

ENGINEERING

Scalable metasurface-enhanced supercool cement

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Structural materials with the capability for passive daytime radiative cooling (PDRC) show promise for the sustainable cooling of buildings. However, developing durable PDRC structural materials with optical robustness, ease of deployment, and scalability remain a challenge for civil engineering applications. We synthesized a metasurface-enhanced cooling cement using a universal, scalable pressure-driven fabrication strategy during a low-carbon production process. The self-assembly of multiple-sized reflective ettringites as main hydration products toward the metasurface, coupled with hierarchical pores, guaranteed high solar reflectance (96.2%), whereas raw materials containing alumina- and sulfur-rich function groups leveraged inherent mid-infrared emissivity (96.0%). This photonic-architected cement achieved a temperature drop of 5.4°C during midday conditions with a solar intensity of 850 watts per square meter. This supercool cement featured intrinsic high strength, armored abrasive resistance, and optical stability, even when exposed to harsh conditions, such as corrosive liquids, ultraviolet radiation, and freeze-thaw cycles. A machine learning-guided life-cycle assessment indicated its potential to achieve a net-negative carbon emission profile.

INTRODUCTION

Buildings contribute nearly 40% of global energy consumption and 36% of carbon emissions throughout their life cycle, with the operational stage as the largest contributor and a key focus of research. During the operational stage, the need for space cooling has been growing rapidly due to global warming, population growth, and rising living standards. The ensuing reliance on energy-intensive air-conditioning systems is therefore projected to account for 30 to 50% of peak electricity demand and could triple related carbon emissions by 2050 (1–3). Action on energy-efficient building cooling is therefore urgently required. Radiative cooling has emerged as a substantial strategy by reflecting sunlight and emitting thermal radiation through atmospheric window without additional energy input. Integrating radiative cooling technology into terrestrial structural materials, such as pavements and roofs, can potentially reduce building cooling energy needs by up to 60% and offset ~44 billion tonnes (Gt) of CO₂ emissions globally by enhancing urban albedo (4, 5), thereby promoting sustainability.

However, implementing daytime radiative cooling in buildings is challenging due to the overheating by solar radiation under direct sunlight (6–8). Current methodologies, including optical metamaterials and nanophotonic designs with sophisticated microstructures, enable light-matter interaction for high solar reflectance and long-wave infrared emissivity (9, 10). Nevertheless, transitioning these technologies from laboratory to real-world applications faces challenges due to the high costs and limitations in durability and constructability (11). Random photonic structures, such as polymeric films (12–15) and coatings (16–18), also struggle with durability

challenges (19). All-inorganic photonic ceramics (primarily Al₂O₃ particles) (4, 20) have been engineered for high radiative cooling performance and environmental stability. However, these coatings may flake off due to insufficient adhesion to the substrates, and the high-temperature post-sintering fabrication process would lead to excessive energy input. It remains a substantial challenge to fabricate scalable and durable radiative cooling structural materials that meet the criteria of low-carbon, high-strength, and structural integrity in civil engineering.

Cement, with a production of 4.1 Gt in 2023 (21), far exceeding that of other building materials (~50%; fig. S1), is the most widely used man-made material globally (22). Despite this, cement's cost-effectiveness, mechanical robustness, and high emissivity from infrared-active groups (23–26) position it as a promising candidate for radiative cooling applications in operational stage as well. However, the production of cement is another key contributor influencing the life-cycle sustainability of buildings, which is responsible for ~8% of global CO₂ emissions. Urgent decarbonization is therefore crucial for sustainable production practices. Furthermore, during the later operational stage, cement products' relatively low solar reflectance ($R_{\text{solar}} \sim 30\%$) leads to considerable heat absorption and undesired overheating, which would further exacerbate air-conditioning energy consumption. Current approaches to enhancing cement's radiative cooling performance rely heavily on incorporating functional additives or non-cementitious coatings. These methods overlook cement's intrinsic potential and inherent advantages, which compromises the structural integrity and durability, thereby limiting their suitability for large-scale engineering applications. Therefore, harnessing the intrinsic radiative cooling potential of cement while addressing the limitations of its solar reflectance is essential for achieving sustainable building cooling in a large-scale perspective.

In this work, we developed an intrinsically supercool cement particle that requires no fillers or additives to achieve its cooling properties. We further nanoengineered the material via a universal cavitation strategy, creating a cement-based radiative cooler with integrated optical metasurfaces (defined as a supercool cement; Fig. 1, A and B). The

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self-assembly of highly reflective ettringite crystals on metasurfaces and hierarchically distributed nano/micropores (Fig. 1C) leverage its high R_{solar} (96.2% on average). Functional bonds (Fig. 1D) from suitable raw minerals guarantee inherent blackbody-like emissions (96.0%). This nanoarchitected supercool cement could overcome the issue of scalability (Fig. 1, E and F), achieving robust radiative cooling performance (Fig. 1, G and H) and fulfilling the urgent engineering demand of manufacturability and durability for sustainable building cooling (Fig. 1I).

RESULTS

Bottom-up strategy to design cool matrix

To design an additive-free, hierarchically structured radiative cooling matrix, we adopted a bottom-up strategy, beginning with the careful selection of calcium-rich alumina-sulfur-silicate compounds for the cement clinkers (figs. S2 and S3). Using a custom phase diagram (fig. S4), we optimized the calcination process (fig. S5) to eliminate impurities while retaining key elements—Ca, Al, Si, and S—that are essential for radiative cooling properties (fig. S6 and table S1). This

