

RESEARCH ARTICLE

Mesoporous Organosilica-Based Infrared Metamaterials for Radiative Cooling of Windows

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ABSTRACT

Efficient radiative cooling requires an unobstructed view of the sky. However, the vertical facades of buildings see terrestrial features, which act as broadband infrared emitters of heat in the summer. As a result, for conventional materials, including building windows, terrestrial heat gain in the summer can negate or overwhelm a vertically-oriented surface's narrowband long-wave infrared (LWIR) emission to space. To address this challenge, here we develop a visibly transparent, LWIR-selective mesoporous metamaterial suitable for use on windows that is designed to emit selectively within the 8–13 μm atmospheric transparency window. This material, a highly porous organosilica network dried under ambient conditions known as an ambigel, leverages phonon-polariton resonances to achieve high and selective emittance in the transparency window. Experimental characterization and modeling suggest that the bond angle and length distributions of amorphous SiO_2 in this ambigel differs significantly from those in fused silica, highlighting a new degree of freedom for optical design. Field tests demonstrate that the ambigel maintains surface temperatures approximately 0.5°C lower than a conventional broadband emitter in representative conditions. Our results demonstrate a material design strategy for a visibly transparent infrared selective material that can enable effective radiative cooling for windows and building facades.

1 | Introduction

Cooling consumes nearly 20% of the electricity used in buildings worldwide today with this energy consumption expected to grow as the effects of climate change continue to intensify [1]. Motivated by the need to develop energy-efficient cooling methods, radiative cooling has emerged as a potential solution by enabling passive daytime cooling through emission of thermal

radiation to the cold of space [2–6]. By taking advantage of the atmospheric transparency window between 8 and 13 microns [7], surfaces can be designed to selectively emit thermal radiation through Earth's atmosphere and into space.

Most work in radiative cooling has typically considered sky-facing environments [2, 3]. For these coolers, high IR emissivity and good solar rejection are the key figures of merit. However, a large

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fraction of buildings' surface area is vertically oriented. Recent research has thus extended the application of radiative cooling to the vertical facades of buildings [8, 9], demonstrating that vertical facades such as walls and windows typically exchange heat with the ground across broadband long-wave infrared wavelengths but lose heat to the cold sky only through the 8–13 micron atmospheric transparency window, a fact that can cause significant heating in materials with broad band IR absorption/emissivity [8]. To overcome this challenge, spectrally selective emitters have been shown to outperform conventional broadband emitters by reflecting much of the heat emitted by the ground while simultaneously losing heat to the sky in the atmospheric window [10–12]. Moreover, in winter months, such spectrally selective materials can minimize heat loss to the ground, enabling a thermoregulatory effect across seasons. However, while many conventional materials exhibit some degree of selectivity within the atmospheric window [3, 13, 14], and have been used for radiative cooling applications, engineering selectivity by design in a scalable way remains a key challenge. Additionally, it is difficult to achieve this capability for transparent elements of facades such as windows while maintaining visual clarity.

One potentially effective approach to enable infrared selectivity within the atmospheric window has been to take advantage of the phonon polariton-driven resonances supported by a range of materials including amorphous oxides. For instance, various polymorphs of silica (SiO_2) have high emissivity within the atmospheric transparency window due to their phonon-polariton resonances and have been used for radiative cooling applications [15, 16]. It is generally assumed that the phonon polariton-driven dielectric function of silica and other oxides are fixed. Instead, one typically seeks to optically design around these constraints, a fact that has limited the performance enhancement. When fundamental materials properties can be tuned, more dramatic changes can be achieved. For example, it has been found that reducing the refractive index of silica has the potential to mitigate IR reflectance while maintaining high IR emission [17]. Previous work has also shown that a silicate material's infrared absorptance can be controlled by its underlying structure and the synthesis route used to make the material [18].

From a scalability and materials flexibility perspective, sol-gel chemistry is a particularly appealing synthesis technique to use in the design of silica-based materials. Taking advantage of decades of research into the sol-gel process, many aspects of silica-based systems can be designed [19]. Porosity is a well-controlled property in sol-gel silica, which has a direct impact on the gel's refractive index. The kinetics of the sol-gel process lead to the formation of a highly porous network, especially under rapid condensation reactions catalyzed by the addition of base [20–22]. From this point, the porosity of the product heavily depends on the procedures used to dry the gels [23–25]. Depending on the drying conditions used, sol-gel silica can produce xerogels with typically 40%–50% porosity [26–28], supercritically-dried aerogels with porosity values in excess of 90% [29, 30], and aerogels dried under ambient conditions, known as ambigels [31]. In contrast to supercritical drying, the silica network is dried under ambient conditions from a nonpolar, high-vapor-pressure solvent. The porosity of ambigels is generally tunable but can reach values as high as 90%, similar to the supercritical

drying route [32, 33]. The ambigel approach is of particular interest in terms of its scalability, which improves the prospects of widescale adoption.

