA, WCAF before burning test.

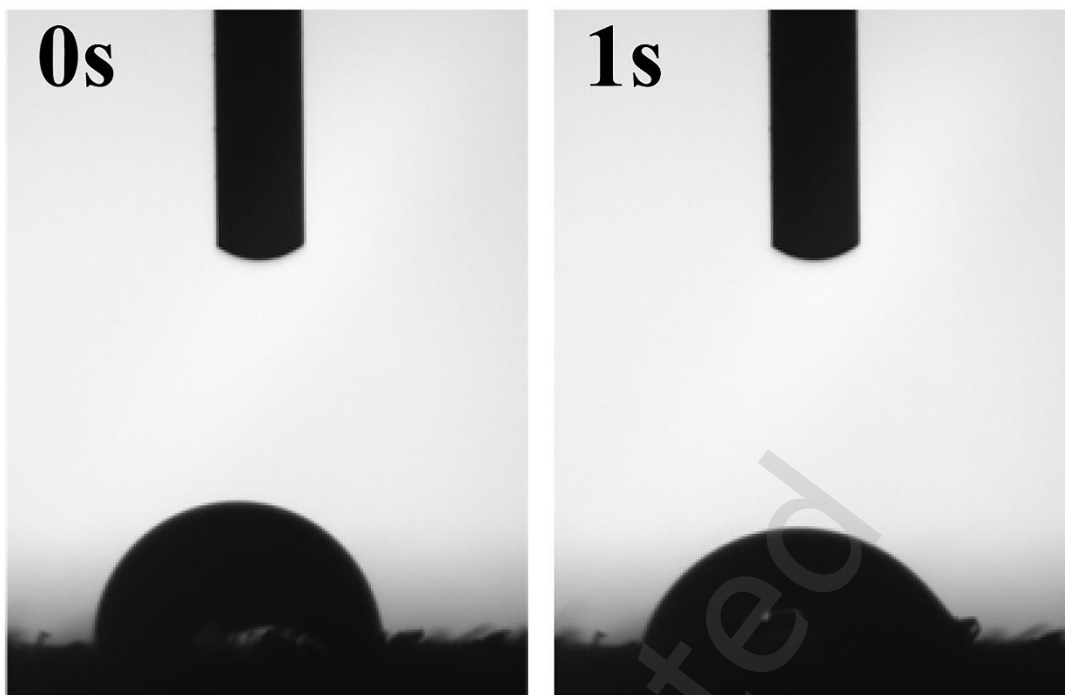


Figure S16. The contact angle test of NW.

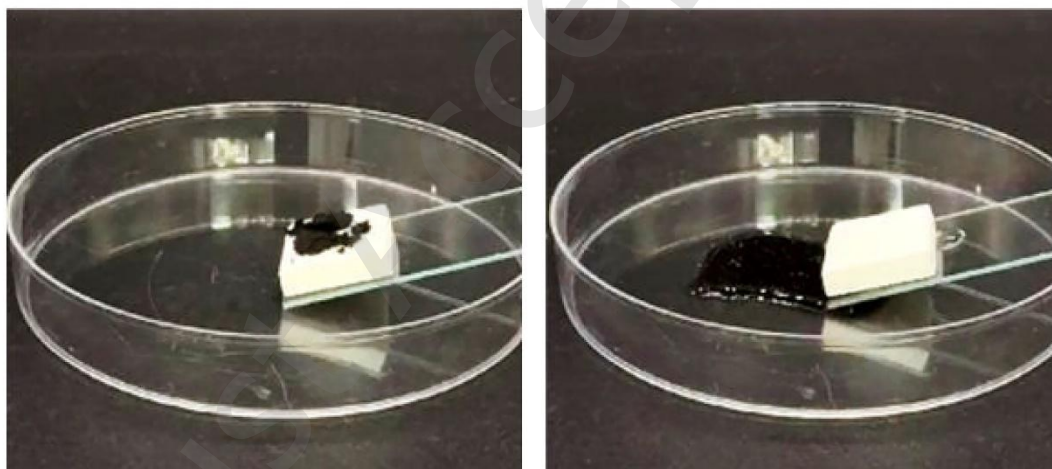


Figure S17. The self-cleaning property of WCAF.

