

Development of a Thermally Insulating Passive Daytime Radiative Cooler Using Hierarchical Silica Aerogels

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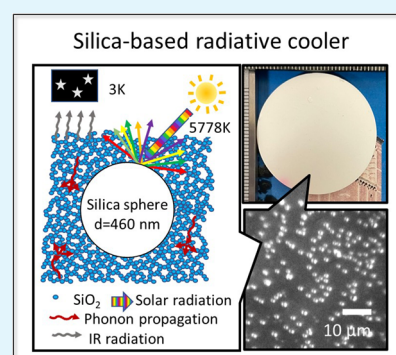
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ABSTRACT: Passive radiative cooling is an energy-free technique for dissipating heat to space (3 K) through the atmospheric transparency window (8–13 μm); it can be used for applications such as enhancing thermal efficiency in buildings or refrigeration. Silica is a promising radiative cooling material due to its wide bandgap, high mid-IR emissivity, and low cost. However, it has two major disadvantages. First, its moderate refractive index across the solar spectrum limits optical reflection and allows for transmission of solar radiation, and second, the Reststrahlen effect is observed in the atmospheric transparency window, which limits IR emittance of bulk silica. In this study, we developed a hierarchical organosilica-based material that mitigates both disadvantages by integrating solar-scattering silica spheres into an IR-emissive and thermally insulating silica aerogel. The large particles effectively backscatter solar radiation due to their high refractive index mismatch with the low effective refractive index aerogel matrix. Furthermore, the porous silica-based aerogel matrix emits in the mid-IR almost ideally by mitigating the Reststrahlen effect through its reduced refractive index. A 3.5 mm-thick ambiently dried aerogel with 15.6 particle vol % of silica showed large solar-weighted reflectance ($97.8 \pm 0.5\%$) and high emissivity ($99.2 \pm 0.3\%$) in the atmospheric transparency window while maintaining the thermally insulating properties of aerogels ($0.055 \pm 0.010 \text{ W/m}\cdot\text{K}$). In the proof-of-concept outdoor experiment, the surface of the aerogel exposed to direct sunlight was 10 °C lower than the ambient temperature and 26 °C lower than a bare Al plate under the same condition, demonstrating significant passive daytime radiative cooling promising for refrigerated transport. In addition, the aerogel showed resistance to UV light exposure, water, dirt, and steam, indicating exciting potential for practical applications.

KEYWORDS: silica, aerogels, organosilica, ambigel, daytime radiative cooling, porous materials, hierarchical structure



1. INTRODUCTION

The year 2024 was hotter than any year humans have experienced.¹ Heat waves significantly raise the risk of heat strokes and food-borne diseases.² Unfortunately, the immediate solution of air conditioning and refrigeration requires energy and inevitably releases heat to the surroundings, contributing to the urban heat island effect. This further exacerbates inequality by causing more damage in less developed areas where the energy to afford a comfortable living temperature is not readily accessible. Hence, energy-free thermal management technology, such as passive cooling, has garnered much interest.³

Passive daytime radiative cooling is a potentially carbon-neutral cooling technique using the cold space as a natural heat sink (Figure 1a).⁴ An object cools when its outgoing heat transfer rate exceeds the incoming one. For a sky-facing material on Earth, the net cooling power during the day, $P_{\text{net}}^{\text{I}}$ (in W/m^2), is defined as⁴

$$P_{\text{net}}^{\text{I}} = P_{\text{rad}}^{\text{I}} - P_{\text{sol}}^{\text{I}} - P_{\text{atm}}^{\text{I}} - P_{\text{non-rad}}^{\text{I}} \quad (1)$$

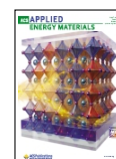
where $P_{\text{rad}}^{\text{I}}$ is the outgoing radiative power of the surface, and $P_{\text{sol}}^{\text{I}}$, $P_{\text{atm}}^{\text{I}}$, and $P_{\text{non-rad}}^{\text{I}}$ are the incoming power from absorbed solar radiation, absorbed atmospheric radiation, and non-radiative heat transfer processes, including conduction and convection, respectively. The Earth receives non-ideal blackbody radiation from the sun in the ultraviolet (UV)-visible to near-infrared (IR) range and emits long wavelength infrared (LWIR) radiation into cold space (Figure 1a). Greenhouse gases such as carbon dioxide, methane, and water trap some outgoing radiation by absorbing and re-radiating it back to the surface. For radiation to directly reach outer space, its spectral energy should lie in the atmospheric transparency window (ATW) corresponding to the wavelength range from 8 to

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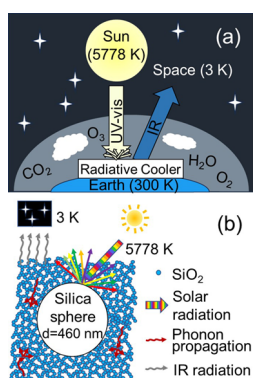


Figure 1. Schematics of (a) the passive daytime radiative cooling principle, (b) the specific daytime radiative cooler designed, synthesized, and investigated in this study. In our design, silica spheres of different length scales create a hierarchical structure comprised of silica spheres with a diameter of about 460 nm embedded in a silica aerogel with a uniform pore size of about 10 nm. The large spheres effectively scatter incoming solar radiation, while the mesoporous aerogel matrix provides high mid-IR emission and thermal insulation.

13 μm , where the atmosphere has weak IR-active modes. Therefore, an ideal sky-facing daytime radiative cooler should emit strongly in the ATW to maximize the emitted radiation, P''_{rad} , combined with minimal absorption at solar wavelengths to minimize absorption of incoming radiation from the sun, P''_{sol} . In addition to possessing low solar absorbance, the cooler should effectively reflect or backscatter incoming solar radiation. To achieve high solar reflectance and IR emissivity, a wide range of materials have been designed in the form of paint,⁵ film,⁶ slab,⁷ or textile⁸ for applications in buildings, clothing, solar cell coating, or moisture harvesting.³

Another parameter to be optimized is the non-radiative heat gain, $P''_{\text{non-rad}}$, which arises from conductive and convective heat exchange between the radiative cooler and its surroundings. Although the simplest form of a cooler that is designed to cool a hot object would benefit from a high thermal conductivity, there are many applications that involve keeping an already cold object from heating up where a thermally insulating cooler would provide superior performance. As a result, development of both thermally insulating and thermally conductive radiative coolers is needed. For instance, if the substrate temperature is hotter than the ambient, such as in cooling textiles, heat would be dissipated more effectively with a thermally conductive radiative cooler.^{6,9} By contrast, if the substrate temperature is significantly colder than the ambient, such as in an air-conditioned buildings in the summer or for application in extreme environment such as space or desert, parasitic heat gain can be reduced with a thermally insulating radiative cooler.¹⁰ To increase the energy efficiency in buildings, various porous materials with low thermal conductivity have been studied.^{11,12–17} In porous materials, the low thermal conductivity of air in the pores and high degree of phonon scattering at the air-solid interface maintains an intrinsic thermal barrier.¹² Various porous polymers such as polyethylene,^{7,13} polyurethane,¹⁴ polyvinylidene fluoride (PVDF),¹⁵ polyvinyl alcohol (PVA),¹⁵ and polydimethylsiloxane (PDMS)¹⁶ have shown to be effective thermally insulating radiative coolers at different working conditions. However, since organic materials are prone to UV-induced

degradation over time, porous inorganic materials would be more favorable for daytime applications.¹⁷

Unfortunately, while it is ideal for a radiative cooler to have low absorbance and high refractive index in the solar spectrum, these two properties are inversely related in semiconductors.¹⁸ This is because electromagnetic waves that are not absorbed by a material create a phase lag, resulting in a high refractive index mismatch between air and the material.¹⁹ Silica is a popular material with a suitably wide bandgap of 8.4 eV, which corresponds to short-wave UV where solar radiation is insignificant (~ 147 nm), and broad absorption in the ATW at around 9.7 μm arising from the strong Si-O-Si vibrational mode. It is also sustainable, with a high natural abundance and low cost.^{20,21}