Another design aspect made possible by sol-gel synthesis is the facile synthesis of materials with organosilane moieties. Organosilane precursors and surface passivating agents have been used to increase the porosity and flexibility of ambigels [34]. Alkyl groups directly bonded to silicon atoms act as a chain-terminating agent by preventing Si–O–Si bond formation and increasing porosity. Surface passivating agents react with silanol groups (Si–OH) on the surface of the gel, preventing condensation reactions and allowing greater spring-back effect [35]. Previously, Butts et al. optimized the synthesis of highly porous ambigels with optical transparency [31]. They used various ratios of organosilane precursors and surface modifying agents to prepare organosilane-based monoliths with ~ 1 mm thickness, $> 80\%$ porosity, and small pores < 20 nm with a narrow pore size distribution. However, incorporation of various functional groups also introduces IR absorptions outside the atmospheric transparency window, and that fact, combined with the large sample thickness, made those ambigels broadband emitters.

Because of the very strong phonon-polariton resonance in SiO_2 , thinner samples have the potential to saturate the silica mode without providing significant absorption outside of the atmospheric transparency window from other vibrational modes. Unfortunately, preparing ambigels with a thickness in this range (from tens to hundreds of micrometers), while maintaining a high porosity, is challenging. While micrometer-thick films (< 10 μm) of ambigels on flat substrates have been fabricated by multi-layer deposition of precursor solutions using spin-coating and dip-coating, or by doctor-blading [36–38], making a substrate-adhered ambigel with a thickness greater than 1 μm is challenging because the high volumetric shrinkage during gelling and drying results in cracking and peeling [39]. At the other extreme, free-standing ambigels can be readily made using a non-interacting substrate, where the ambigel can freely shrink. Ambigel monoliths millimeters to centimeters thick can be made in this manner, but a thickness less than 1 mm is challenging due to the fragile nature of silica ambigels. Recently, Bar et al. reported the synthesis of a 130 μm -thick free-standing aerogel with 90% porosity via sol-gel chemistry at the interface of two immiscible liquid molds [40]. However, they supercritically dried the gels, making the aerogel optically opaque. To the best of our knowledge, optically transparent ambigels in this thickness range have not been reported to date.

In this work, we report the synthesis of novel optically transparent, highly porous silica ambigels with ~ 90 μm thickness as a selective infrared emitter for radiative cooling of vertical surfaces. These ambigels take advantage of low-energy processing through sol-gel synthesis and ambient drying. We characterize their optical properties with Fourier Transform Infrared Spectroscopy (FTIR) and Raman spectroscopy, supported by an inverse design methodology to extract the silica ambigel's wavelength-dependent permittivity from measured infrared transmittance spectra. This approach allows us to accurately model the internal bonding structure within the ambigel, enabling optimization of thickness and porosity for enhanced infrared selectivity. Our ambigels achieve radiative cooling of $\sim 0.5^\circ\text{C}$ compared to a

broadband infrared emitter, corresponding to a net radiative cooling power of 50 W/m² emitted through the atmospheric transparency window and into space.

2 | Results and Discussion

2.1 | Synthesis and Characterization of Ambigel Monoliths

The emissivity observed for ambigels is generally broad-spectrum in nature due to the high thicknesses (> 1 mm) of most ambigel monoliths. By decreasing the ambigel thickness and increasing porosity, however, infrared selectivity could be enabled. Indeed, the absorbance of a material generally follows the Beer–Lambert Law, where absorbance (*A*) of a material is a product of its spectral absorptivity (*a*), optical length (*l*), and concentration (*c*) [31]. For the same material, overall absorbance decreases by decreasing thickness or increasing porosity, which correlates to reducing the optical path length and concentration. In organosilane-based gels, absorptivity arising from the Si–O–Si stretching vibrational mode in the atmospheric transparency window (8.3–11.1 μm, 900–1200 cm^{−1}) is orders of magnitude higher than absorption from organic functional groups, and the absorption band reaches saturation at just a few microns thickness [31, 41]. Therefore, by reducing the thickness of ambigel slabs, and/ or by increasing the porosity, it should be possible to reduce the undesirable absorption arising from the organic groups, while keeping the IR absorption from the silica backbone saturated.

To experimentally explore this possibility, ambigels were synthesized using a modification of an established sol–gel method that uses tetraethoxy orthosilicate (TEOS) and methyltrimethoxysilane (MTES) as silicate precursor and trimethylchlorosilane (TMCS) as a surface silylating agent [31]. For this work, the challenge was to find a substrate with the proper surface energy to make free-standing thin ambigels with a uniform thickness. Historically, substrates with a low surface energy have been used to make thick ambigels, enabling easy removal of the ambigel from the substrate, while substrates with high surface area have been used to prepare thin films with uniform thickness. To prepare thin, free-standing ambigels, a substrate should be inert enough to prevent covalent bond formation with our precursor solutions while also not being so hydrophobic that it causes the solution to bead up instead of spreading out. Polyethylene (PE) films were chosen for their balanced surface energies, as seen from the image of sessile drops of the major solvent in the solution, ethanol and water (Figure S1a,b). The contact angles measured from ethanol and water were 19° and 90°, respectively (Figure S1c). The moderate surface wettability enabled the synthesis of thin monolithic ambigels.