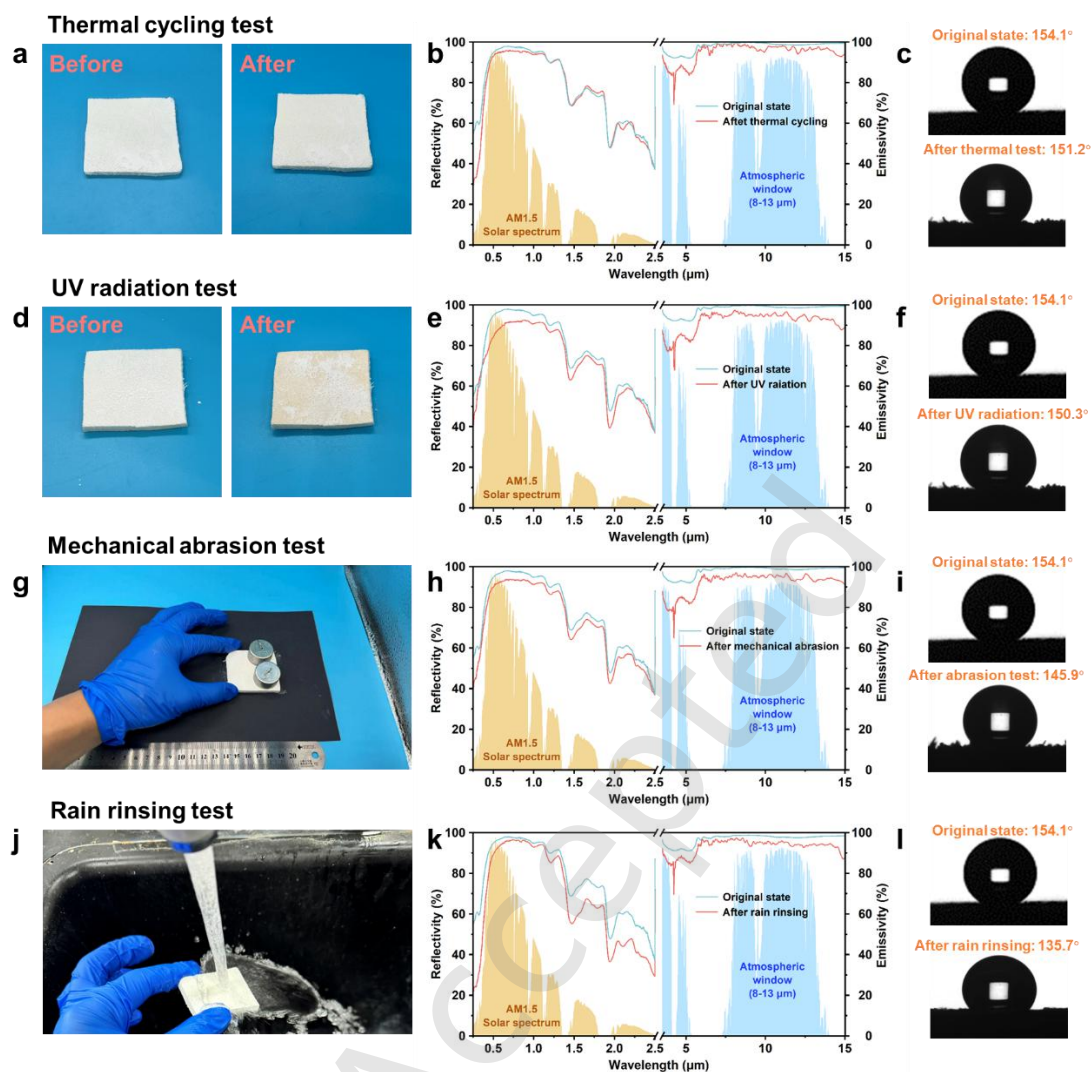


Figure S18. Retention of optical properties and hydrophobic performance of WCAF after simulated outdoor aging tests: (a, b, and c) thermal cycling test, (d, e, and f) UV radiation test, (g, h, and i) mechanical abrasion test, (j, k, and l) rain rinsing test.