Silica's moderate refractive index of 1.5 at optical wavelengths enables the design of materials that are either weakly or strongly scattering, depending on their structure. For example, fused quartz can be used to form optical materials in various forms, including bulk,^{22–24} thin films,⁴ nanospheres with a diameter below 100 nm,^{25,26} microsphere,^{27–29} nanoshell,³⁰ coatings on ceramic,³¹ and porous networks.^{32,33} However, complete solar reflectance R_{sol} and high emittance in the ATW, ϵ_{ATW} , using silica alone are limited by its moderate refractive index in the UV-visible region resulting in modest solar reflectance R_{sol} and by the Reststrahlen band in the IR region which reduces the emissivity ϵ_{ATW} .³⁴ The Reststrahlen band arises from the strong phonon-polariton resonance at the surface of bulk silica due to Si-O-Si stretching, which reduces the permittivity in the ATW. To improve the solar reflectance, researchers have integrated silica with materials with higher refractive index, such as Ag coating,^{22,28,35} layers of metals,⁴ and ceramics,^{31,33,36} or more light scattering interfaces such as air-solid interface created by porous polymers.²⁹ To enhance the emissivity in ATW, surface patterning was found to be effective without affecting optical properties, by lowering the refractive index at the air-silica interface.^{23,24} Making a composite material with polymers with vibrational modes in the ATW such as polyvinyl alcohol (PVA),²⁵ polydimethylsiloxane (PDMS),^{22,30} polymethyl pentene (TPX),³⁵ polyvinylidene fluoride (PVDF),²⁹ and metal oxide³¹ also enhanced the IR emissivity.

In addition to experimental studies, the theoretical radiative cooling power of silica-based materials has been calculated to design promising metamaterials. For example, Yalçın et al.²¹ numerically predicted the ideal configuration of silica spheres and fiber in an air medium at 4 vol %. Results showed that silica spheres with a diameter on the order of optical wavelengths, and silica fiber with a smaller diameter of 150–550 nm could achieve both high solar intensity-weighted scattering coefficient and low blackbody intensity-weighted scattering coefficient, yielding a theoretical net radiative cooling power of 178 W/m^2 . When a polymeric binder was used as a matrix instead of air to simulate a more realistic design, both the solar reflectance and IR-emittance were greatly reduced due to the small refractive index mismatch between the polymeric matrix and the silica spheres and fibers, and the surface IR reflection at a low incident angle and total internal reflection at a large incident angle, respectively. In another study, Feng et al.³⁵ modeled silica spheres embedded in the optically transparent polymer TPX with a 200 nm-thick silver coating. The authors found that a sphere diameter of 600 nm was optimum when the net cooling power was positive. They also showed that increasing the volume

fraction of the particles was as effective as increasing the thickness of the sample at improving the cooling power. In addition, new 2D and 3D surface patterns on bulk silica have also been proposed to further engineer the emissivity.³⁷

Porous inorganic materials, such as silica aerogels, have been widely studied for radiative cooling and other thermal control applications owing to their low thermal conductivity. However, achieving both high solar reflectance and high emissivity within the atmospheric transparency window remains challenging, due to the inherent trade-off associated with their microstructure.³⁸ For instance, aerogels with smaller nanoscale porosity exhibit high emissivity and low thermal conductivity, but are not suitable for daytime cooling applications due to the high visible light transmittance.³⁹ Macroporous aerogels with larger building blocks exhibit higher visible light reflectance due to Mie scattering, but also exhibit higher IR reflectance and higher thermal conductivity due to gas conduction in the pores.⁴⁰ Notable studies have developed hierarchically structured ceramic-based materials optimized for high solar reflectance R_{sol} and high emissivity in the atmospheric transparency window ϵ_{ATW} .^{41–43} However, the emissivities of ceramic radiative coolers are often limited by the Reststrahlen bands inherent to wide-bandgap semiconductors.⁴⁴ In some approaches, polymers are incorporated to enhance the emissivity,⁴⁵ which may compromise long-term outdoor durability.⁵⁴

Herein, we report a hierarchically structured silica-based aerogel with limited organic components, exhibiting near-complete emissivity within the atmospheric transparency window, while maintaining a high solar reflectance a low thermal conductivity. Specifically, 460 nm silica particles were embedded in a homogeneous mesoporous organosilica aerogel containing small (<20 nm) pores with a narrow pore size distribution (Figure 1b) to form a hierarchical structure featuring three distinct length scales. At the atomic scale, the material utilizes the strong emissivity arising from the Si-O-Si vibrational mode of silica, which is optimally positioned for IR emission within the atmospheric transparency window. At the small nanoscale (2–20 nm), the mesoporous network formed from few-nanometer primary particles reduced the refractive index of the silica monolith and provided thermal insulation. Finally, at the large nanoscale (400–500 nm), large silica spheres effectively scattered UV and visible light to mitigate the heating effects of incoming solar radiation. The hierarchical structure, composed of small pores optimized for IR emissivity and thermal conductivity, and large Mie-scattering particles for optimized visible light scattering, enabled the high solar reflectance of the aerogel while preserving high emissivity and low thermal conductivity, without the use of polymers.⁵⁵

Ambiently dried organo-silica aerogel matrices present four advantageous properties for radiative cooling: (1) their ultra-low refractive index results in a significant refractive index mismatch between the aerogel matrix and embedded solid silica particles, (2) they provide mitigation of the Reststrahlen effect at 9–10 μm thanks to their high porosity (>85%), and (3) they show large directional IR emissivity at wide incident angles characteristics of dielectric materials. In addition, the high porosity of aerogel results in (4) a low thermal conductivity that can effectively maintain a thermal gradient. The synergy of high solar scattering from the silica spheres, and low thermal conductivity and high mid-IR emissivity from organo-silica aerogel create an optimal daytime radiative cooler analogous to the model of silica spheres embedded in air.²¹ Moreover, the very limited

organic fraction makes the present radiative cooling surface more resistant to UV degradation and fire.²⁰ As a result, the present work helps establish a framework for employ hierarchical structures to create synergistic combinations of properties in radiative cooling materials.

2. MATERIALS AND METHODS

2.1. Materials

Methyltrimethoxysilane (MTMS, 97%), chlorotrimethylsilane (TMCS, 98%), and tetraethyl orthosilicate (TEOS, 98%) were purchased from Thermo Scientific. Formamide ($\geq 99.5\%$) was purchased from Fisher BioReagents. Ammonium Hydroxide (28–30%, ACS Grade) was purchased from VWR Chemicals. Hydrochloric acid (36.5–38% in water, Certified ACS plus) and n-heptane (HPLC grade) were purchased from Fisher Chemical. Ethanol (200 proof) was purchased from Decon Labs. All materials were used as received without further purification.

2.2. Aerogel Preparation

Stöber synthesis was used to prepare non-porous silica nanospheres.⁵¹ Aerogel preparation followed the synthesis from the literature.⁴⁶ To prepare aerogels embedded with silica particles, acidic solution with hydrolyzed MTMS and TEOS was mixed in with a basic solution containing silica particles. After aging the gel for 2 days, the surface of the gel was silylated using TMCS, and the solvent was exchanged to heptanes before it ambiently dried.

2.3. Material Characterization

2.3.1. Optical Microscopy. Optical microscope images were taken using the AxioCam ICc 5 attached to an Axiotech 100 reflected-light microscope in $\times 1000$ magnification.