Figure 1 shows examples of data collected on six different ambigel samples with varying thicknesses and consistent porosity; thickness was controlled by dispensing varying volumes of solution onto the substrate and then aging for 2 days. The thicknesses of the ambigel samples were measured to be 75, 90, 120, 280, 380, and 420 μm from cross-sectional optical microscope images (Figure S5). Figure 1a and Figure S4a show the normalized normal-normal transmittance of the 6 ambigels. At all thicknesses, the IR transmittance of the ambigels was saturated at 8–10 and 11–13 μm

due to the high extinction coefficient of the Si–O–Si asymmetric and symmetric stretching, respectively. In contrast, the transmittance outside of the atmospheric transparency window decreased with decreasing thickness, thereby enhancing the selectivity of the IR emissivity.

The selectivity was further enhanced by increasing the porosity of the ambigel. The porosity of the ambigel samples increased from 84% to 88% and 91% by increasing the aging time from 2 to 3 and 7 days, respectively (Figure S6a). The peak of the pore size distribution also increased from 10.2 to 11.5 and 13 nm, respectively (Figure S6b). Increases in mean pore sizes and porosity with extended aging are consistent with literature results and are attributed to a sturdier silica backbone obtained from prolonged condensation during aging, which results in greater spring-back of the pores after drying [42, 43]. Compared to the 75 μm-thick ambigel slab with 84% porosity, the 80 μm-thick slab with 88% had increased transmittance outside of the atmospheric transparency window despite the slightly greater thickness, further increasing the selectivity (Figure 1b, Figure S4b). However, as the pore size and porosity increased, the ambigels became more opaque and fragile. Thus, ambigels with 88% porosity were selected as the ideal compromise of selectivity and optical transparency and used for more in-depth studies as a selective IR emitter for radiative cooling.

For the optical studies presented below, a 2 cm × 2 cm ambigel monolith with ~90 μm thickness and 88% porosity was synthesized (Figure 2). The ambigel was optically transparent, having > 96% transmittance and < 11% haze in the visible spectrum (Figure 2d). Further haze measurements are shown in Figure S14. N₂ porosimetry revealed a typical type IV isotherm, with a high BET surface area of 945 m²/g, and a narrow pore size distribution (Figure 2e,f). SEM images of the ambigel confirmed a uniform distribution of mesopores throughout the whole gel, with no signs of macropores (Figure 2c, Figure S7). The hydrophobic nature of the ambigel slab was confirmed by a water contact angle of approximately 128°. (Figure S2)

To understand the IR absorption contributions from organic functional groups in our ambigels, the IR absorbance was compared with fused silica and ambigels that were calcined at temperatures hot enough to remove all organic functional groups. (Figure 3a–c). The absorbance of each sample was calculated as Absorbance = 1–Transmittance Reflectance, as shown in Figure S8. As expected, fused silica had a reflectance peak centered around 9 μm due to a surface phonon-polariton resonance. This reflectance was significantly reduced in both the ambigel and the calcined ambigel due to their porosity and reduced refractive index. The minimal IR reflectance of ambigels and calcined ambigels supports their viability for radiative cooling applications. Calcination at 500°C in air burned off the organic moieties in the ambigels, leaving behind silanol groups on the gel's surface. IR absorption peaks and the relevant vibrational modes are tabulated in Table 1. The undesirable absorptions at 7.1, 7.8, and 7.9 μm associated with Si–C stretching vibrational modes were removed by calcination, however, the newly formed Si–O–Si bonds broadened the bond angle distribution in the calcined ambigel, broadening absorption beyond our desired IR window and offsetting the selectivity gained by removing the organic groups. In the atmospheric transparency window,