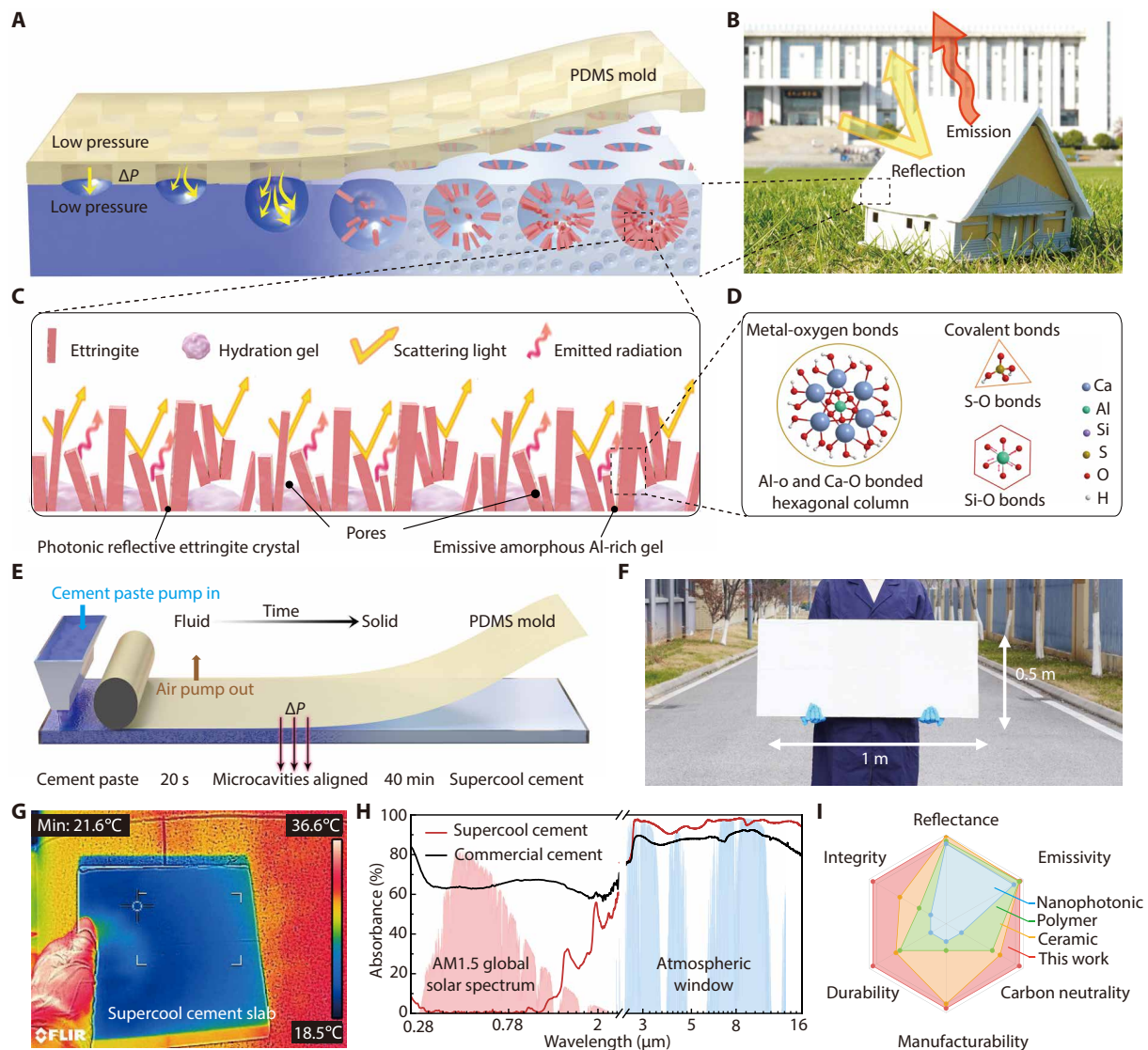


Fig. 1. Scalable structural supercool cement. (A) Schematic of the surface engineering strategy for supercool cement during the hydration process, consisting of 20-s air bubble expansion and 40-min air cavity stabilization for crystal accumulation. (B) Illustration of supercool cement for building roofs with high emission and reflection. (C) Self-assembly of the highly reflective ettringite crystal hydration products on the microcavity surfaces due to the microscale free space for atom nucleation via metasurface engineering. (D) Abundant infrared-active vibration modes by Al-, Ca-, and S-rich functional groups in hydration products for high mid-infrared emissivity. (E) Scalable and simple fabrication illustration. (F) Nanoarchitected supercool cement slab. (G) Infrared image demonstrating the low surface temperature of the supercool cement (21.6°C) under direct sunlight compared to the ordinary building surface in the background. (H) Spectral absorbance [$R(\lambda) = 100\% - \epsilon(\lambda)$] of the as-fabricated supercool cement against AM 1.5 global solar spectral irradiance and the atmospheric transparency window. R_{solar} (96.2%, $\lambda \sim 0.25$ to $2.5 \mu\text{m}$) and ϵ_{LWIR} (96.0%, $\lambda \sim 2.5$ to $16 \mu\text{m}$) served as breakthrough optical properties for cementitious materials compared to commercial gray or white cement composites. Transmittance was set as zero due to the thickness reaching 15 mm. (I) This versatile metasurface-enhanced supercool cement exhibited environmental durability, manufacturability, structural integrity, and carbon neutrality.

approach enhanced both the purity and functionality of the clinkers and set the foundation to form a stable, effective cooling matrix during hydration. Consequently, these clinkers reacted rapidly with water to form early-generated ettringite crystals and amorphous gels as the skeleton and the glue in a stable structural framework (figs. S7 to S10). The hydration products (table S2), enriched in Al and S, displayed infrared-active vibrational modes that effectively triggered mid-infrared emissions like a blackbody radiation, thus yielding high long-wave infrared emissivity (ϵ_{LWIR}) (figs. S11 and S12). In addition, the activated formation of ettringites ensured high whiteness in appearance, excellent solar reflectance (R_{solar}) especially in the visible range, and a moderate bandgap (~ 5.32 eV), preventing ultraviolet (UV) absorption while promoting efficient light scattering, thus providing a solid foundation for the development of supercool cement (figs. S13 to S17).

This bottom-up strategy focuses on designing the cooling matrix from the ground up by selecting and optimizing the composition of the cement clinker. Unlike approaches that rely solely on functional fillers or external additives, our method inherently controls and integrates material properties at the fundamental level, yielding an additive-free, hierarchically structured matrix with an unprecedented synergy in radiative cooling performance.

Metasurface-enhanced supercool cement

In the cool cement matrix, the amorphous gel leveraged cement's inherent high ϵ_{LWIR} as a blackbody-like emitter. However, this heat-absorbing gel tended to envelope the highly reflective ettringite crystals, thereby limiting the overall R_{solar} . To mitigate the masking effect and amplify the role of photonic ettringite crystals, we optimized the distribution and composition of amorphous and crystalline phases within the matrix by a gas difference-driven strategy. Taking the in-place fabrication of a structural element as an example in this dual mold-reversed process (fig. S18), the cooling cement paste (Fig. 2A) was cast into a base mold (Fig. 2B) and then overlaid with a single-patterned polydimethylsiloxane (PDMS) foil featuring concaved microcylinders as a secondary textured mold (fig. S19). Upon applying a negative pressure, air bubbles formed at the gas-liquid interfaces due to the pressure difference between the cylindrical cavities and the cement paste (Fig. 2C and fig. S20) and became stabilized permanently as microcavities. By adjusting key factors during the cavitation, the size and spacing of each cavity could be precisely controlled (tables S3 to S6). The recyclable PDMS mold could then be easily peeled off during demolding (Fig. 2D). The setting time of supercool cement is highly tunable across a wide range (fig. S36), allowing both rapid setting within 10 min (movie S1) and early-strength properties suitable for future roll-to-roll fabrication. Unlike conventional texturing methods (e.g., mechanical engraving, chemical etching, and photolithography), which often involve costly equipment, high energy consumption, or hazardous chemicals, our gas difference-driven cavitation strategy provides a scalable, energy-efficient approach compatible with standard cement hydration, enabling precise microscale structuring.

To further optimize the optical performance of the supercool cement, we adjusted the diameter and spacing of the microcavities on the metasurface by using PDMS molds with different dimensions. From the side view, the as-fabricated metasurface consisted of hexagonally aligned concave microcavities with controllable diameters and spacings, which were consistent with PDMS microcylinders' dimensions (e.g., a cavity diameter of 35 μm and spacing of 50 μm

matched a microcylinder of similar size; Fig. 2E). Given the typical size of cement particles (usually up to several tens of micrometers), a cavity diameter of 35 μm challenged the smallest scale for orderly microstructuring on the cement surface, which still provided sufficient microscale free space for ettringite's surface-oriented assembly (figs. S21 and S22). According to the free energy theory, during cement hydration, the microcavity surface, acting as a crystal nucleus, offered suitable gas-cement interfaces where ions (Al^{3+} , Ca^{2+} , SO_4^{2-} , and OH^-) could spontaneously aggregate for further ettringite crystallization and recrystallization (27). Without surface modification (i.e., conventional fabrication; Fig. 2F), reflective ettringites are typically embedded in the amorphous gel matrix. After surface modification, however, randomly distributed rod-like ettringite crystals spontaneously assembled on the surface and turned into metasurface (cavity diameter of 35 μm ; Fig. 2G). Quantitative analysis and size measurements revealed that more reflective ettringites accumulated with cross-sectional dimensions between 0.1 and 0.6 μm (Fig. 2H and figs. S23 and S24), which could interact with the light of equivalent scale and induce stronger scattering (fig. S25), as further corroborated by the simulations results (Fig. 2I and fig. S26) (28, 29). Pore-size measurements (Fig. 2J) indicated that, in supercool cement, abundant nano/micropores could efficiently promote light scattering and R_{solar} by creating high refractive index contrast at solid-gas interfaces (30). Notably, the supercool cement constituted a continuous Al-, Ca-, and Si-rich inorganic network with reflective nanoarchitectures and hierarchical pores, exhibiting high R_{solar} (96.2% on average) and ϵ_{MIR} (96.0%) in an ultrabroad infrared band (fig. S28).