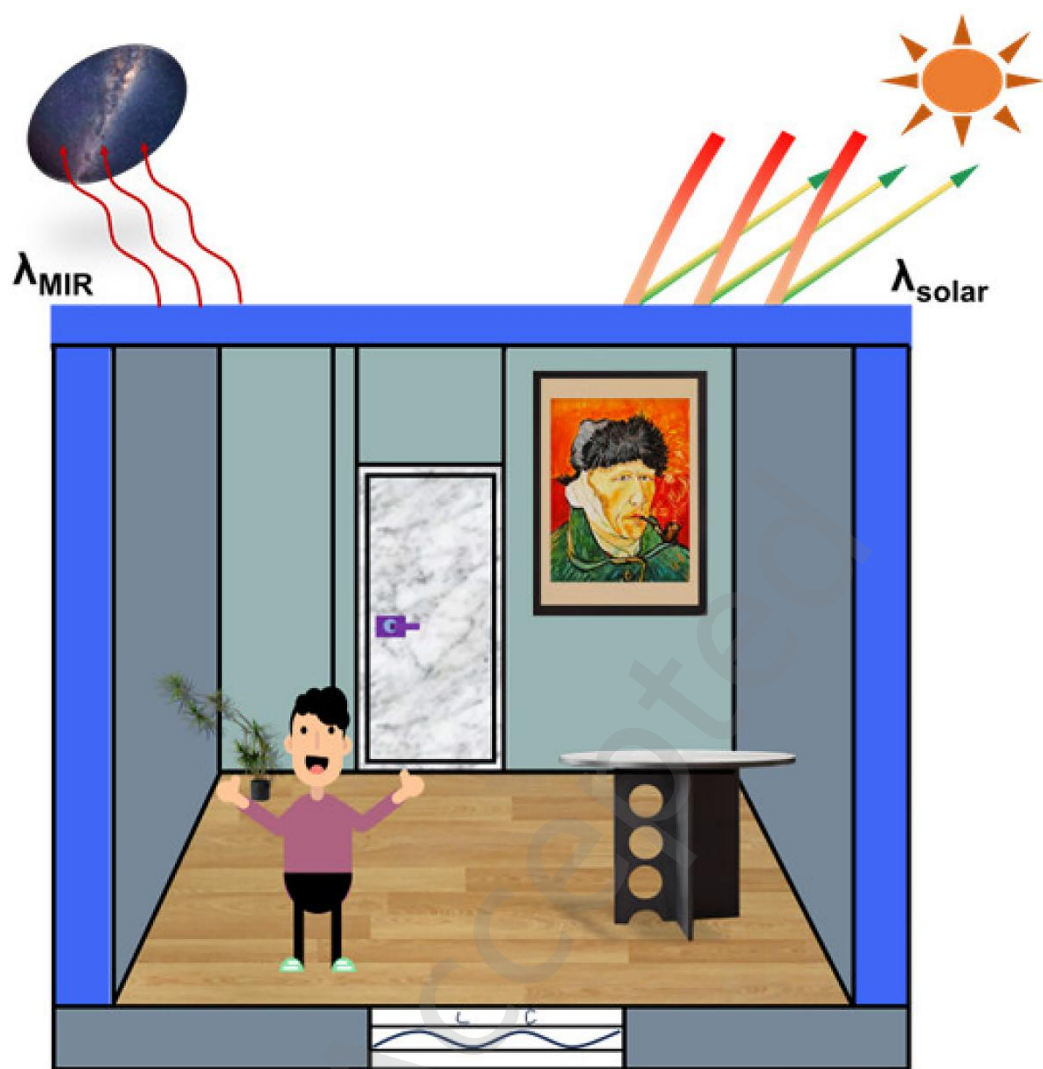


Figure S19. Scheme for energy saving buildings.

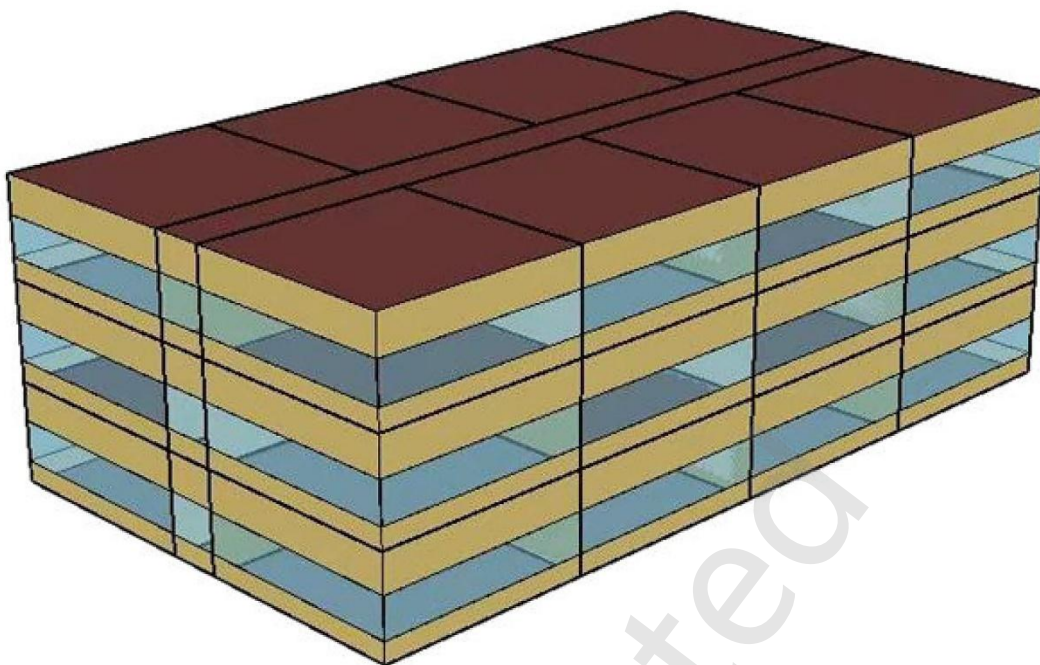


Figure S20. The model used in simulation.