2.3.2. Scanning Electron Microscopy. Scanning Electron Microscopy (SEM) images were collected using a model JEOL JSM-6700F field emission electron microscope with 5 kV accelerating voltage and a secondary electron detector. Powdered white aerogel was placed on a carbon tape to image the white gel, and the concentrated silica particle solution was spin-coated on a plasma-cleaned piece of p-doped silicon to image the silica particles. The average size and distribution of the silica particles were measured using Image J software.

2.3.3. Dynamic Light Scattering. Dynamic light scattering measurements were conducted using Malvern Zetasizer Nano. Samples were prepared by diluting 20 μL of concentrated silica particles with 3 mL of DI water.

2.3.4. Nitrogen Porosimetry. The porosity and pore size distribution were obtained from N_2 adsorption-desorption isotherms at 77 K, using TriStar II 3020 surface area and porosity analyzer (Micromeritics Instrument Corp., Norcross, GA, USA). Both the clear and white aerogels were dried at 110 $^\circ\text{C}$ under vacuum overnight prior to these measurements.

2.3.5. Thermogravimetric Analysis (TGA). The thermogravimetric analysis was conducted using TGA8000 Thermogravimetric Analyzer (Perkin Elmer Inc., USA) under 45 mL/min of air.

2.3.6. Ultraviolet-Visible-Infrared Spectroscopy. The normal-hemispherical reflectance and transmittance of the sample within the wavelength range of 0.3–1.1 μm were measured using a double-beam ultraviolet-visible (UV-Vis) spectrophotometer (Evolution 220, Thermo Scientific Fisher, USA) equipped with an integrating sphere (Evolution ISA-200 Integrating Sphere Accessory, Thermo Scientific Fisher, USA). Spectral reflectance and transmittance were obtained by averaging data collected from three different spots on the sample, and the uncertainty in solar reflectance was estimated from the variation between measurements to be $\pm 0.5\%$. In the IR region, a nitrogen-purged Fourier transform infrared (FTIR) spectrometer (Nicolet iS50, Thermo Scientific Fischer, USA) equipped with an integrating sphere (Upward IntegratIR PIKE Technologies, USA) was

used for measurements of wavelengths between 1.1 and 20 μm . IR measurements at wavelengths between 1.1 and 2 μm were conducted using an InGaAs detector with a CaF_2 beam-splitter, while for wavelengths ranging from 2 to 20 μm , an MCT detector with a KBr beam-splitter was used. Note that the angular-dependent reflectance was not measured in this work. The aerogel was assumed to be optically diffuse and opaque due to its highly scattering structure and dielectric nature. Under this assumption, the measured normal-hemispherical reflectance was used to represent the directionally averaged optical response. Here, a total of 800 scans per wavelength were performed to reduce noise and ensure a low instrument uncertainty. The uncertainty in emissivity was determined from the standard deviation of emissivity calculated across five different aerogel samples to be 0.3%. A diffuse gold standard was used as the baseline in the IR spectrum.

2.3.7. Thermal Conductivity Measurements. The effective thermal conductivity k (in $\text{W/m}\cdot\text{K}$) of aerogels with thickness L was measured at room temperature using a guarded hot plate apparatus in accordance with the C177-13 ASTM standard, operating in a single-sided mode.⁴⁷ The thermal conductivity of each sample was determined from the average of three measurements, and the uncertainty was determined from error propagation. Details of the characterization methods can be found in the [Supporting Information](#).

2.3.8. Ultra-/Small Angle X-ray Scattering (USAXS/SAXS). USAXS and SAXS data were collected on the 9-ID-C beamline at the Argonne National Laboratory Advanced Photon Source.⁴⁸ USAXS data were reduced using the data reduction software provided by the beamline and SAXS data were reduced using the Nika⁴⁹ package of the Igor Pro software (WaveMetrics Inc, Portland, OR). Details of the measurement and the data processing method can be found in the [Supporting Information](#).

2.4. Outdoor Cooling Experiments

The outdoor cooling experiments were conducted four times in two different places. On February 15th, 2023 and June 28th, 2023, the experiment was conducted on the roof of the parking structure 2 at UCLA ($34^\circ 04' 07.3''\text{N}$, $118^\circ 26' 23.4''\text{W}$). On August 2nd, 2023, and August 19th, 2025 to on August 25th, 2025 the experiment was conducted on the roof of the California NanoSystems Institute (CNSI) at UCLA ($34^\circ 04' 05.0''\text{N}$, $118^\circ 26' 33.5''\text{W}$). The photo and schematic of the setup are presented in [Figures S1–S5](#). A total of 6 samples were compared: Al foil, 3 M tape, white paint, white aerogel, clear aerogel, and Styrofoam. Each sample was attached to aluminum substrates, and the temperature of the substrate was measured with a T-type thermocouple (OMEGA, USA). Note that this choice of substrate provides a high thermal conductivity which helps maintain temperature uniformity and enables reliable temperature measurements using an embedded thermocouple. Moreover, Al is often used to build spaces that need cooling, and so it is representative of some situations where radiative cooling may be beneficial. The ambient temperature was measured with a thermocouple in the shade, about 70 cm above the ground. The thermocouple was placed under a plastic table, with cardboard shielding on the sides and an open bottom to prevent direct heating and facilitate convection. The substrate temperature was recorded using two data loggers (Omega, USA/ Huato Electric, China). The surface temperature of the samples was determined using an IR camera (E96, FLIR systems, USA) by using their respective emissivity as input. The uncertainties in the temperatures of the substrate and ambient air measured with a thermocouple and the surface measured with an IR camera were $\pm 1^\circ\text{C}$ and $\pm 2^\circ\text{C}$, respectively. Details of the setup can be found in the [Supporting Information](#).

2.5. Modeling

Radiation transfer through the randomly distributed silica particles embedded in aerogel was simulated to design a radiative cooling material with maximum solar reflectance and maximum emittance in the

ATW, as previously discussed. Then, steady-state energy balance was formulated for different configurations.

2.5.1. Simulation of Solar Reflectance and IR Emissivity of Aerogels. A silica-based aerogel slab of thickness $L = 0.1\text{--}4$ mm and porosity ϕ of 85%, containing randomly distributed non-porous monodisperse silica particles of volume fraction f_v ranging between 1 and 20% and diameter d_s spanning 200–1000 nm was considered. Light transfer simulations of the aerogel monoliths were performed by the commonly used DMRT method,⁵⁰ which accounts for multiple scattering and dependent scattering among silica nanoparticles. Here, the normal-hemispherical transmittance $T_{\text{nh},\lambda}$, reflectance $R_{\text{nh},\lambda}$, and absorbance $A_{\text{n},\lambda}$ of the aerogels were numerically predicted in the 0.3–20 μm spectral range. Then, the corresponding solar-weighted reflectance R_{sol} and atmospheric emittance ϵ_{ATW} were computed according to [eqs S9 and S10](#), respectively. Details of the simulation could be found in [Section S1.4](#) of the Supporting Information.

3. RESULTS AND DISCUSSION

3.1. Theoretical Materials Design

In our design, the diameter d_s of the silica particles determined the magnitude of scattering of each particle, while the particle volume fraction f_v and the thickness L of the aerogel slab together govern interparticle distance, number of scattering centers, and multiple scattering. [Figure 2a,b](#) show the solar reflectance R_{sol} and emissivity in the ATW ϵ_{ATW} for varying thickness L and particle diameter d_s simulated assuming particle volume fraction f_v of 10 vol %. The results indicate that the solar reflectance increases with increasing particle diameter and slab thickness, i.e., with the total number of particles. By contrast, the IR emissivity ϵ_{ATW} exhibits a much weaker dependence on particle size or slab thickness because the overall emissivity of the aerogel was governed by the highly emissive nature of the aerogel matrix rather than the embedded silica particles.