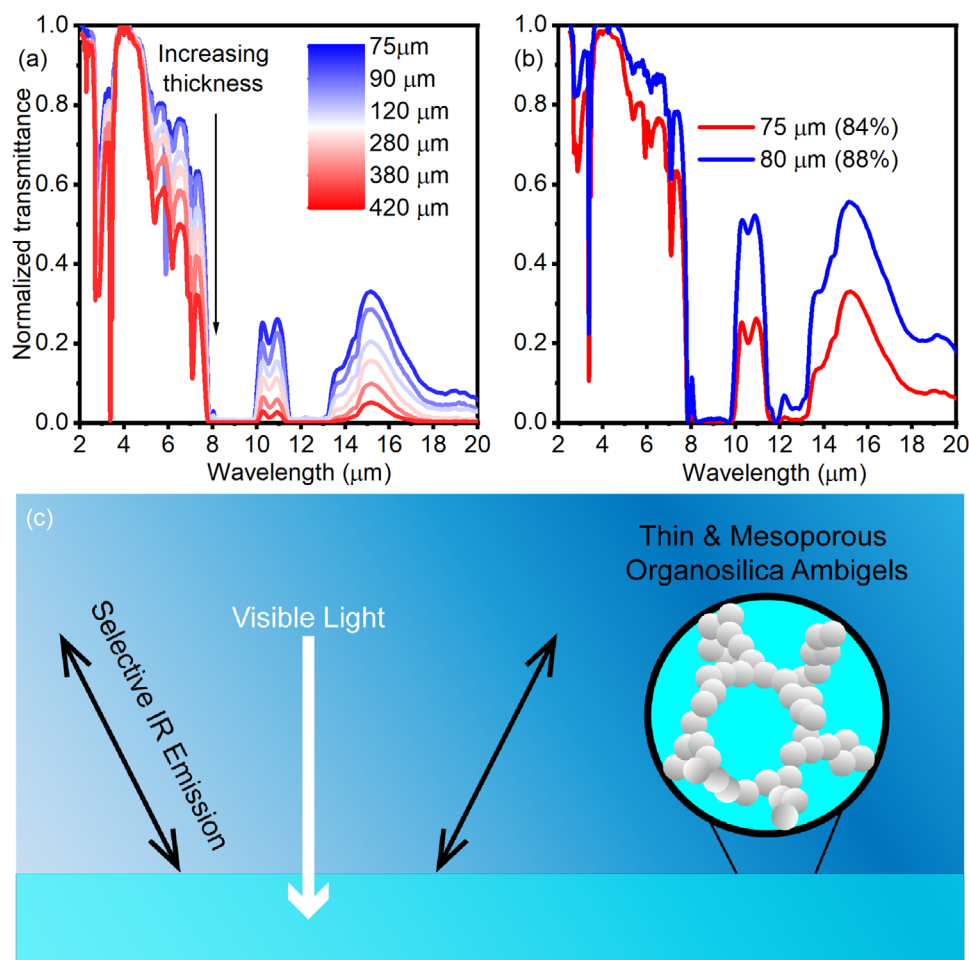


FIGURE 1 | Infrared spectroscopy showing how selectivity of ambigel samples is influenced by both micro- and nanostructure. (a) FTIR spectra of ambigel films synthesized with varying thicknesses at porosity of 88%, showing increased selectivity with decreasing thickness due to reduced absorbance from Si-CH₃ and Si-OH modes outside the 8–13 μm region. (b) FTIR spectra of ambigel samples with varying % porosity, again showing increased selectivity with a higher porosity of 88% compared to 84%. Porosity values were determined from nitrogen isotherms taken at 77 K. (c) Schematic of the proposed use of thin ambigels as a visibly transparent radiative cooler.

calcining reduced absorption associated with Si-C stretching and bending modes in 11.5–13.5 μm while enhanced absorption associated with Si-O stretching mode in silanol groups at ~10.6 μm. Due to the trade-offs in the IR absorption, overall selectivity of calcined ambigels was comparable to that of uncalcined ambigels, although without the hydrophobic organic coating, the chemical stability is assumed to be lower in the calcined materials.

Raman spectroscopy provides further insight into the altered dielectric function of the as-synthesized silica ambigels through a variety of vibrational features (Figure 3). As discussed above, these sol-gel derived ambigels are not purely SiO₂, as organic modifiers (primarily methyl groups) are present throughout the ambigel due to MTES precursor and TMCS surface treatment. The Raman spectrum of the ambigel more clearly shows the peaks attributed to organosilicate vibrational modes than the IR absorption because the Si-O-Si modes are not as strong in Raman. Full labeling of the peaks present in the ambigel is shown in Table 1 and Figure S10. Many of these can be correlated with Si-CH₃ vibrations found in PDMS, as there are similar bonding environments in both materials [47]. Thus, the Raman spectrum of the ambigel appears as a mixture of amorphous silica and

organosilica [56]. This is confirmed by comparison to Raman data collected on the calcined ambigel samples, which do not contain the organic groups. After calcination, the Raman spectrum of the ambigel becomes nearly identical to that of standard amorphous silica. The only major difference between the two is the large peak at 980 cm⁻¹ in the calcined ambigel, which arises from Si-OH groups on the unpassivated silica surface [57].

Another interesting difference between the ambigel and the two pure silica samples is the prevalence of ring subunits in the ambigel samples. These are observed as sharp peaks at 610 cm⁻¹ for a three-Si ring, and at 490 cm⁻¹ for a four-Si ring. As seen in fused silica, these ring peaks are typically much smaller than the standard scattering associated with Si-O-Si bonds, and are typically referred to as “defects” in the literature [58]. Previous studies on silicate glasses have shown that in porous silica, three-Si siloxane rings are especially prevalent [59, 60]. It is known in the sol-gel literature that the three-Si ring in particular is sterically unfavorable due to high strain [61]. These rings are likely a byproduct of the kinetics of sol-gel synthesis; when base catalysis is used, there will be rapid condensation of the silicate network. This can force the Si-O-Si bonds into