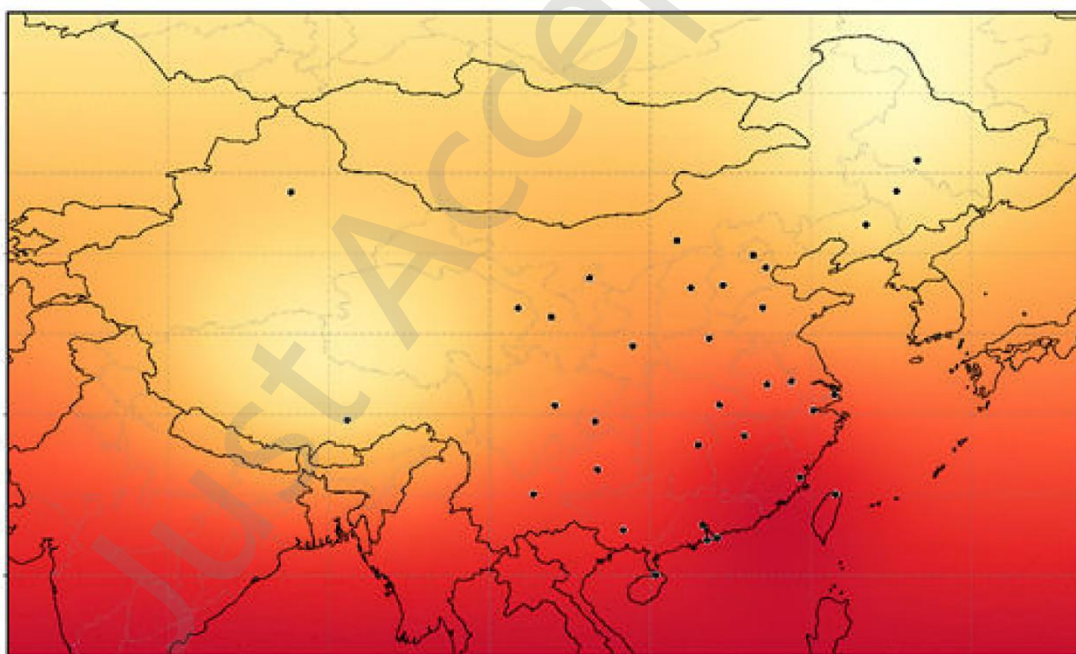


Figure S21. Heat map of china.