[Figure 2c](#) shows the simulated spectral solar normal-hemispherical reflectance $R_{\text{nh},\lambda}$ of a 3.5 mm-thick aerogel containing varying volume fractions f_v of silica particles with a diameter d_s of 400 nm. The results indicate that while the reflectance $R_{\text{nh},\lambda}$ in the solar spectrum increased with increasing particle volume fraction, the reflectance in the UV-vis was high and mostly unchanged for f_v exceeding 10 vol %, with only a slight increase in the near IR with higher particle concentration. These numerical results suggest that the ideal sample to synthesize would be an aerogel slab with a thickness L greater than 3 mm, consisting of randomly distributed silica particles of diameter d_s between 400 and 500 nm, in a volume fraction f_v larger than 10%.

3.2. Synthesis and Characterization of Aerogels

The preparation of aerogel slabs embedded with silica nanoparticles consisted of two main steps: first, preparing silica particles using the Stöber synthesis method,⁵¹ and second, synthesizing the aerogel slab using established acid-base-catalyzed sol-gel chemistry.⁴⁶ After sol-gel transition, the gel was surface-treated with silylation agent and the solvent was exchanged with a low surface tension solvent, followed by drying at ambient pressure and temperature. As a control, a gel without particles was also made using a 2M ammonium hydroxide solution instead of the particle solution. The simulation results presented in [Section 3.1](#) and [Figure 2](#) indicate that an optimal combination of high solar reflectance and high IR emissivity could be achieved using aerogels containing between 10 and 20 vol % colloidal silica with a sample thickness greater than 3 mm, and

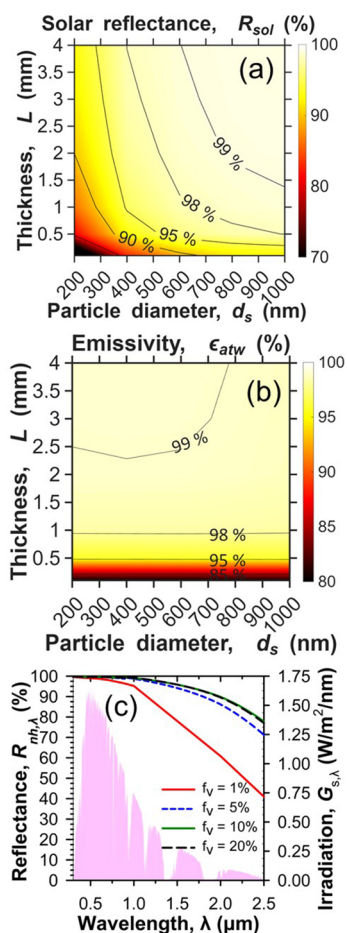


Figure 2. Dependence of the simulated (a) solar reflectance and (b) IR-emissivity on the diameter of silica spheres and the thickness of the aerogel. A particle volume fraction f_v of 10 vol % was used for both calculations. (c) Simulated spectral solar reflectance of four 3.5 mm thick aerogel samples containing silica particle diameter of 400 nm at different concentrations.

colloid diameters of 450–770 nm. In our synthetic design, the dispersion of individual silica particles was crucial in maximizing the solar reflectance of the aerogel slab, as aggregated colloids result in dependent scattering effects, which reduce the overall solar reflectance. Thus, the silica colloid content was chosen to have the highest value that could be fully dispersed in the porous silica monolith. While higher volume fractions could be incorporated, colloid aggregation due to the limits of electric double layer stabilization meant that further silica incorporation did not significantly improve solar reflectance. Considering both simulated results and what is experimentally achievable, the final aerogel had a diameter of 4.2 cm, thickness of 3.5 mm, and a particle volume fraction of 15.6%.

Scanning electron microscopy (SEM) images of the as-prepared silica colloids revealed spherical particles with a diameter centered around 460 nm, as illustrated in Figure 3a,b. No sign of aggregation appeared on the dynamic light scattering (DLS) data of the diluted particle solution, showing a hydrodynamic diameter of 510 nm with a polydispersity index of 0.13 (Figure 3c). The particle size determined by DLS was slightly larger than the size determined from SEM, probably due to a combination of two factors. First, DLS measures the

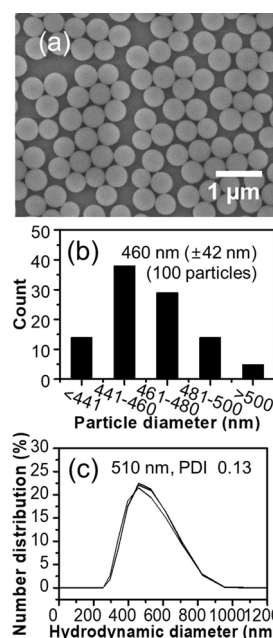


Figure 3. Characterization of silica particles used in this work. (a) SEM image of the colloidal silica spheres. (b) Particle size distribution of the colloidal silica particles from measurement of SEM images. The average diameter of the particles was 460 nm, with a standard deviation of 42 nm. 100 particles were measured for the analysis. (c) The colloidal silica size distribution from dynamic light scattering (DLS) of dilute aqueous silica solutions. The average hydrodynamic size was 510 nm, with the polydispersity index (PDI) of 0.13.

hydrodynamic diameter (including the counterion cloud), while SEM measures the silica core size. Second, each colloid is measured individually from the SEM image, but if two particles were stuck together, they would appear larger in DLS. The agreement between the two values indicates that most colloids were well separated, but a few dimers likely existed.

The dried gel without the particle was transparent (clear aerogel), while that with particles was opaque (white aerogel) (Figures 4a and S6a). The particle-embedded gel in Figure 4 had diameter of 4.2 cm and thickness of 3.5 mm. The particle volume fraction, f_v , calculated from the volume of the gel and the volume fraction of the colloidal solution, was 15.6%. The porosity, ϕ , calculated from the N_2 adsorption-desorption isotherm at 77 K of aerogel without particles, was 84%. This value decreased to 74% with particles, as expected given the volume fraction f_v of silica spheres in the aerogel matrix. The high porosity and the narrow pore size distribution that peaks at 10 nm were maintained upon addition of the larger silica colloids, confirming that adding the silica particles did not change the porous aerogel structure (Figure S7). Ultra-/small-angle X-ray scattering (USAXS/SAXS) of the aerogel samples with and without silica particles further confirmed that the mesoporous architecture was well preserved upon addition of the large silica colloids. The data show nearly identical patterns for both samples at $q > 0.015 \text{ \AA}^{-1}$, a range that corresponds to structural features below 42 nm (Figure S8a). At lower q , scattering from the well-separated large silica colloids can be clearly observed only in the composite white aerogel. In the mass fractal regime, the radius of gyration R_g , representing the cluster size, and the mass fractal dimension D_{mv} , representing the tortuosity of the aerogel network, were 8.7 nm and 2.2, respectively, for both