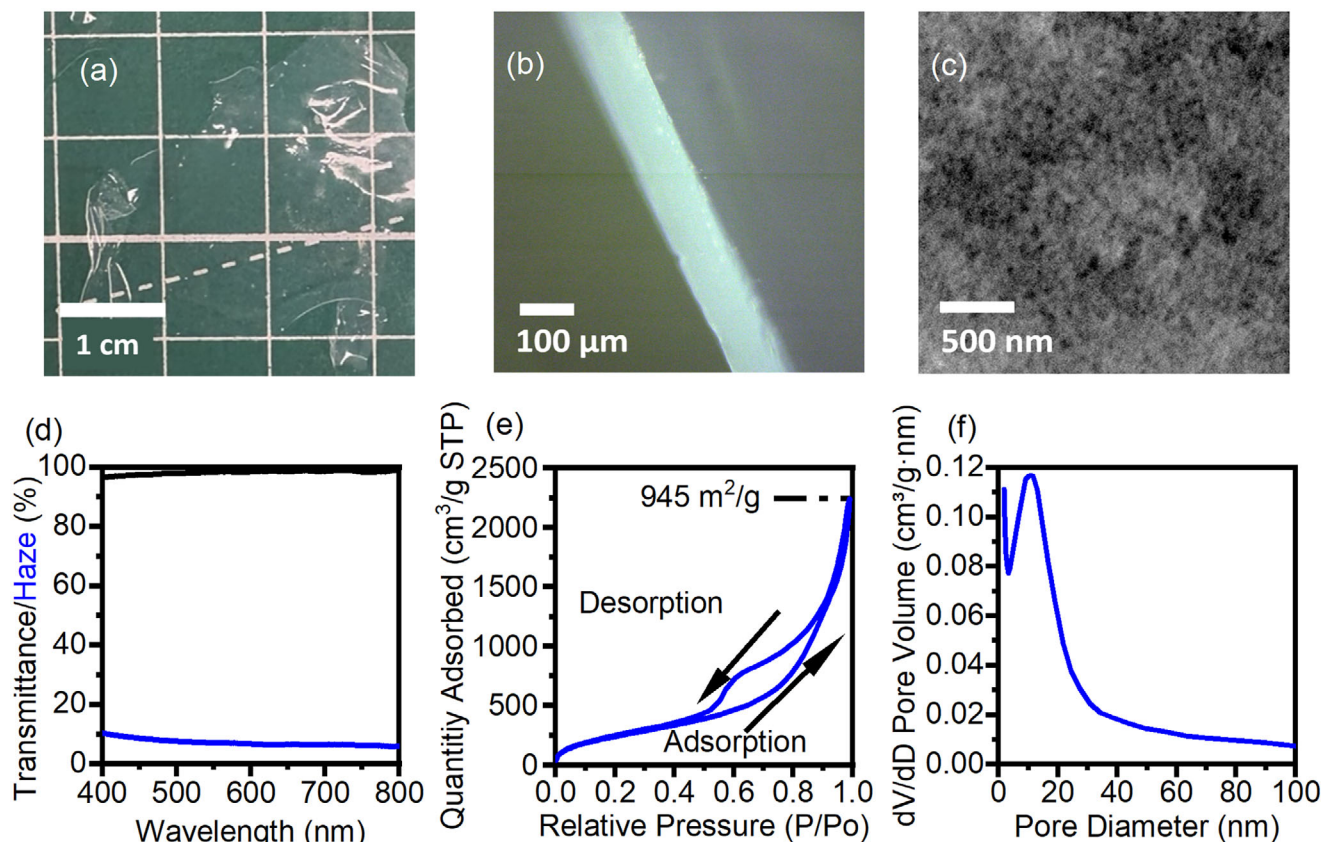


FIGURE 2 | Physical characterization of ambigel slabs. (a) Optical image of an ultra-thin ambigel, with a size of roughly $2 \times 2 \text{ cm}^2$, and a high degree of optical transparency. (b) Cross-sectional optical microscope image of an ambigel sample, showing a thickness of roughly $100 \text{ }\mu\text{m}$. (c) SEM image of ambigel, showing a uniform mesoporous nanostructure. (d) Transmittance and haze measured on the ambigel in the visible region. (e) Nitrogen adsorption/desorption isotherms for the as-synthesized ambigel. (f) Ambigel adsorption pore size distribution calculated from the adsorption shown in (e).

high-strain configurations. After calcination, the number of three-Si rings decreases dramatically, but the 490 cm^{-1} peak remains more intense than in fused silica. Overall, the Raman data indicate that as-synthesized ambigels have higher amounts of both three-Si and four-Si siloxane rings compared to non-porous amorphous silicates.

While viewing the ambigel as a mixture of amorphous silica and PDMS is a useful model, it is not an entirely accurate picture of the nature of organics in the ambigel system. PDMS contains entirely D^2 silica, with each silicon atom bonded to two organic groups and two other Si atoms through Si—O—Si bridges. The reactants used in ambigel synthesis are not expected to provide D^2 silica. MTES is used as a silicate precursor, which can be expected to form T^3 coordination, in which three Si—O—Si bonds can be formed alongside one Si— CH_3 group. Similarly, TMCS is expected to form M^1 coordination, where one Si—O—Si bond is formed in addition to the three existing Si— CH_3 groups. Of key importance in these distinctions is that the different atomic coordinations are expected to cause small shifts in the Raman spectra of the silicates [62]. As computed by Shimizu and Murata, the Si— CH_3 symmetric stretch and asymmetric bend in M^1 TMCS will be slightly blueshifted in comparison to either D^2 PDMS or T^3 MTES [63]. Indeed, the measured Raman spectra support this expectation: peaks at 1275 and 1416 cm^{-1} corresponding to CH_3 bending modes show a clear blueshifted shoulder, indicating that

multiple unique silicate bonding environments are present in the ambigels.