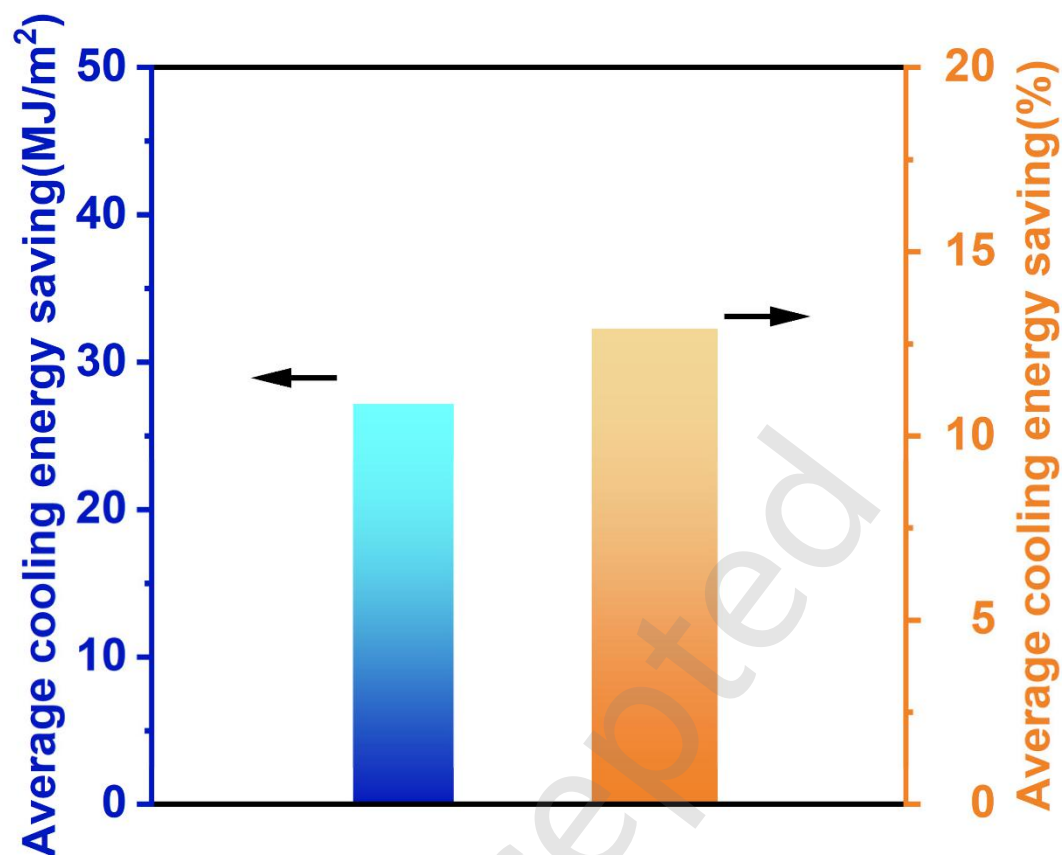


Figure S22. The average cooling savings of WCAF.

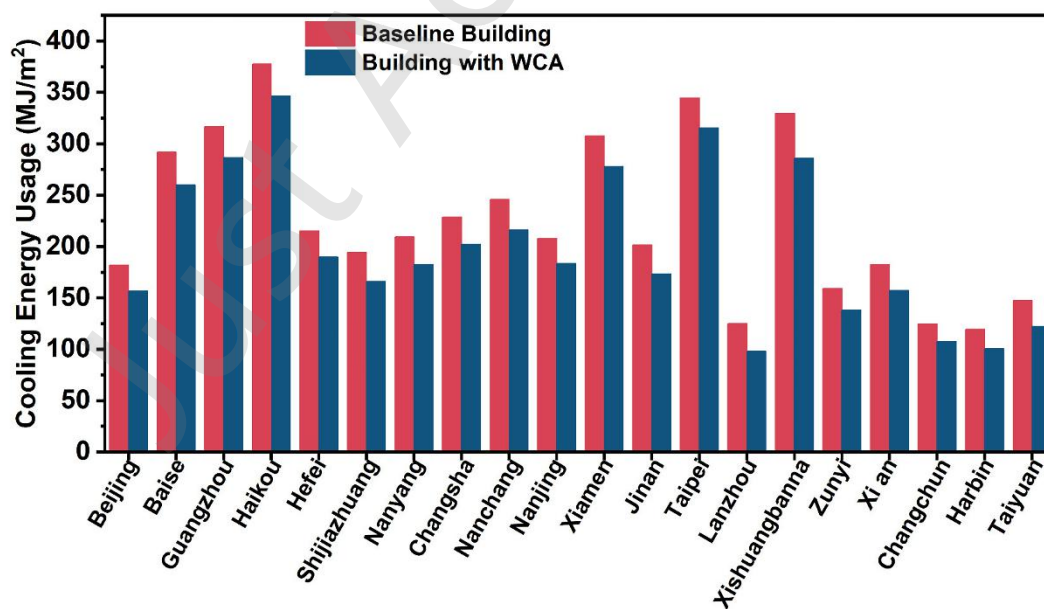


Figure S23. Cooling energy usage (MJ/m²) for the baseline building and the building covered by WCAF.