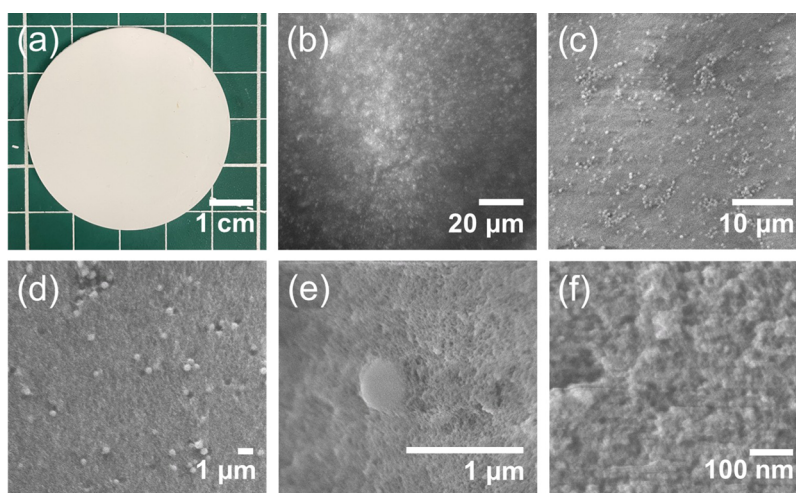


Figure 4. The hierarchical structure of the white aerogel is revealed from images taken at varying magnifications. In the white aerogel, few-nanometer-sized silica particles form a mesoporous network that tightly holds few-hundred nanometers silica spheres. (a) Photo of the aerogel. The aerogel appeared white due to the visible light scattering by large silica particles (b) Dark field optical microscope image. Light scattering from individual silica spheres appears as dust-like specs. (c–f) Scanning electron microscope images with increasing magnification. (c) The distribution of silica spheres is in good agreement with the white specs shown in (b), confirming that the visible light is scattered by the large silica spheres rather than the silica aerogel matrix. (d) Well-dispersed silica colloids in the aerogel matrix are observed. (e) Individual large silica spheres are well embedded in the porous matrix. (f) A zoomed-in image of the porous aerogel matrix shows very small silica particles (<10 nm) forming a well-connected network with uniform pores of 10–20 nm.

samples, in good agreement with previously reported values (Figure S8b).^{46,52,53}

The thermal conductivity of the aerogel slabs also remained low, with or without silica particles, and was measured at room temperature as 0.055 ± 0.010 and 0.051 ± 0.010 W/m·K, respectively with one standard deviation confidence intervals. (Table S1) These values of thermal conductivity are consistent with previous reports in the literature.⁴⁶ The similarity in the measured thermal conductivity suggests that the large silica particles were well enough separated by the porous aerogel matrix that they do not increase the phonon mean free path.

The hierarchical nanostructure of the aerogel was revealed from images taken at different magnifications. (Figure 4) The large Mie-scattering silica spheres embedded in the mesoporous aerogel matrix make the particle-embedded aerogel appeared uniformly white (Figure 4a). In a dark field optical microscope image, these larger nanoscale scattering centers were resolved, looking like specs of white dust on a dark background (Figure 4b). The dispersion of particles in the optical microscope image suggested a homogeneous distribution of the large Mie-scattering particles within the optically transparent aerogel matrix. SEM images of the aerogel further confirmed that the particles were mostly separated, with only a few pockets showing higher particle concentrations (Figure 4c,d). A magnified image of a single embedded particle shows that it was tightly held by the porous aerogel matrix (Figure 4e). When the image was further zoomed in on the aerogel matrix, the smaller nanoscale pores were revealed, where silica particles with diameters below 10 nm formed a homogeneous porous network with pores below 20 nm (Figure 4f).

The reflectance and transmittance spectra of the white and clear aerogels were collected across UV-visible and IR wavelengths (Figures 5 and S6b). In the IR region, the spectral hemispherical emissivity was assumed to be equal to the spectral hemispherical absorptivity according to Kirchhoff's radiation

law for a diffuse emitter; the spectral absorptivity, in turn, was calculated by subtracting the measured spectral hemispherical reflectance and transmittance from unity. The experimentally measured aerogel transmittance of the colloid-loaded white aerogel was below 1.5% across the full solar spectrum (0.3–3.0 μm), and near-total reflectance was observed from 0.35 to 1.3 μm , where the peak intensity of the solar radiation lies. (Figure 5a) In the IR region, the white aerogel showed broadband emissivity, with nearly complete emissivity across the atmospheric transparency window. (Figure 5b) The solar-weighted reflectance R_{sol} and the emissivity in the ATW ε_{ATW} was 97.8 ± 0.5 and $99.2 \pm 0.3\%$, respectively. The net cooling power calculated from the solar absorbance and emittance of white aerogel was 160 W/m^2 , making it one of the highest among reported daytime radiative cooling materials to date (Table 1).

To further validate the theoretical model used in Section 3.1, the spectral reflectance and transmittance of our specific material were simulated using the measured particle size 460 nm, particle volume fraction of 15.6%, and aerogel slab thickness of 3.5 mm (Figure S9). The simulated solar reflectance R_{sol} and emissivity ε_{ATW} were $\sim 99\%$ and $>99\%$, respectively, which were quite close to the experimental values of 97.8 and 99.2%, respectively. The experimental and simulated spectra showed good agreement in most spectral windows, although the aerogel was slightly more absorptive between 1.3 and 5 μm . In the DMRT model, the aerogel matrix was assumed to be a homogeneous mixture of silica and air, without accounting for the presence of organic surface groups and water. The surface methyl and hydroxyl group on the aerogel and adsorbed water likely contributed to the slight discrepancy between simulation and experiment. Specifically, vibrational modes associated with C-H stretching of the methyl groups from the MTES precursor and TMCS treatment ($2800\text{--}3000 \text{ cm}^{-1}$, 3.3–3.6 μm), Si-OH (4372 cm^{-1} , $\sim 2.3 \mu\text{m}$), broad absorption from O-H stretching

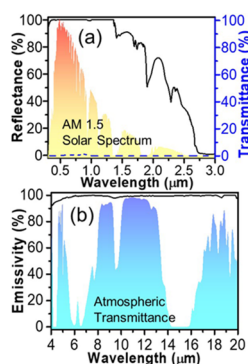


Figure 5. Spectroscopic characterization of the white, colloid containing porous silica aerogel. a) Spectral reflectance in the solar spectrum, and (b) spectral emissivity in the IR region. The colored regions in the background of (a) and (b) corresponds to the AM 1.5 solar spectrum and the transmission spectrum of the Earth's atmosphere, respectively. The reflectance is high across visible wavelengths, and emissivity in the ATW is uniformly high. The white aerogel had a diameter of 4.2 cm, a thickness of 3.5 mm, a porosity of 74%, and a pore size of ~ 10 nm.

at (~ 3500 cm^{-1} , 2.9 μm), H-O-H bending (5254 cm^{-1} , 1.9 μm), and combination band of water (~ 2000 cm^{-1} , 5 μm) and overtones of water (6896 cm^{-1} , 1.45 μm) all contributed to the broad absorption in the near-IR.^{62,63} Such modes are an intrinsic part of a high surface area, porous material, and they fortunately fall in a spectral window with weak solar radiation. As a result, the discrepancies have minimal effect on the solar reflectance ($\sim 1\%$), allowing the model to remain a valid tool to design future radiative cooling materials.

3.3. Outdoor Performance Evaluation

Studies of the radiative cooling property of the white, particle-embedded aerogel began on a sunny winter day (February 15th, 2023). The white aerogel was adhered onto an aluminum plate substrate and secured on a styrofoam-embedded plastic cup, covered with a polyethylene film to minimize convection effects. The substrate temperature T_{sub} underneath the thick aerogel slab was recorded using an embedded thermocouple, while the surface temperature T_{surf} was measured hourly with an IR camera (Figures 6a,b and S1). The ambient temperature was measured with a thermocouple in the shade. From 9 a.m. to 2:30 p.m., The substrate temperature was about 1 $^{\circ}\text{C}$ higher than the ambient temperature, on average. However, the surface temperature of the aerogel remained 9 – 11 $^{\circ}\text{C}$ lower than the ambient temperature. This suggests that the white aerogel backscattered most of the incoming solar radiation, and the high thermal resistance of the aerogel slab inhibited additional heat loss from the substrate to the radiatively cooled surface.