Engineered with metasurfaces, the supercool cement is designed as a matrix-directed radiative cooling material suitable for use as both a radiative cooler and a structural material in buildings for roofs and walls (fig. S29). This metasurface engineering strategy offers a universal solution that is applicable even for conventional commercial Portland cement, which also takes on a surface ettringite-enrichment manner toward higher R_{solar} (fig. S30). However, the enhancement in R_{solar} was limited due to the insufficient ettringite content. Particularly in gray Portland cement, which contains more hazardous iron (Fe) element, inappropriate diameter and spacing choices of the metasurfaces would not induce the assembly of reflective crystals, thus failing to enhance R_{solar} . Beyond radiative cooling application, this strategy was applicable to even more cement systems and yield-stress fluid with time-dependent rheology for complex surface engineering. In terms of sustainability, this method can be extended to other binder systems such as slag-based cement or alkali-activated geopolymers, which are of increasing interest for low-carbon construction.

Versatility and aesthetic

In radiative cooling research, durability is a critical factor influencing long-term stability and cost-effectiveness. Supercool matrix intrinsically has robust mechanical strength [ASTM C109 (31) and ASTM C348 (32)] due to the amorphous gel generated through hydration ($\sim 50\%$) as a strong binder. It demonstrated compressive strength higher than 100 MPa and flexural strength higher than 12.0 MPa, which is far beyond traditional building materials and both increased with time (figs. S31 to S33). In addition, the concave structure on the surface acted as armor to protect the functional structures inside against abrasion (Fig. 3A and fig. S34), which agreed well with those from finite-element (FE) simulations (Fig. 3,

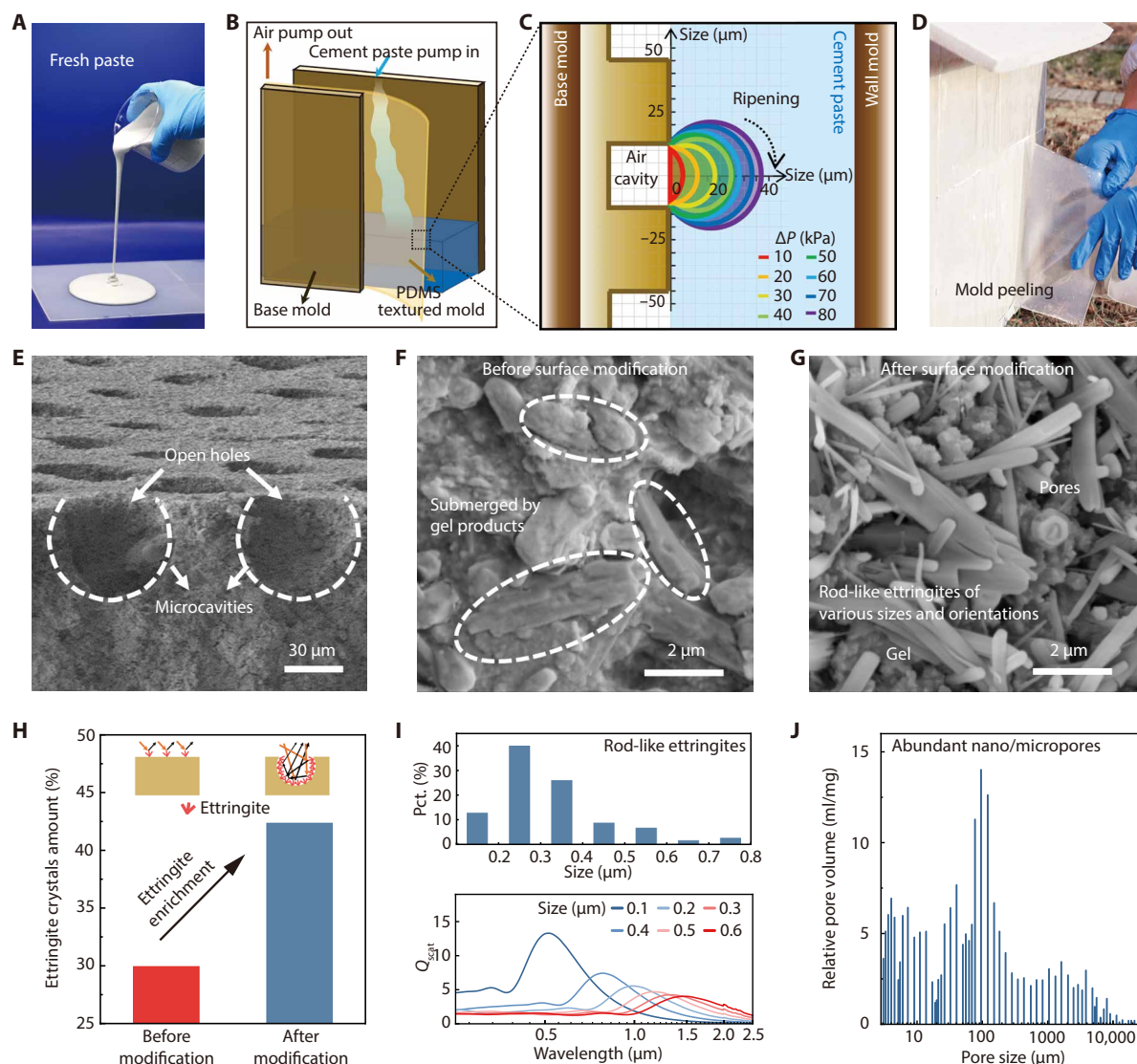


Fig. 2. Fabrication and morphology of supercool cement with engineered metasurfaces. (A) The good flowability of the fresh cement paste guaranteed flexible manufacturability for real-world construction. (B) Illustration of the cast-in-place wall fabrication using a wall and PDMS mold. (C) Controllable bubble evolution by a pressure difference–driven cavitation strategy to create bubbles with controllable sizes. (D) The recyclable PDMS mold could be easily peeled off from the fabricated metasurfaces. (E) Side view of the supercool cement with well-designed metasurfaces consisting of open holes and concave chambers in SEM images. (F) Submerged crystals in gels without metasurface modification. (G) Outside-oriented and overlapped crystal ettringites beneath the amorphous gel with surface modification. (H) Differences in surface structures and ettringites contents before and after the metasurface were engineered in the surface/near-surface region. (I) Cross-sectional size distribution of rod-like ettringites on the surface and the simulated scattering efficiency of ettringites with corresponding cross-sectional sizes. Ettringites of different sizes comprehensively scattered all solar wavelengths, resulting in high reflectance. (J) Size distributions of the nano/micropores in the composites in the range from several nanometers to 10 μm .