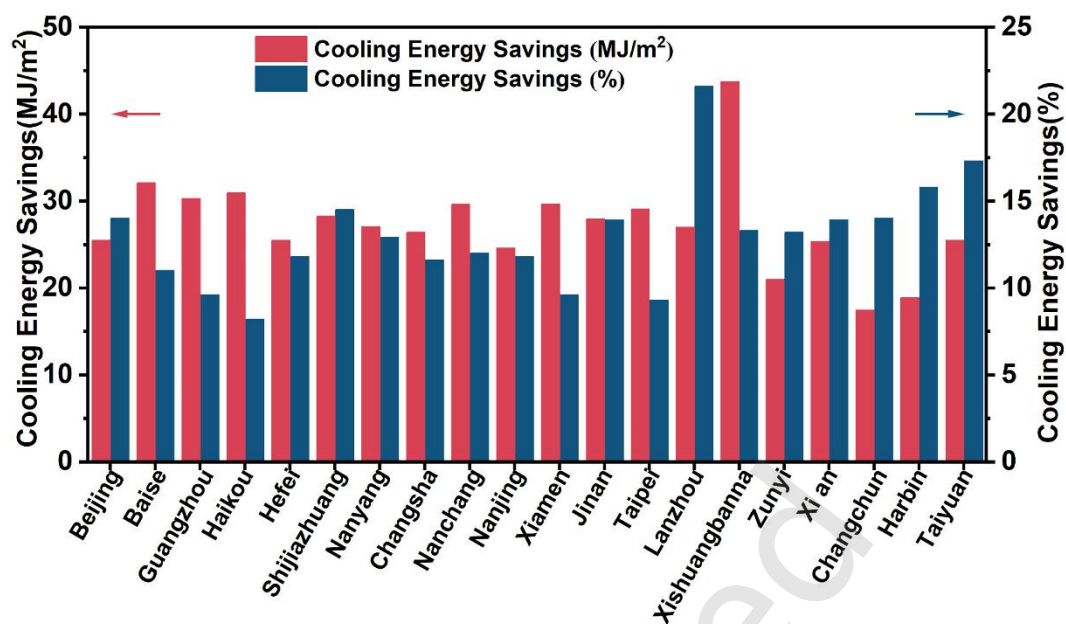


Figure S24. Energy savings for the baseline building and the building covered by WCAF.

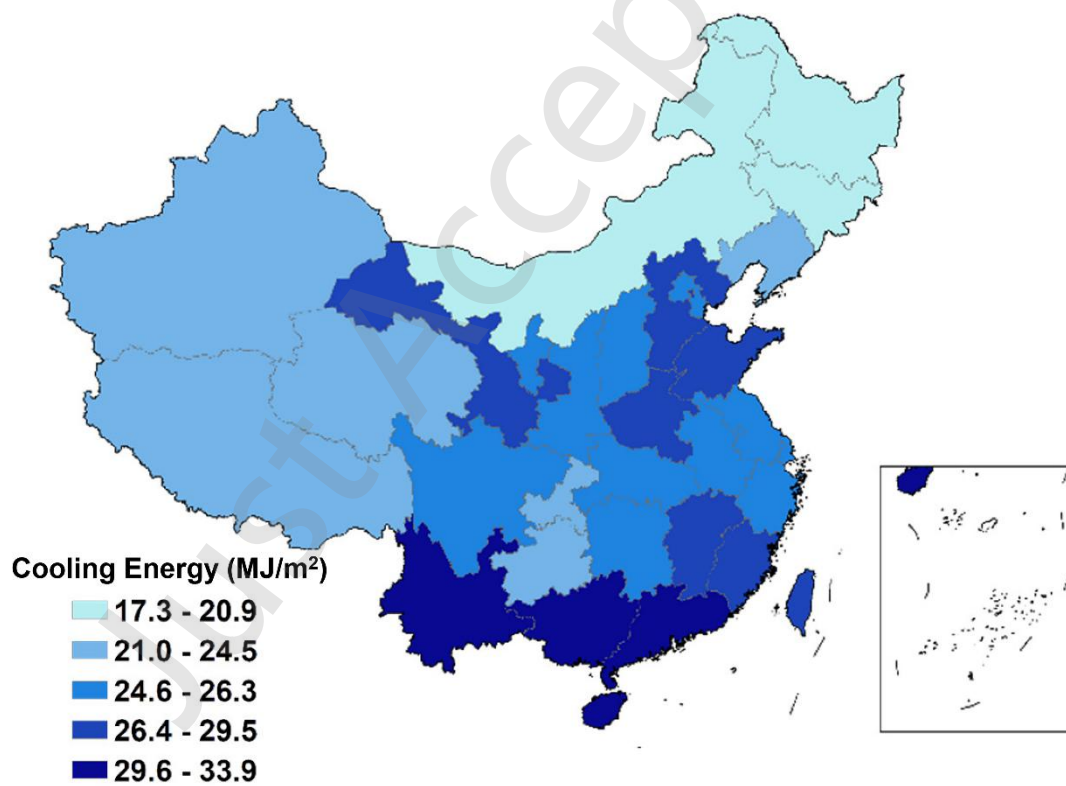


Figure S25. The predicted cooling map of China.