Finally, peak widths observed in the Raman spectra provide insight into the bond-angle distribution of Si—O—Si bonding in the ambigel. The porosity of the ambigel should allow strained bonds to relax, resulting in a bond angle distribution that should be narrower than in a dense amorphous material, such as fused silica. This was analyzed via the Si—O—Si asymmetric stretching mode in both the ambigel and fused silica. This mode is four times narrower in the ambigel, thus confirming the structural difference (Figure S11). The other relevant parameter that can be measured with Raman spectroscopy is the average bond angle, which is determined from a shift in the peak location. In the ambigel, Si—O—Si asymmetric stretching peak shifts appreciably, moving from 1064 to 1097 cm^{-1} . This blue shift corresponds to an increase in bond angle, which is also supported by a minor red shift of the Si—O—Si symmetric stretch from 810 to 798 cm^{-1} , as detailed by Geissberger and Galeener [64]. The blue shift is not observed in the calcined sample, suggesting that the increased bond angle is metastable and relaxes upon heating, like the three-Si siloxane rings. Another key point is that the high strain present in three-membered and four-membered siloxane rings means that those Si—O—Si bonds will not undergo standard network vibrations, and thus they will not affect the bond angles studied through the standard phonons of amorphous silica [64].

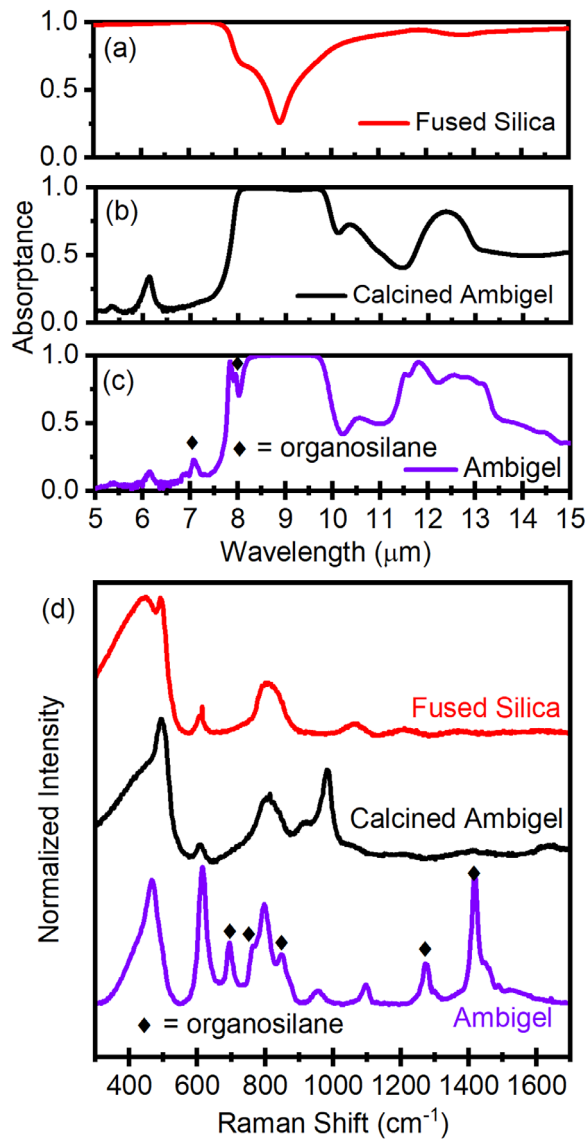


FIGURE 3 | Vibrational characterization of ambigels. (a–c) ATR-FTIR absorbance and (d) Raman scattering of ambigel samples (as-produced and calcined at 500°C to remove organic groups), and a fused silica reference.

2.2 | Modeling and Simulation

The experimental data shown here make a strong case that organosilane ambigels are a distinctive class of materials with strong potential for radiative cooling applications since their refractive index can be engineered through controlled variations in both thickness and porosity. To fully exploit these materials in optical applications, a model of their optical behavior is needed that fully captures the differences between the ambigels and materials such as pure amorphous SiO₂ or PDMS. Fortunately, the ambigel pores are sufficiently small (~10 nm) that they will not produce scattering in the LWIR region. Thus, the Maxwell-Garnett approximation [65] can be used to determine the dielectric permittivity of the ambigel and is given by

$$\epsilon_{eff} = \epsilon_m \frac{2\delta(\epsilon_i - \epsilon_m) + \epsilon_i + 2\epsilon_m}{2\epsilon_m + \epsilon_i - 2\delta(\epsilon_i - \epsilon_m)} \quad (1)$$

Here, ϵ_{eff} represents the dielectric permittivity of the bulk ambigel, and ϵ_m is the dielectric permittivity of the organo-silicate backbone that forms the ambigel matrix. The organo-silicate backbones are composed of amorphous SiO₂, organics, and water. Thermogravimetric analysis (TGA) shows that the ambigels are by weight 3% water, 6% organics, and 91% amorphous SiO₂ (Figure S12). The closest polymer whose optical constants can describe the organics within the ambigel is polydimethylsiloxane (PDMS) [66]. While D² coordinated Si atoms in PDMS are not an exact match to the M¹ and T³ bonding environments displayed in the ambigel, as discussed above, Raman spectra confirm that this difference is relatively small. Thus, PDMS is a suitable starting point to model our organo-silicate modes. About 40% of the weight of a PDMS monomer is amorphous SiO₂. Thus, the dielectric permittivity of the organo-silicate backbone can be modeled as a weighted average of the weight percent of its constituents: 15% PDMS, 3% water, and 82% amorphous silica. Here we use ϵ_i as the dielectric permittivity of air that fills the deep-subwavelength pores and δ as the porosity, i.e., volume fraction of air.