B and C, and fig. S35). Thus, mechanical robustness and radiative cooling performance are not compromised but rather synergistically enhanced. By tuning the solidification time of supercool cement in a wide range of time (see fig. S36), it can be easily applied to existed building surfaces due to its rapid setting. Supercool cement, when cast as a coating, exhibits high adhesive strength to various building surfaces, including concrete, metals, and ceramic tiles [Fig. 3D, fig. S37 and movie S2; ASTM D4541 (33)].

The all-inorganic composition, dense microstructure, and ettringite-enriched framework synergistically contribute to long-term stability across diverse climates and service environments. Supercool cement

remained optically stable after the 30-day immersion tests in corrosive liquids with different pH values (Fig. 3E and fig. S38), namely, H_2SO_4 solution (pH = 3.5), MgSO_4 (pH = 7.0), seawater (pH = 8.4), and NaOH solution (pH = 13.0), which were otherwise harmful to ordinary inorganic cement and organic structural materials, such as wood, polymeric films, and coatings (figs. S39 to S41). Our supercool cement exhibited high stability after 1080 hours of UV accelerated weathering test (equivalent to 6 months of natural UV exposure) [ASTM G154 (34)], with only a slight drop in R_{solar} and ϵ_{LWIR} (Fig. 3F and fig. S42). We conducted a freeze-thawing test to simulate the severe circumstances buildings might face. After immersion in NaCl solution (3 wt %)

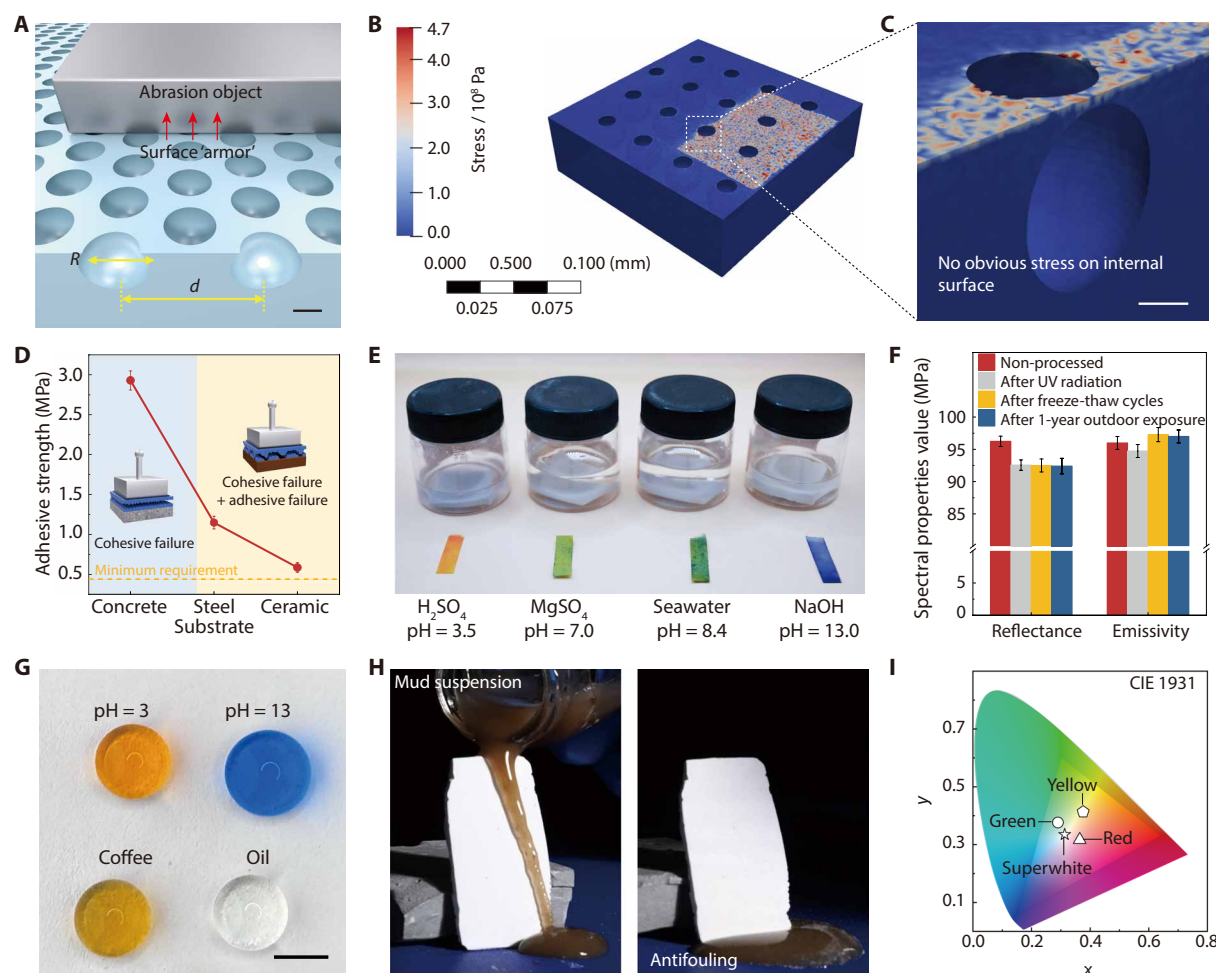


Fig. 3. Versatile performances in mechanical robustness, environmental stability, and aesthetic design. (A) Illustration of the side view of the concave microcavities as armor to protect the interior ettringites against abrasion forces. Scale bar: 20 μm . (B) Simulated stress distribution of the supercool cement surfaces. (C) Enlarged stress distribution, demonstrating that abrasion would not affect the surface inside the microcavity. Scale bar, 10 μm . (D) Adhesive strength results of supercool cement on different substrates (concrete, steel, and ceramic tile), which are all higher than the minimum requirements (0.4 MPa) in the standard. Supercool cement shows robust adhesion with concrete, leading to a cohesive failure in the test, whereas with steel and ceramic tile, the failure type is partially adhesive failure. (E) Images of the supercool cement immersed in corrosive liquids with different pH values. (F) R_{solar} and ϵ_{LWIR} changes before and after selected environmental influence, including UV radiation and freeze-thaw cycle. The reflectance showed little change. Error bars represent means $\pm$ SD. (G) Stain drops (acidic solution, alkaline solution, coffee, and oil) in air, demonstrating robustness of the amphiphobic property. Scale bar, 2 mm. (H) Demonstration of antifouling properties of the supercool cement with hydrophobicity against muddy water. Scale bar, 15 mm. (I) Chromaticity of the colored supercool cement. According to the color inside the circle, reflectance could maintain $>90\%$.

for 50 cycles of temperature changes between 3° and -16°C , R_{solar} remained nearly unchanged, and the emissivity even increased to 97.28% due to the saturated state, exhibiting good durability in building's service life (figs. S42 and S43). In addition, a full-scale panel exposed to natural outdoor conditions for 1 year exhibited negligible degradation (fig. S44). Such durability makes the supercool cement a strong candidate for scalable and resilient building envelope applications.

To avoid degraded optical properties due to surface contamination (fig. S45), a supercool cement with amphiphobicity and antifouling properties (Fig. 3, G and H, and fig. S46) was obtained by the integration of functional fluoroalkylsilane and SiO_2 particles (~ 30 nm).

For aesthetic reasons, we modified the color appearance of the supercool cement (yellow, green, and red) by mixing the unhardened paste with inorganic phosphor dyes [5 wt %, Ce:YAG,

$\text{MgAl}_{11}\text{O}_{19}:(\text{Ce},\text{Tb})$, and $\text{CaAlSiN}_3:\text{Eu}^{2+}$]. Our colored supercool cement simultaneously achieved high reflectance up to 90% and all colors were in the s-RGB gamut color space (Fig. 3I and fig. S49). We also investigated the trade-off between visual appearance and radiative cooling performance by simulating the net cooling power at various temperature differences. The results revealed that even colored supercool cements with lower reflectance ($\sim 90\%$) can retain a net cooling effect under full sun. This aesthetic appeal along with cement's plastic nature (fig. S50) could offer more creativity for complex scenarios in advanced civil engineering.