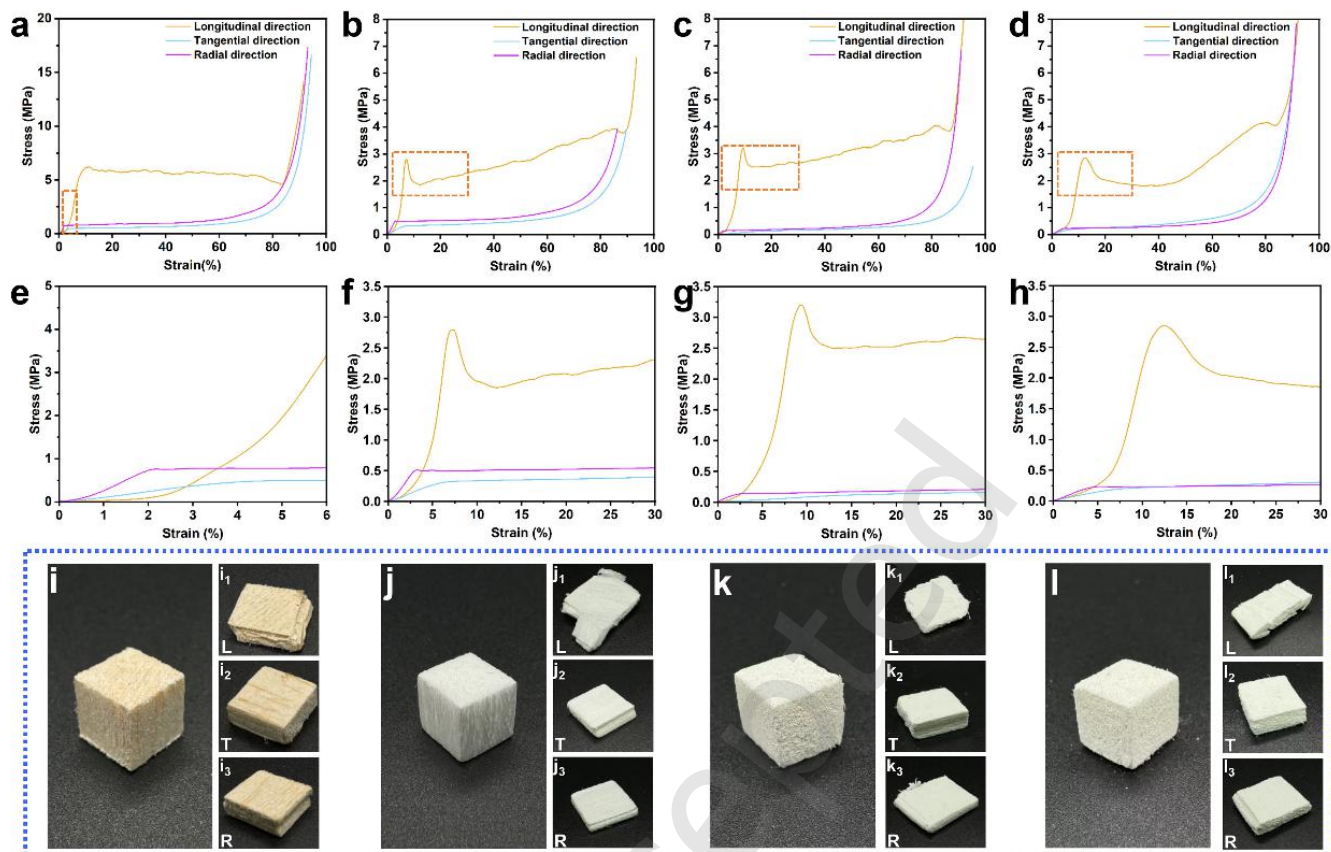


Figure S26. Mechanical anisotropy and optical images of samples before and after compression. (a-d) compressive stress-strain curves in the longitudinal, tangential, and radial directions of NW, DW, WCA, and WCAF. (e-h) Partially amplified compressive stress-strain curves in the longitudinal, tangential, and radial directions of NW, DW, WCA, and WCAF. (i-l) Appearance of NW, DW, WCA, and WCAF. (i₁-i₃, j₁-j₃, k₁-k₃, and l₁-l₃) Appearance of NW, DW, WCA, and WCAF after compression in three directions, respectively.

Table S1. The index used in performance radar chart.

Samples Performance	WCAF	CNC cooler	Cooling wood	Durable cooling wood
Cooling temperature (°C)	11.58	9.5	9	8
Superhydrophobicity (°)	154	138	150	137
Fire resisistance	64.9	0	0	10
Optical performance	94	94	88	95
Sustainability	85	60	70	60
Lightweight	94	99	0	17

Table S2. The composition and density of NW and DW.

Samples	Density(mg/cm ³)	Composition content (%)		
		Cellulose	Hemicellulose	Lignin
NW	135.2±12.5	47.8±2.4	25.2±1.1	27.0±1.2
DW	73.2±8.5	79.3±2.1	12.8±1.1	7.9±0.6

Table S3. Thermo-fluid properties of the WCAF used in the simulation.