To verify the experimental surface temperature, the theoretical surface temperature was calculated from the experimentally measured substrate temperature. (Figure 6b) The schematics and parameters used to calculate the surface temperature are shown in Figure S10. Four assumptions used in Section S1.4.2 and two additional assumptions were used:⁵ Heat transfer by convection was suppressed.⁶ The aluminum substrate was sufficiently insulated and there was no heat loss from its bottom. Then, the energy balance applied at the aerogel surface can be written as

$$q''_{\text{r,s-a}} - q''_{\text{cond}} - q''_{\text{atm}} = 0 \quad (2)$$

where $q''_{\text{r,s-a}}$ denotes the heat flux (in W/m^2) due to solar absorption retrieved from eq S13, with the aerogel spectral absorptivity being equal to its experimentally measured emissivity in accordance with Kirchhoff's law for a diffuse and opaque surface ($\alpha_{\lambda, \theta} = \epsilon_{\lambda, \theta} = \epsilon_{\lambda}$).

The heat flux due to conduction $q''_{\text{cond,a}}$ (in W/m^2) was given by

$$q''_{\text{cond,a}} = \frac{k_a}{L} (T_{\text{surf}} - T_{\text{sub}}) \quad (3)$$

where k_a and L are the white aerogel thermal conductivity and thickness taken as 0.055 $\text{W}/\text{m}\cdot\text{K}$ and 3.5 mm, respectively. Additionally, the heat flux due to radiation exchange with the atmosphere q''_{atm} was computed from eq S14, where eqs S17 and S18 were used to calculate the corresponding emitted radiative power P''_{rad} at a given aerogel surface temperature T_{surf} and the absorbed atmospheric radiative power P''_{atm} at a given sky temperature, T_{sky} . The atmosphere had a temperature $T_{\text{sky}} = 0.552T_{\infty}^{1.5}$,⁶⁴ where T_{∞} is the experimentally measured ambient air temperature using eq S19.

Finally, to solve for the surface temperature T_{surf} , the experimentally measured air temperature T_{∞} and the substrate temperature T_{sub} were used as prescribed boundary conditions at a given time of day. Then, Newton's method was employed to solve the energy balance iteratively. Predictions of the aerogel slab surface temperature fell within the experimental uncertainty of the measured ambient and substrate temperatures. (Figure 6b) The substantially cold surface of the white aerogel confirms its radiative cooling properties.

However, cooling of the substrate below ambient temperature was not evident due to thermal conduction from the ground and the experimental setup, which elevated the air temperature near the sample above ambient. Currently, there is no standard method for conducting outdoor cooling measurements, which makes it challenging for researchers to custom-build setups where the temperature of the enclosed environment matches the ambient temperature. Indeed, the solar absorptivity and thermal conductivity of the experimental setup greatly influence the air temperature immediately surrounding the samples, often resulting in a higher temperature than the ambient.⁶⁵ The heating of our setup and the cooling of the sample's substrate below the enclosed temperature will be discussed in the following section. However, we acknowledge that the details of cooling below the inner temperature of the setup are not equivalent to the details of cooling below ambient, as the former temperature difference can be influenced by the physical properties of the setup and by heat exchange with the cooler ambient air outside the setup. In addition to heating of the setup, the relatively high ambient humidity during this summer outdoor experiment likely reduced atmospheric transparency, limiting the radiative cooling effect. This is a known issue with radiative cooling, which makes the process more effective in drier climates. Because of this, absolute comparisons between our aerogel coolers and the ambient temperature were performed during drier periods, and higher humidity seasons were used only for relative comparisons between different cooling materials. In the

Table 1. Comparison of Net Radiative Cooling Power (P_{net}''), Solar Reflectance (R_{sol}), Emissivity in the Atmospheric Transparency Window (ϵ_{ATW}), and Thermal Conductivity (k) of State-of-the-Art Ceramic-Based Radiative Coolers with This Work^a

material	P_{net}'' (W/m ²)	R_{sol} (%)	ϵ_{ATW} (%)	k (W/m·K)	ref
SiO ₂ (this work)	160	97.8 (±0.5)	99.2 (±0.3)	0.055 (±0.01)	
Si ₃ N ₄ /BN	78.55	~95	~95	3.75	54
Zn ₂ SiO ₄	97.4	97	95	N/A	55
ZrO ₂ /Al ₂ O ₃	125	97.7	95.6	N/A	41
Al ₂ O ₃	>130	99.6	96.5	N/A	42
Al ₂ O ₃ /glass	60	>96	~95	N/A	33
Al ₂ O ₃ /SiO ₂	133.1	95	93	0.03	56
SiO ₂	N/A	93.2	92.4	0.035	32
SiO ₂	128.52	97.8	92.3	N/A	43
SiO ₂ /PVA	44.54	93.7	98.19	0.039	57
TiO ₂ , PVDF, PDMS	80–103	97.3	>98	N/A	45
SiO ₂	93.2	96.2	97.5	0.023 (radial) 0.093 (axial)	58
SiO ₂ /cellulose	103.3	98.1	92.1	0.03	59
SiO ₂ /cellulose	~112	96	97	0.032–0.041	60
SiO ₂ /CaSiO ₃	96	94	96	0.0854	61

^aPVDF, PDMS, and PVA refer to polyvinylidene fluoride, polydimethylsiloxane, and poly vinyl alcohol, respectively.

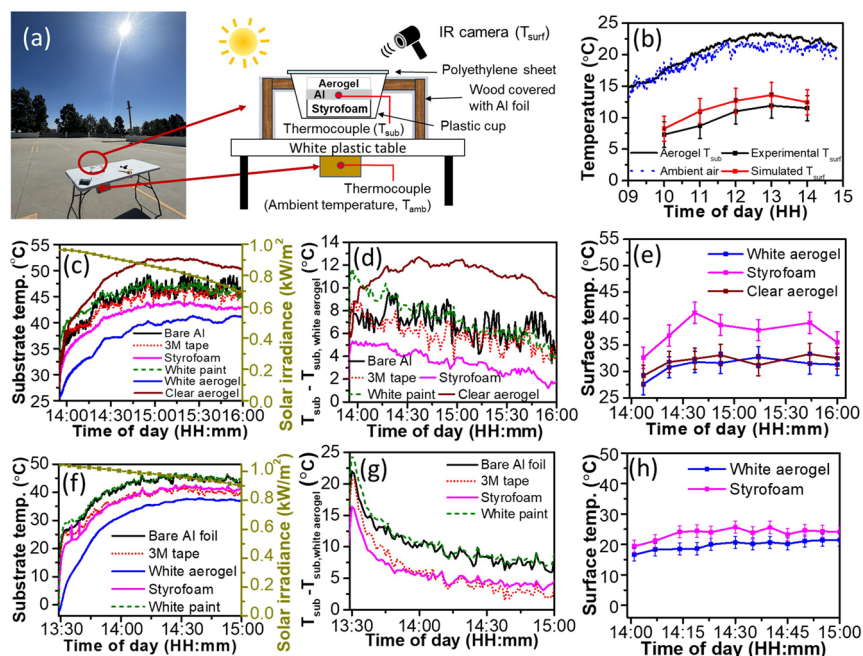


Figure 6. Proof-of-concept outdoor experiments conducted in Los Angeles, CA. (a) Photo and schematics of the setup and (b) experimental and simulated substrate and surface temperatures collected from the white aerogel on a sunny winter day. (c) The substrate temperature (T_{sub}), (d) the difference between T_{sub} for each control sample and T_{sub} for the white aerogel, and (e) surface temperature (T_{surf}) data collected on a colloid containing white aerogel sample, along with a family of control samples. Data was collected starting from ambient temperature on a sunny summer day. (f) T_{sub} , (g) the difference between T_{sub} for each control sample and T_{sub} for the white aerogel, and (h) T_{surf} data collected on a colloid containing white aerogel sample, along with a family of control samples with all samples and the experimental set-up starting at refrigerated temperature. Data was again collected on a sunny summer day.

following experiments, the sample chamber temperature was used for a qualitative comparison of cooling performance, and a range of other control samples were also used for comparison.