With the published refractive index data for water [67], silica [68], and PDMS [66], the dielectric permittivity for the ambigel matrix can be calculated using Equation (1). Solving the Fresnel equations via the Transfer Matrix Method (TMM) for a slab of ambigel with air on either side yields a theoretical transmittance response. Comparing the simulated transmittance function with an ambigel with equivalent parameters shows a large discrepancy (Figure S15). The difference between experiment and simulation suggests that this model for the dielectric permittivity does not accurately capture the phonon-polariton modes in fused silica or the organic vibrational modes. Raman and FTIR spectroscopy similarly showed that the bond angle distribution of the phonon-polariton resonances in the organosilane ambigel has shifted relative to what is seen in fused silica (Figure 3).

To understand how the dielectric permittivity of silica changes, it must first be modeled analytically. Prior work has demonstrated [69] that a Lorentz model [70, 71] can effectively capture the phonon-polariton resonances of conventional bulk amorphous silica as

$$\epsilon_{SiO_2}(\omega) = \epsilon_\infty \left(1 + \sum_{j=1}^3 \frac{\omega_{LO,j}^2 - \omega^2 - i\gamma_j\omega}{\omega_{TO,j}^2 - \omega^2 - i\gamma_j\omega} \right) \quad (2)$$

The Lorentz model employs three oscillators, each characterized by a longitudinal optical phonon ω_{LO} , transverse optical phonon ω_{TO} , and a dampening factor γ_j . Additionally, ϵ_∞ represents the high-frequency dielectric constant. This results in a total of nine parameters – six phonon frequencies and three dampening factors that define the dielectric permittivity of amorphous silica. Using inverse design, a parameter sweep can identify the correct combination of phonon frequencies and dampening factors that yield a dielectric function which, when applied through the Maxwell Garnett equation and TMM, reproduces the experimentally measured transmittance spectrum. We aim to explore this parameter space using a genetic algorithm, a widely used optimization method inspired by Darwin's principle of survival of the fittest [72].

TABLE 1 | Vibrational modes observed in as-synthesized ambigels (see Figure 3).

Wavelength (μm)	Wavenumber (cm^{-1})	Vibrational Mode	Activity	Refs.
20.8	440	Si—O—Si network bend	R, IR	[44, 45]
20.4	490	Four-membered siloxane ring	R	[46]
16.3	614	Three-membered siloxane ring	R	[46]
14.6	687	Si-CH ₃ symmetric stretch	R	[47]
14.1	708	Si-CH ₃ symmetric rock	R	[47]
12.8	787	Si—C asymmetric stretch	R, IR	[47, 48]
12.5	798	Si—O—Si symmetric stretch	R, IR	[45, 49]
11.7	862	Si-CH ₃ symmetric rock	R, IR	[47, 50]
10.6	954	Si-OH stretching	R, IR	[50, 51]
9.1	1096	Si—O—Si asymmetric stretch	R, IR	[45, 52]
7.8	1275	CH ₃ symmetric bend	R, IR	[47]
6.9	1416	CH ₃ asymmetric bending	R, IR	[47]
6.2	1622	H ₂ O	IR	[53]
5.9	1691	Si—O stretch (SiOH)	IR	[50]
3.4	2962	C—H stretching	IR	[54]
2.9	3500	O—H stretch in H ₂ O	R, IR	[55]
2.7	3650	SiO-H stretch	R, IR	[50]

The genetic algorithm is initialized by generating a population of parameter sets consisting of randomly generated values of 9 parameters (ϵ_∞ , ω_{LOj} , ω_{TOj} , γ_j with $j = 1, 2, 3$). From there, transmittance spectra are simulated for a 100-micron-thick ambigel with 88% porosity using the previously described procedure. The fitness of each candidate dielectric function is then evaluated by calculating the sum of differences between the simulated and experimentally measured transmittance spectra for the same ambigel configuration. A lower sum indicates a more accurate simulation, and therefore a more accurate dielectric function for the fused silica in the ambigel, corresponding to a higher fitness for a given set of parameters (an individual in the population).

After evaluating the fitness of each dielectric function, the genetic algorithm selects high-fitness parameter sets (and applies genetic crossover to generate a new population of parameter sets. Recombination is done via uniform crossover, in which each parameter is treated independently, and a random decision is made at each parameter position whether to swap values between two parent parameter sets. This process is repeated until a new population of the same size, but with a recombined set of parameters, is generated. Following crossover, random mutations are introduced by selecting parameters at random and replacing them with new, random values. This mutation step helps prevent premature convergence of local minima and encourages exploration of the broader parameter space in search of a global minimum. Each full cycle of fitness evaluation, selection, crossover, and mutation constitutes one generation.

Using this approach, we can numerically determine the dielectric function shown in Figure 4a. When used as input into the TMM, the output transmittance spectrum closely matches the experimentally measured data for the ambigel, as shown in Figure 4b.