Property	Value
Thickness [mm]	0.0253
Thermal Conductivity [W/m·K]	0.1121
Specific Heat Capacity [kJ/kg·K]	1900
Thermal Absorptance [-]	0.991145
Solar Absorptance [-]	0.0868
Visible Absorptance [-]	0.0463

Table S4. Properties of structural materials in building energy consumption simulation.

Name	Thickness (m)	Conductivity W/m·k	Density (kg/m ³)	Thermal absorptance	Solar absorptance	Visible absorptance
1/2IN Gypsum	0.0127	0.16	784.9	0.9	0.4	0.4
1IN Stucco	0.0253	0.6918	1858	0.9	0.92	0.92
8IN Concrete HW	0.2033	1.7296	2243	0.9	0.65	0.65
Wall Insulation	0.0794	0.0432	91	0.9	0.5	0.5
Roof Membrane	0.0095	0.16	1121.29	0.9	0.7	0.7
Roof Insulation	0.2941	0.049	265	0.9	0.7	0.7
Metal Decking/ Metal Roofing	0.0015	45.006	7680	0.9	0.6	0.6
G01a 19mm gypsum board	0.019	0.16	800	0.9	0.4	0.4
G05 25mm wood	0.0254	0.15	608	0.9	0.5	0.5
MAT-CC05 4 HW CONCRETE	0.1016	1.311	2240	0.9	0.85	0.85

Table S5. Region-specific grid carbon emission factors for electricity.

Province	FCO ₂ (kgCO ₂ /KWh)
Beijing	0.5580
Guangxi	0.4044
Guangdong	0.4403
Hainan	0.4184
Anhui	0.6782
Hebei	0.7252
Henan	0.6258
Hunan	0.4900
Jiangxi	0.5752
Jiangsu	0.5978
Fujian	0.4092
Shandong	0.6410
Taiwan	0.5366
Gansu	0.4772
Yunnan	0.1073
Guizhou	0.4989
Shanxi	0.6558
Jilin	0.4932
Heilongjiang	0.5368
Shanxi	0.7096

Table S6. Energy usage in Baseline Building.

Energy Cities	Cooling (kWh)	Lighting (kWh)	Equipment (kWh)	Heating (kWh)
Haikou	181113.6	59575	62655.6	0
Guangzhou	152054.4	59575	62655.6	76.8
Changsha	109651.2	59575	62655.6	4459.2
Nanjing	99801.6	59575	62655.6	5760
Changchun	59908.8	59575	62655.6	57600
Harbin	57302.4	59575	62655.6	67833.6

Table S7. Energy usage in Building with WCAF.

Energy Cities	Cooling (kWh)	Lighting (kWh)	Equipment (kWh)	Heating (kWh)
Haikou	166241.7	59575	62655.6	0
Guangzhou	137582.4	59575	62655.6	110.4
Changsha	97036.8	59575	62655.6	5112
Nanjing	88056	59575	62655.6	7454.4
Changchun	51556.8	59575	62655.6	62668.8
Harbin	48292.8	59575	62655.6	73574.4

Table S8. Thermo-fluid properties of the WCA used in the simulation.

Property	Value
Thickness [mm]	0.0253
Thermal Conductivity [W/m·K]	0.1119
Specific Heat Capacity [kJ/kg·K]	1900
Thermal Absorptance [-]	0.992026
Solar Absorptance [-]	0.0839
Visible Absorptance [-]	0.0424

Table S9. Comparison of cooling performance, compression strength, fire resistance and hydrophobicity.

	Cooling performance	Compression strength	Fire resistance	hydrophobicity
WCAF (This work)	11.58 °C	2.85Mpa	√	154°
SiO ₂ /PVA ⁶	14.1 °C	Not mentioned	√	×
L-D-2 ⁷	3.5 °C	Not mentioned	×	152°
CS-MA/PA-30 wt% ⁸	4.3 °C	314Kpa	√	×
GE-DNA ⁹	16 °C	Not mentioned	√	×
CNC aerogel cooler ¹⁰	9.2 °C	Not mentioned	×	138°
sANA ¹¹	3.3 °C	Not mentioned	×	143.5°
GMS0.6 ¹²	4.8 °C	679Kpa	√	√
AFTPU-10 ¹³	7.7 °C	Not mentioned	×	126°
P4-G1 ¹⁴	22.7 °C	Not mentioned	×	Not mentioned
FCCA ¹⁵	8.2 °C	624.8Kpa	×	148°
FhNTs ₄₀ /(FSiO ₂) ₁₀ ¹⁶	12.8 °C	Not mentioned	×	154.75°
PC metacotton fabric ¹⁷	8.8 °C	Not mentioned	×	159.7°

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