A comparative benchmarking of the supercool cement against both conventional cementitious materials and other classes of passive daytime radiative cooling (PDRC) materials (data S1) promoted the supercool cement as a versatile radiative cooling candidate. According to detailed techno-economic analyses (Supplementary Text and tables S16 and S17), these results underscore the practical

viability of integrating supercool cement into existing building systems with a competitive cost-performance ratio. Further improvement in infrastructure efficiency and reduction in cost is anticipated with potential roll-to-roll automated production, integration into precast panel workflows, and development of low-cost micropatterning substrates.

Worldwide life-cycle assessment toward CO₂ emission reduction

The high R_{solar} and ϵ_{LWIR} enabled efficient building cooling. An in-field test measuring the temperature drop was conducted on a Purdue University campus roof in West Lafayette, USA (40°25'21" N,

86°55'12" W) from 30 July 2023 to 31 July 2023 (Fig. 4, A and B). At noon, when the solar irradiance was the highest (~850 W/m², between 1 and 2 p.m.), a temperature drop of 5.4° and 26.0°C occurred compared to ambient temperature (38.4°C) and commercial cement (59.0°C), respectively. The wind speed was 0.98 m/s and the relative humidity was 38.5% (fig. S51). At night, between 9 and 10 p.m., the temperature drop was 7.0°C compared to the ambient temperature (28.2°C) at a wind speed of 0.16 m/s and relative humidity of 61.8%. On the basis of radiative cooling principles (fig. S52), and considering the influence of wind speed (fig. S53) (35), we estimated that the net cooling power of the supercool cement approximated 96 W/m² at noon. Both field test results and infrared images (fig. S54) verified

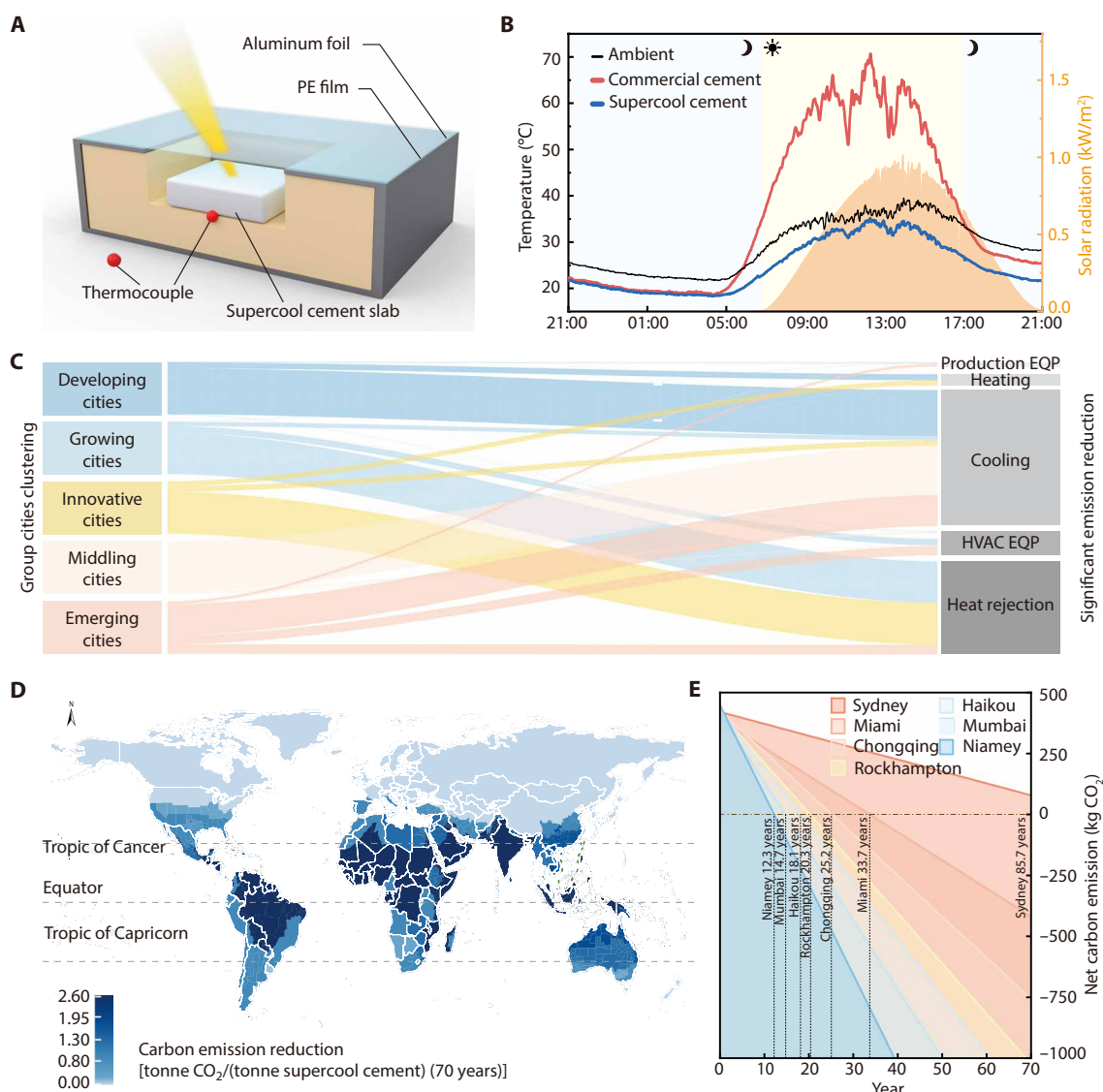


Fig. 4. Real-world cooling performance of scalable supercool cement and LCA toward carbon emission reduction. (A) Apparatus of in-field radiative cooling performance. (B) Measured temperature data of the supercool cement under direct sunlight compared to ambient and commercial Portland cement composites. The outdoor field test was conducted on the campus of Purdue University, West Lafayette, USA (40°25'21"N, 86°55'12"W) from 9 p.m. on 30 July 2023 to 9 p.m. on 31 July 2023. (C) The Sankey diagram showed the sensitivity analysis of the LCA results, which reflected the contribution each module made toward net total carbon emission values. For all five city clusters, the carbon emissions in the use stage, especially for cooling, were substantial in terms of net total carbon emission values. (D) CO₂ emission reduction considering the LCA from production to operation stage (compared to commercial cement composites). (E) Years required for the six chosen cities (Niamey, Mumbai, Haikou, Rockhampton, Chongqing, and Miami) to gain zero or even negative emissions when using supercool cement envelopes.

the all-day radiative cooling performance of the supercool cement when applied in urban buildings. In terms of denser urban environments, the sky view factor should be considered for vertical surfaces due to the radiative heat exchange between adjacent buildings (36).