To better understand the cooling properties of these unique insulating gels, in a subsequent experiment, the radiative cooling power of the white, colloid-embedded aerogel was compared with that of a bare aluminum substrate and four different

control materials deposited on Al substrates: 3 M tape, Styrofoam, white paint, and clear aerogel without embedded silica particles (Figure S10, Table S2). Bare aluminum was a reference material that is not expected to show passive daytime cooling due to its absorptance in the solar spectrum ($R_{\text{sol}} = 0.89$) and high reflectance in the mid-IR ($\epsilon_{\text{ATW}} = 0$). This sample is analogous to a thermocouple exposed to direct sunlight, which is often compared with daytime radiative coolers in the literature.^{66,67} 3 M tape on aluminum was proposed by Raman and co-workers⁶⁸ as an easy-to-prepare and inexpensive radiative cooler that can be used as a standard radiative cooler due to the emissive property of the polymer-based tape ($R_{\text{sol}} = 0.76$, $\epsilon_{\text{ATW}} = 0.92$). Styrofoam was a thermally insulating white material with a reasonable reflectance ($R_{\text{sol}} = 0.82$), but which lacks emissivity in the ATW ($\epsilon_{\text{ATW}} = 0.64$). The white paint was diffusely reflective in the visible range ($R_{\text{sol}} = 0.75$) and emissive in the ATW ($\epsilon_{\text{ATW}} = 0.92$). Note that we used standard all-purpose white paint, rather than a paint formulated for cooling roof applications. Details of the preparation and spectral characterization of the white paint can be found in the Supporting Information (Figure S11, Table S2). Finally, the clear aerogel has solar absorptivity, IR emissivity, and thermal conductivity similar to white aerogel, but is transparent to solar radiation. The solar reflectance, mid-IR emittance, and thermal conductivity of these samples are tabulated in Table S1. Similar to the previous setup, all samples were prepared in a plastic cup filled with thermally insulating Styrofoam and silica fiber and had a PE cover. The outside of the wooden sample holder was additionally covered with silica fiber to reduce conduction to the samples.

This second, larger dataset was collected on a sunny summer day in Los Angeles. The substrate temperature of all the samples and the surface temperature of all thermally insulating samples were collected between 1:54 and 4 pm (Figure 6c–e). Among the six samples, the temperature of the substrate underneath the clear aerogel was the highest, while that of the substrate under the white gel was the lowest. Interestingly, the surface temperatures of the two aerogel samples were identical. The high substrate temperature of the clear aerogel sample was attributed to the fact that solar radiation easily transmitted through the clear aerogel and was subsequently absorbed by the Al substrate. The absorbed heat was then trapped in the Al substrate because it was sandwiched between the thermally insulating aerogel on the top and Styrofoam on the bottom. In contrast, the colloidal silica spheres embedded in the white aerogel effectively backscattered the incoming solar radiation, preventing direct heating of the substrate. Both aerogels experienced similar radiative heat loss to space on the surface, which resulted in similar surface temperatures. The nearly 20 °C temperature gradient on the clear aerogel shows that heat cannot readily dissipate through the thick aerogel due to its large thermal resistance. The substrate temperature of Styrofoam was the second lowest and was 2–5 °C greater than that of the white aerogel. The temperatures of the substrate covered with the 3 M tape or coated with white paint were similar and 4–11 °C hotter than the substrate covered with the white aerogel. These results indicate that the substrate temperature under direct solar irradiance was mostly controlled by the solar reflectance, rather than by the emissivity, which agrees with previous reports where radiative cooling through emissivity is negated by solar absorption in materials with low R_{sol} .⁶⁹ The surface temperature of the Styrofoam

was not significantly lower than that of the substrate underneath due to its low emissivity.

To study the effect of conductive heat transfer to the samples, the temperature of the wooden setup and the air temperature in an empty cup were collected simultaneously. To prevent direct solar absorption by the thermocouple, it was placed inside a white paper straw, which was sandwiched between Styrofoam and further covered with an aluminized mylar and a PE film (Figure S4). Compared to the temperature from the local weather station, which remained at 28 °C (± 1 °C), the temperature of both the setup and the air in the cup significantly increased upon exposure to direct sunlight, reaching 46–48 °C and 42–44 °C from 3 to 4 pm, respectively (Figure S12). From the IR camera, the temperature of the ground exceeded 60 °C, more than 30 °C above the air temperature (Figure S13). Such intense heat was likely conducted to the samples on the sample holder through the silica fiber and Styrofoam layers, resulting in elevated substrate temperatures, particularly for thermally insulating samples. This edge-effect was also observed in the IR images, where the rims of the samples are hotter than the center of the samples (Figure S14). Still, the substrate temperature of the white aerogel slab stayed 2–3 and 6–7 °C colder than the air in the cup and the setup, respectively, owing to its excellent solar reflectance, high emissivity, and thermal insulation.

The outstanding thermal insulation that could maintain a large temperature gradient, the high solar reflectance that prevents heat absorption from the sun, and the radiatively cooling surface that further inhibits heat gain, collectively make the colloid containing white aerogel an ideal material for keeping cold objects cold. The final set of experiments thus examined how well the different configurations could keep a cold substrate cool. (Figure 6f–h). The setup was loaded with five samples: bare aluminum substrate, aluminum substrate with 3 M tape, Styrofoam, white paint, and white aerogel and was cooled in a freezer overnight before being exposed to the sun. Here, the setup was only insulated on the bottom to ensure thermal equilibration of all the samples with the setup. Although a large ice pack was used to maintain the cold temperature of the samples while transporting the setup from the freezer to the outdoor setting, all samples warmed up to slightly different temperatures due to inherent handling issues. The initial temperature of bare Al, 3 M tape, white aerogel, styrofoam, and white paint were measured to be 3.8, 1.2, –2.2, 4.4, and –1.8 °C, respectively.

Upon exposure to direct sunlight, the initial rise in the substrate temperatures appeared to be driven by the solar absorption of the wooden setup, which subsequently conducted heat to each sample. Thus, each sample's initial rate of temperature increase was primarily dictated by the samples' thermal resistance. The temperature of the bare Al substrate, Al substrates covered with 3 M tape, and Al substrate coated in white paint increased rapidly at a similar rate, followed by that with Styrofoam and then by the Al substrate covered with the white, colloid-loaded, aerogel slab. While the temperature of the Al substrate with or without white paint remained similar throughout the experiment, the temperature of the substrates covered with 3 M tape and Styrofoam converged within 30 minutes. At all times, the white aerogel's substrate temperature was lower than all reference samples, by 4 °C compared to 3 M tape and Styrofoam, and by 8 °C compared to the bare Al substrate and with white paint during the 2–3 pm time window. The difference between the white paint and 3 M tape may be explained

by differences in their spectral selectivity and thickness. Although ϵ_{ATW} was the same, the broad absorption of the white paint beyond the ATW resulted in the absorption of the atmospheric radiation, thus diminishing its cooling power from radiation to space. This effect was more pronounced when the setup was cooled before the start of the experiment. As observed from the previous experiment, Styrofoam stayed cooler than the other conventional materials at both the top surface and the substrate side, despite having low ϵ_{ATW} , due to its high solar reflectance and high thermal resistance. The effect of the low ϵ_{ATW} was observed with Styrofoam whose surface temperature remained about 4 °C higher than that of the white aerogel slab. Interestingly, the top surfaces of both insulating slabs (Styrofoam and the white aerogel) in Figure 6h were able to maintain cooler substrate temperatures than those shown in Figure 6e thanks to their high thermal resistance which inhibited heat transfer from the hot surroundings.