Large-scale industry-compatible fabrication can be achieved in both the production process of novel clinkers and metasurface-enhanced cement products. First, the fabrication of supercool cement clinkers was applicable to cosmically industrial production by using existing calcination equipment such as ball mills and rotary kilns (fig. S55). A preliminary commercialization has been achieved, enabling a transition from laboratory-scale to commercial-scale production. For supercool cement production, both large-scale cast-in-place (Fig. 2B) and prefabricated applications (Fig. 1E) can achieve extended dimensions while maintaining desired uniformity in metasurfaces (figs. S56 and S57 and movies S3 and S4). These materials can serve as coatings for existing building envelopes or function as structural components for future sustainable construction (fig. S58). In addition, on the basis of their time-dependent and adjustable theological properties, the construction methods can be further integrated with advanced roll-to-roll processes. By the gradual deployment of flexible PDMS molds on a roll as a substrate, metasurfaces could be continuously and efficiently (fig. S59). For future development, the metasurface-enhanced strategy and this supercool cement can integrate with photovoltaic systems to offer an opportunity to combine passive cooling and solar energy harvesting.

Scalable cooling envelopes can exert a negative radiative force in cities and assist in mitigating related carbon emissions resulting in the greenhouse effect (37). To evaluate the potential carbon emission reduction performance of buildings with cooling envelopes, we performed the life-cycle assessment (LCA) from a cradle-to-grave perspective (38, 39) toward cement industry on a comprehensive landscape. Inside the system boundary of four stages (40, 41, 42) (fig. S60), 15 modules (P1 to P15; fig. S61) representing different carbon emission sources were considered based on the collected fundamental data (tables S9 and S10 and data S2) and carbon emission factors (tables S9 and S10). In the production and construction stage (P1 to P8), the fabrication of novel cement clinkers led to a reduction in the equivalent carbon emission (CO_2e) by ~25% compared to that of commercial Portland cement (735 kg CO_2e /tonne from Chinese standards) due to the decreased calcium carbonate content in raw minerals and less fuel burned (~200°C lower sintering temperature). Supercool cement clinker demonstrates the best CO_2 emission reduction capability among various construction binders during production (table S11), demonstrating notable potential in advancing the path toward carbon neutrality. In the use stage (P9 to P15), we used EnergyPlus to model the energy consumption within two different building types (office and apartment use; fig. S62) in over 100 worldwide cities. We set conventional gray Portland cement-based buildings as the benchmark and then calculated the individual carbon emission and emission reduction between cooling buildings and conventional buildings by changing the optical properties of roofs and walls (table S12) derived from measured values. Net total carbon emission (net carbon) was defined and calculated as the sum of carbon emissions from the material production and construction stages (positive value, P1 to P8), and carbon emission reduction from the use stage (negative value, P9 to P15). Because carbon emissions from the production and construction stages were massive and inevitable, this value highlighted the

importance of the supercool cement's passive cooling effect on the overall carbon footprint.

Meteorological-responsive building design strategies are both adaptive and efficient for the future design of radiative cooling materials. To conduct a more targeted analysis of the carbon reduction priorities in different cities around the world, cities sharing highly similar meteorological, economical, and energy-related characteristics were clustered into five groups using a Gaussian mixture model (fig. S63) under an unsupervised modeling framework for machine learning (table S13); these five groups include developing, growing, innovative, middling, and emerging cities (fig. S64 and table S13). In each group, we conducted sensitivity analysis by calculating the feature importance of 15 energy-consuming modules via extreme gradient boosting. These feature importance values could reflect the contribution of each module, which was concluded in a Sankey diagram (Fig. 4C and fig. S65). It can be concluded that supercool cement is promising to substantially influence the total CO_2 emission value in a wide market including developing, middling, and emerging cities. Global color maps (Fig. 4D and fig. S66) exhibited remarkable performance for cooling buildings in cutting down on equivalent CO_2 emissions in the use stage. We conducted an in-depth investigation on seven cities (figs. S67 and S68) representing different categorized climate zones (43), namely, Mumbai (0A), Niamey (0B), Haikou (1A), Rockhampton (2A), Chongqing (3A; table S14), Miami (5A), and Sydney (6A). For every unit of cooling cement consumed, office buildings in Chongqing and Niamey (tables S14 and S15) achieved a carbon emission reduction of over 1183.34 and 2585.853 kg CO_2e in the use stage, respectively. Notably, on the basis of standardized buildings and controlled operational assumptions, a total carbon emission reduction of 2867.78 kg CO_2e could be achieved for office buildings in Niamey. The net carbon of seven cities demonstrated a notable radiative cooling effect to offset the inevitable carbon emissions in the energy-consuming production stage (fig. S69). In addition, we estimated the years required for global cooling buildings to compensate for carbon emissions in the early stage (Fig. 4E and fig. S70). When equipped with supercool cement products as envelopes, office buildings in nearly half of the cities in our calculations could achieve offset within their life span, achieving zero net carbon or even negative net carbon, demonstrating high potential for our supercool cement in energy-efficient applications.

DISCUSSION

In this study, we designed a novel type of cement raw particles, and further engineered a metasurface-enhanced supercool cement with self-assembled reflective crystals on light-interactive surfaces. This cement achieved high solar reflectance (96.2%) and blackbody-like emissivity (96.0%) in the long-wave infrared spectrum. Extensive performance tests confirmed its high mechanical robustness under compressive, flexural, abrasive, and adhesive forces, as well as its amphiphobicity, plasticity for complex shapes, and overall design versatility. Its cost-effectiveness and scalable fabrication processes give it unparalleled advantages over other materials, making it suitable for use in coatings, structural roofs, and walls, even in severe environments. In a cradle-to-grave machine learning-assisted LCA, supercool cement was estimated to reduce carbon emissions by up to 2867.78 kg/tonne over a 70-year life span compared to ordinary Portland cement. We have innovatively transformed cement

materials from heat absorbers to heat reflectors using a bottom-up approach. This breakthrough holds the potential to turn the heavy cement industry into a negative-carbon emission system, where supercool cement could play a key role in driving an energy-efficient, carbon-free future for the construction industry.

MATERIALS AND METHODS

Chemicals

Al_2O_3 , CaCO_3 , CaSO_4 , and SiO_2 particles were used for the fabrication of supercool cement clinkers after several treatment processes. Seawater, sodium chloride (NaCl), magnesium sulfate (MgSO_4), and sodium hydroxide (NaOH) were used in an environmental stability test.

Fabrication of cement clinkers as raw materials of supercool cement

To eliminate the influence of impurity ions, the supercool cement clinker was prepared using the chemical reagents CaCO_3 (C), CaSO_4 (CS), Al_2O_3 (A), and SiO_2 (S) derived from minerals as raw mixes. In lab-scale production, A, C, CS , and S particles were precisely weighted to match the reactant ratio and milled together in a ball mill machine for 1 hour at a speed of 400 rpm to achieve a fine and homogeneous mixture. After adding some water to the mix, the powders were pressed into cylinders (~3 cm in diameter and ~5 cm in height). These cylinders were then calcined in a muffle furnace (SGM-M31/13, SIGMA SHANGHAI), heated from room temperature to 1350°C at an increase speed of 5°C/min, and held at 1350°C for an hour. After calcination, the cylinders (now clinker) were rapidly cooled to room temperature. The clinkers were then milled with gypsum (450 rpm for 1 hour) to produce uniformly fine raw materials for supercool cement.