For practical applications, a daytime radiative cooler should be able to withstand environmental weather conditions. To evaluate the practicality of our white aerogel for outdoor use, basic durability against UV radiation, water, dirt, and steam was tested. To test UV resistance, the white aerogel slab was left outdoors under direct sunlight for about 6 days during an extreme heatwave (Figures S5 and S15). The solar reflectance showed minimal changes after the testing, with a slight improvement in the near-UV range (Figure 7a). The increased reflectance in the near-UV was more pronounced in an old sample that had been poorly stored for over 3 years (Figure S16). The old sample exhibited reduced reflectance prior to solar exposure but returned to the higher reflectance after exposure. The decreased reflectance was likely due to the presence of UV-absorbing organic particles, such as dust or oil, which accumulated on the surface. These contaminants appear to have decomposed during the prolonged UV exposure. Importantly, in both cases, intense radiation from the sun did not induce adverse changes in the radiative cooling properties.

Next, we explored the stability of our porous radiative cooler to a range of common (and less common) environmental hazards. Water resistance was tested by contact angle measurement and actively submerging the gel into the water. (Figure S17, Movies S1 and S2) The contact angle measured for a sessile drop of water on the white aerogel was 124°. (Figure S17a) Due to this hydrophobicity, water droplets rolled off the surface when dispensed at a 45° angle. (Movie S1) The gel further stayed dry when fully submerged in water, forming a thin layer of air on the surface of the aerogel (Figure S17b). The combination of nano-sized pores and hydrophobic surface functionalization effectively blocked water from infiltrating the porous network, and the porous cooling materials were found to repeatedly floated once the force used to submerge them was released (Figure S17b,c, Movie S2). In addition, the small and uniform pore sizes made it difficult for micron-sized dust particles to penetrate through the pores. This fact, in combination with hydrophobic nature of the surface functionalized silica pores, provided some degree of self-cleaning properties. Upon pouring dirty water onto the aerogel, most of it rolled off or was removed by pouring clean water (Movie S3). The aerogel maintained its structural integrity when exposed to pressurized steam (Movie S4). While real materials are unlikely to be exposed to these conditions, pressurized steam can be considered a form of simulated aging, given that hydrolysis is the main degradation pathway in silica-based aerogels.³⁸ In addition, the

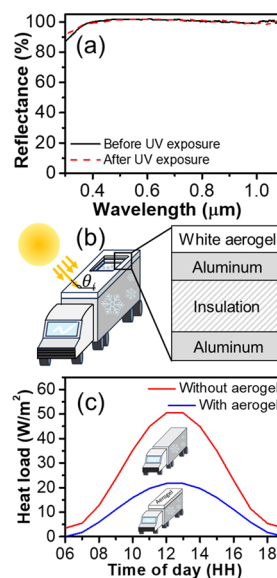


Figure 7. (a) UV-vis reflectance of the white aerogel before and after the prolonged UV exposure. (b) Schematics of a truck driving through a desert with an aerogel cover assumed in the simulation. The walls of the truck was assumed to be comprised of two aluminum layers with insulation in the middle. (c) Simulated heat load on the roof of a truck driving through a desert, with and without a white aerogel cover.

aerogel's ability to withstand local steam pressure of 3–3.5 bar demonstrates its structural robustness. The high mechanical strength of our aerogel arises from the durable aerogel matrix, which showed no discoloration or fracture throughout 20 days of harsh thermal cycles between −30 and 60 °C in the presence of humid air (Figure S18). The TGA data showed high thermal stability of the white aerogel, with minimal weight loss (~0.5 %, attributed to water) up to 300 °C, and about ~8.2% weight loss between 350–700 °C due to oxidation of organic components. These results suggest that hydrophobicity should be maintained up to 300 °C (Figure S19a). The white aerogel also exhibited fire-resistance, showing no discoloration or charring upon direct flame exposure (Figure S19b).

With this data with hand, we can consider applications where this material would find most utility. The combination of properties observed for the colloid-containing white aerogels, which show strong IR emissivity and prevent heat conduction and solar absorption, makes them interesting candidates for solar-reflective insulation to mitigate the heat load acting on refrigerated buildings or containers, especially in areas with large daily temperature swings such as a refrigerated truck passing through a desert. To examine this in more detail, this hypothetical situation was simulated using experimentally collected data on the white aerogel as an input. The heat load and the surface temperature of the roof of the refrigerated truck traveling across the desert with and without a white aerogel cover was modeled, using an average temperature swing from −4 to 38 °C and the solar irradiance measured in the Mojave Desert (Figure 7b,c).⁷⁰ Detailed schematics of heat fluxes, temperature profiles, and the thermal circuit used in the simulation are provided in Figures S20–S23. As observed from the outdoor cooling experiments, an Al substrate would heat up significantly above the air temperature during the day and would reach a peak temperature of 74.1 °C at 1 pm. This temperature was reduced by about 46 °C, to 27.6 °C by covering the Al roof with

the white aerogel slab. Similarly, the maximum heat load on the refrigerator under the bottom Al layer could be reduced to about 43%, from 50.5 to 21.7 W/m² by adding the white aerogel slab. These results illustrate that the intense solar irradiation absorbed from the exposed Al surface could be conducted through the 50 mm-thick insulation, despite having almost half the thermal conductivity and 14 times greater thickness than the white aerogel, imposing a significant heat load on the refrigerated container. The reduction in heat load with the aerogel cover is primarily associated with its high solar reflectance and infrared emittance, which prevents heating of the aluminum surface and maintains a lower thermal gradient. The low thermal conductivity of the aerogel further helps maintain the thermal gradient, together with the thick insulation layer. The reduction in energy consumed by the refrigeration unit simply by covering the roof with a white aerogel demonstrates a practical application where thermal insulation complements radiative cooling properties.

4. CONCLUSIONS

This study proposes thermally insulating radiative cooling monoliths consisting of size-controlled, well-dispersed colloidal silica embedded in ambiently-dried aerogel synthesized by inducing gelation of a solution containing colloidal silica and silica-based sol-gel precursors. The resulting white aerogel slab featured high solar reflectance (97.8%), high mid-IR emissivity (99.2%) and low thermal conductivity (0.055 W/m·K), making it an interesting daytime radiative cooler, that could simultaneously back-scatter most of the incoming solar radiation, create a cold outer surface, and thermally insulate low temperature samples. When the aerogel was placed directly under sunlight outdoors, its surface temperature remained 9–11 °C lower than the ambient temperature. In addition, both the surface temperature and the substrate temperature of the aerogel were lower than a range of promising control samples, including Al metal, 3 M tape, white paint, Styrofoam, and clear aerogel. Aerogel also showed strong durability under environmental conditions, highlighting its potential for practical outdoor use. In a hypothetical setting of an aluminum truck traveling across the desert, the peak heat load on the inner wall of the truck was calculated to be reduced to 43% when covered with the white aerogel. The white, colloid-containing aerogel's unique mixture of optical scattering, minimal absorption, radiative cooling, and thermally insulating property makes it ideal for applications where a temperature lower than ambient needs to be retained, such as water preservation or refrigeration. While further engineering efforts are needed to enable large-scale, low-cost production, the hierarchical aerogel architecture provides a promising design strategy for future radiative cooling materials and other applications that require tailored spectral reflection and absorption.

At a more fundamental level, this study shows that established building blocks, such as organo-silica gels or size-controlled colloidal silica, can be combined to create materials with new and interesting properties. While many material properties were carefully optimized for this work, the basic components have been known for decades. This work thus demonstrates the power of creative materials design, coupled with high level modeling, to repurpose established materials into designed composites that can help address the many energy challenges currently facing our society.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsaem.6c01136>.

Additional experimental and characterization details including aerogel preparation, details on outdoor cooling measurements, experimental methodology, simulation methodology, optical and thermal measurements, porosimetry, USAXS analysis, infrared imaging, durability testing, and reference material characterization (PDF)

Water droplet repellency test (MP4)

Water immersion test (MP4)

Anti-soiling test (MP4)

Steam exposure test (MP4)

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Notes

The authors declare no competing financial interest.

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