Fabrication of supercool cement with metasurfaces by two mold-reversed processes

The fabrication of supercool cement consists of two mold-reversed processes (see fig. S18), where the first is from the silicon mold to PDMS foil as the mold, whereas the second is from the PDMS mold to supercool cement with micropatterned surfaces. In the first mold-reversed process, the dual-component PDMS (SYLGARD 184 silicone elastomer base and curing agent, wt % ratio 10:1) was cast on the silicon wafer's surface, which was imprinted with a periodic aligned protuberant micropillar. After 4-hour curing at 70°C, the PDMS foil was peeled off from the silicon wafer and could act as the PDMS mold with concaved microcylinders on the surface. In the second mold-reversed process, a pressure difference-driven cavitation strategy was introduced for metasurfaces engineering. With one PDMS substrate as the mold, the mixed paste was cast onto PDMS's sophisticated side. The diameter of each micropillar and spacing between the PDMS mold is set to be tens of micrometers. Limited by the particle size of the hydration products, it did not fall completely into the micropillars. Thus, the remained gas still maintained normal atmosphere pressure between the micropillar and the surface of PDMS foil. Put them into the desiccator chamber and lower the pressure inside, the gas pressure difference will squeeze the paste up, forming microbubbles aligned at the boundary of PDMS and the cementitious side. Because of the elaborately chosen and designed of this fast-setting cool cement, it

had hardened in place before it fell. The specimens thus have aligned microcavities on the surface.

Fabrication of colored cooling cement composites

The phosphor dyes of yellow, green, and red [Ce:YAG , $\text{MgAl}_{11}\text{O}_{19}$: (Ce,Tb), and $\text{CaAlSiN}_3\text{:Eu}^{2+}$, respectively] were used as functional fillers with a mass ratio of 5 wt %. Because cement paste could accommodate many fillers, so the dyes could be directly mixed with the fresh cooling cement paste. The colored cooling cement composites share the same fabrication process as the white ones, only with the addition of dyes at the mixing process, and after the cavitation and hardening stage, colored cooling cement composites were then obtained with high simplicity.

Fabrication of the amphiphobic cooling cement composites

SiO_2 nanoparticles (~30 nm) and 1H,1H,2H,2H-perfluorodecyltrimethoxysilane (PFDTTS) were uniformly mixed with ethanol in a specific proportion to create a solution. The supercool cement surface was dipped into the solution and air-dried to form amphiphobic cooling cement composites.

Material composition characterization

An x-ray fluorescence (XRF; Panalytical Zetium) spectrometer was used for relatively chemical composition in the form of oxides. X-ray diffraction (XRD; Bruker D8 Discovery diffractometer with $\text{CuK}\alpha$ radiation at 40 kV and 30 mA) was conducted for phase identification of the unhydrated and hydrated specimens. The 10 wt % ZnO powder was used as an internal standard by mixing with paste for quantification analysis. For quasi-quantification analysis, XRD and XRF results were combined based on TOPAS (tables S7 and S8) Fourier transform infrared (Thermo Nicolet 5700) was performed to analyze the chemical bonds in the hydration products with wave number range of 400 to 4000 cm^{-1} and resolution of 0.1 cm^{-1} . One milligram of vacuum-dried paste powder was mixed with 100 mg of KBr to prepare pellets for this spectroscopic method. This type of vibrational spectroscopy can determine the vibration frequencies of constituent atoms or their coordination groups and is suitable for short-range order amorphous gel, which is otherwise not feasible by the XRD method. Thermogravimetric analysis (TGA; STA449F3 thermogravimetric analyzer, NETZSCH, heating rate of 10°C/min) was conducted to analyze the chemical composition.

Morphology characterization

Scanning electron microscopy (SEM; Hitachi Regulus 8100) was used to analyze the external morphology and nano/microscale structure of the cooling cement composite specimens. Elemental maps by energy-dispersive x-ray spectroscopy were obtained simultaneously. Crushed pieces were placed on holders by conductive tape and coated with Pt for 360 s (Quorum Q150T Plus). Mercury intrusion porosimetry (Micromeritics AutoPore V 9620) was used to measure the pore distribution of supercool cement, with measurable pore size ranging from 4 nm to 350 μm . A laser particle size analyzer (Malvern Mastersizer 2000) was used for the determination of cement clinker's particle size.

Optical characterization

Spectral reflectance of powders (cement, functional fillers, and hydration products) and hardened pastes were measured in the range

of 0.28 to 2.5 μm using an UV/visible (VIS)/near-infrared (NIR) spectrometer (Shimadzu 3600-plus) equipped with an integrating sphere with a test step of 5 nm. Solar spectrometer tests conform to standard test methods. Note that any reflectance greater than 1 means that the measured reflectance exceeds that of standard reference, thus we assumed the reflectance value of 0.99 for the exceeding part. Spectral emissivity of powders and hardened pastes were measured in the range of 2.5 to 20 μm using a Fourier transform infrared spectrometer (Nicolet 6700) equipped with a gold-coating integrating sphere (temperature assumed to be 25°C unless otherwise stated). The solar reflectance $R_{\text{solar}}(\lambda)$ of each tested sample is calculated as

$$R_{\text{solar}}(\lambda) = \frac{\int_{0.28}^{2.5} I_{\text{solar}}(\lambda) \times R(\lambda) d\lambda}{\int_{0.28}^{2.5} I_{\text{solar}}(\lambda) d\lambda} \quad (1)$$

where $I_{\text{solar}}(\lambda)$ is the air mass (AM) 1.5 solar irradiance (44). Note that, to measure the absolute reflectance with accuracy, we use a calibrated white plate [polytetrafluoroethylene (PTFE)] as a reference to calculate on the relative measured value as the equation

$$R(\lambda) = \frac{R_{\text{reference_absolute}}(\lambda)}{R_{\text{reference_relative}}(\lambda)} \times R_{\text{radiator_relative}}(\lambda) \quad (2)$$

where $R_{\text{reference_absolute}}(\lambda)$ and $R_{\text{reference_relative}}(\lambda)$ are the absolute and relative reflectance value (measured by the spectrometer) of reference white plate, whereas $R_{\text{radiator_relative}}(\lambda)$ is the relative reflectance value of the tested radiator.

Refractive index measurements were taken for supercool cement using HORIBA France SAS ellipsometers with the wavelength ranging from 0.28 to 16 μm . Images of samples and apparatus were taken using Sony ZVE-10 (visible) cameras. A macro lens (LAOWA, 25 mm, $f = 2.8$) was integrated when taking macro images of bubble formation. Whiteness values of the specimens were measured using an Intelligent Digital Whiteness Meter WSB-3A from Shenzhen ThreeNH Technology Co. Ltd. Infrared thermal images were taken using ZLV-FLIRE6390 in a hot region. A handheld optical microscope (Tipscope CAM V2) was used to testify the uniformity of microcavity alignment in scalable supercool cement's metasurface.

Calculation of bandgap

The bandgap of a material describes the energy needed to excite an electron from the balance band to the conduction band. The most accepted and widely used method to estimate the bandgap energy of semiconductors is proposed by Tauc (45) in 1966 as

$$(\alpha \cdot h\nu)^{1/\gamma} = B(h\nu - E_g) \quad (3)$$

where h is the Planck's constant, ν is the photon's frequency, E_g represents the bandgap energy, and B is another constant. According to the theory of Kubelka–Munk (K-M) function presented in 1931, the measured diffuse reflectance spectra can be used in bandgap calculation by being transformed to the corresponding absorption spectra as

$$F(R_{\infty}) = \frac{K}{S} = \frac{(1 - R_{\infty})^2}{2R_{\infty}} \quad (4)$$

where R_{∞} represents the measured diffuse reflectance of an infinitely thick specimen, whereas K and S are the absorption and scattering coefficient. Combine two equations (eqs. S1 and S2), we have

$$[F(R_{\infty}) \cdot h\nu]^{1/\gamma} = B(h\nu - E_g) \quad (5)$$

Plot $[F(R_{\infty}) \cdot h\nu]^{1/\gamma}$ against energy ($h\nu$), which is known as the Tauc plot, and the x -axis intersection point of the tangent of the Tauc plot represents the value of bandgap energy.

Scattering efficiency simulation

Numerical simulation based on Comsol Multiphysics (46) was performed to model the absorption behavior of a single rod-like ettringites with different cross-sectional sizes (ranging from 0.1 to 0.6 μm) and different incident light directions (vertical or horizontal). Two-dimensional Helmholtz wave equation was used as

$$\nabla \times (\nabla \times E) - k_0^2 \epsilon_r E = 0 \quad (6)$$

where ∇ is the Hamiltonian, $k_0 = \frac{2\pi}{\lambda}$ is the wave number, $\epsilon_r = n + ik$ is the complex refractive index containing real part n and imaginary part k . Assuming that the electromagnetic wave travels in the z direction, and the propagation factor equals $e^{(ikz^2 - i\omega t)}$, thus the electric field $E(x, y, z)$ can be expressed as

$$E(x, y, z) = E(x, y) e^{-ikz^2} \quad (7)$$

The computation consists of two steps by first calculating the background electric field $E^{bg}(r)$ in the air without the ettringite and, second, evaluating the scattering field $E^{scat}(r)$ with the rod-like ettringite. $E(x, y)$ represents the field components in rectangular coordinates (x, y, z) . The total field $E(r)$ can be represented as the sum of the background and scatters fields as

$$E(r) = E^{bg}(r) + E^{scat}(r) \quad (8)$$

Perfectly matched layers are used at the simulation boundaries to eliminate unrealistic reflections that add to the scattering field solution (47). The input electric field is set to be 1 V/m, with light power square of it. The scattering cross section was calculated using two Poynting vector components, which were then integrated and normalized to the input power.

FE simulation for abrasion tolerance

To testify the abrasive tolerance performance of supercool cement under real loads, an abrasion tolerance numerical simulation was performed using FE simulation. First, the simplified geometric models were designed with fixed microcavities' diameters (25 μm) and changing spacings (25, 50, and 75 μm) etched on the surface of the substrate. After refining the meshes, we simulated the finished models for numerical abrasive calculation. Note that the geometric modeling, mesh refinement, and FE modeling were all performed based on ANSYS LS-DYNA 17.2 (48). A horizontal load using a displacement-controlled method with a strain rate of 1 s^{-1} was applied in the FE model on supercool cement's metasurface. The friction coefficient was set to be 0.50. LS-Prepost was used for the postprocessing.

Field tests of temperature drop

The field test demonstrates the temperature drop of the supercool cement under direct sunlight compared to ambient conditions and

commercial Portland cement composites. The apparatus used for in-field radiative cooling performance included aluminum foil and expandable polystyrene (EPS) foam (to minimize heat conduction), polyethylene (PE) film (to reduce heat convection), and a data logger connected to thermocouples for temperature measurement. Accurate temperatures were captured using a weather station (Delta OHM HD52.3DP17R).

Mechanical tests

All the mechanical tests were conducted at room temperature and follow a standard compressive strength testing method (49, 50) on supercool cement, which were fabricated into cubes with dimensions of 50.8 mm by 50.8 mm by 50.8 mm using a universal testing machine (SUNS, UMT5105) with loading rate of 250 N/s. Note that the thrust surface is the side of the metasurface to testify its mechanical robustness as armor. A standard flexural strength testing method (32) was adopted with a specimen size of 40 mm-by-40 mm-by-160 mm prism. In the abrasion tolerance test, supercool cement cubes with the size of 50.8 mm by 50.8 mm by 50.8 mm were placed on the platform with the metasurface facing downward in direct contact with a 240 mesh sandpaper sheet. A weight of 200 g was placed on the sandpaper as an extra load for the abrasion. Pull to the sandpaper and the weight to a certain direction with the cube still, then the metasurface would be rubbed by the rough sandpaper as a horizontal load. Thirty cycles are exerted on the supercool cement's metasurface to verify armor's protection. A standard test method for the pull-off strength of coatings using portable adhesion testers (SHENGSHIWEIYE, SW-6000C) was conducted to evaluate the pull-off strength (commonly referred to as adhesion) of a coating on rigid substrates, including concrete, metal, and ceramic tile. This method involves attaching a disk-shaped loading fixture to the coating with epoxy resin adhesive at first, then firmly holding the instrument with one hand, and then turning the handwheel clockwise using as smooth and constant motion as possible. The maximum force required to pull the coating off the substrate is recorded and expressed as a measure of the coating's adhesion strength, typically reported in megapascals (MPa). The calculation formula of adhesive strength R in the unit of MPa is

$$R = \frac{F}{S} \times 10^3 \quad (9)$$

where F is the maximum force applied to detach the tile, measured in kilonewtons (kN). S is the area of the surface of the tile that was adhered to the substrate, measured in square millimeters (mm^2) or square meters (m^2).

Wetting characterization

The droplet dripping test was conducted by extruding a droplet onto superhydrophobic supercool cement's metasurface, which was leaned at an angle. The images were recorded using a high-speed camera (Revealer, 5F04). The static water contact angle measurement was performed using a video contact angle meter (Chengde Dingsheng JY-82C). A sessile drop (5 μl , oil and water) was dispensed onto the test surface, and the side-view images were captured with the integrated cameras in a static state. An antifouling test was conducted by pouring a mud suspension (a mixture of mud and water) onto the superhydrophobic supercool cement's surface.

Color calculation

The coordinates in CIE 1931 color space (x, y) were derived from tristimulus values X , Y , and Z , which were measured by a fluorescence spectrometer to determine the visually perceived dominant spectral wavelength of the given sample. $x = \frac{X}{X+Y+Z}$ and $y = \frac{Y}{X+Y+Z}$.

Corrosive liquid immersion test

Artificial seawater, sodium chloride (NaCl) particle, sodium hydroxide (NaOH) particle, and magnesium sulfate (MgSO_4) particle were used in a corrosive liquid immersion test. The artificial seawater was prepared in accordance with standard substitute ocean water for laboratory tests, with a pH value of 8. The NaCl solution was prepared with a concentration of 2.5 M and pH value of 7. The NaOH solution was prepared with a concentration of 0.1 M and pH value of 13. The MgSO_4 solution was prepared with a mass ratio of 3 wt % and pH value of 8. In the immersion tests, the supercool cement slabs (size of 30 mm by 30 mm by 3 mm) underwent five cycles of being immersed entirely into the corrosive solution for 24 hours and dried for another 24 hours.

UV radiation aging test

A UV radiation aging test was conducted under a common exposure condition, which has been used for roofing materials (34). With the lamp of UVA-313 and under a typical irradiance of $\sim 0.62 \text{ W/m}^2\text{-nm}$, five cycles are conducted consisting of 20-hour UV at a $80^\circ \pm 3^\circ\text{C}$ black panel temperature and 4-hour condensation at a $50 \pm 3^\circ\text{C}$ black panel temperature.

Freeze-thaw cycle

A freeze-thaw test was performed for 1025.5 hours to determine cooling cement composites' stability under various conditions through a series of rapid temperature changes. The test conditions consist of temperature drop and recovery between 3° and -16°C in a speed of 2°C/hour . All the specimens were held at 3° and -16°C for 1 hour during each cycle.

Supplementary Materials

The PDF file includes:

Supplementary Text
Figs. S1 to S72
Tables S1 to S18
Legends for movies S1 to S4
Legends for data S1 and S2
References

Other Supplementary Material for this manuscript includes the following:

Movies S1 to S4
Data S1 and S2

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