A common challenge in electromagnetic simulations is the issue of nonuniqueness, where multiple dielectric functions can yield similar transmittance spectra. To address this, parameter space is constrained to a physically realistic set of values determined by Gunde [56] for silica phonon-polariton resonances. Rather than allowing arbitrary or random values, the phonon frequencies ω_{LOj} and ω_{TOj} , and the dampening factors γ_j are limited to a subset of values that correspond to physically realizable vibrational modes in silica.

The dielectric function in Figure 4a reveals insights into the structural changes in the amorphous silica. Compared to the experimentally determined dielectric function of fused silica [68], the silica in the ambigel exhibits a dampened low-frequency phonon, increased intensity of the 800 cm^{-1} phonon peak, and a slight damping and blueshift of the high-frequency phonon. These structural changes are validated by the Raman spectra of the ambigel in Figure 3. The Raman studies reveal a broad range of vibrational modes between 400 and 520 cm^{-1} . This broadening is attributed to increased structural disorder within four-membered Si—O—Si rings, which exhibit a wider distribution of bond lengths. Compared to fused silica, the ambigel also shows a much sharper peak around 800 cm^{-1} , indicating a narrower bond angle distribution associated with the symmetric stretching of Si—O—Si bonds (Figure S11). Additionally, the blue shift of the high-frequency peak suggests an increase in average bond angle [64].

2.3 | Outdoor Radiative Cooling Experiment

After synthesizing 1–4 cm^2 ambigel monoliths with a thickness of about 90 μm , a porosity of 88%, and optimal long-wave

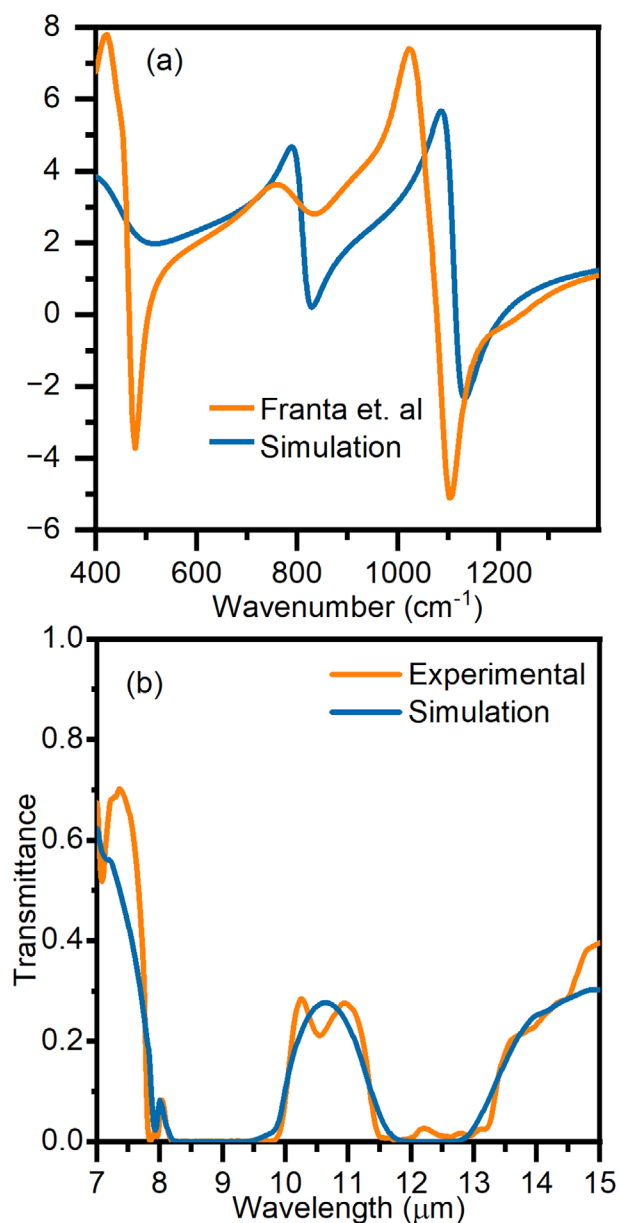


FIGURE 4 | Part (a) shows the dielectric function of fused silica [50], alongside the result obtained through inverse design using a genetic algorithm. (b) The simulated transmittance spectrum, based on the inferred dielectric function, exhibits excellent agreement with experimental measurements.

infrared (LWIR) selectivity, a controlled outdoor experiment was performed to quantify their radiative cooling performance relative to a broadband emitter when facing a warm ground. Figure 5a presents a schematic of the experimental setup and Figure 5b displays a photograph of the ambigel device (left) and the reference device (right) arranged for outdoor measurement. IR emittance data, shown in Figure 5d, demonstrate that the ambigel exhibits strong LWIR selectivity within the 8–13-micron atmospheric transparency window, while the reference thick PDMS sample behaves as a broadband emitter with comparable UV and visible reflectance to the ambigel (Figure S13). Figure 5c shows the view facing the ambigel and reference devices, with the warm swimming pool maintained at 24°C to provide a

constant ground temperature. Measurements were conducted over a 5 h period, from 1 to 6 a.m., to model the thermal gradient experienced between building facades and the ground under typical summer conditions [8]. During this time, the ambient air temperature steadily decreased from 9.1 to 6.7°C (Figure S20), while the dew point remained stable at −4°C.

Figure 5e shows the temperatures of the ambigels and the reference broadband emitter the ambigels, which are both significantly below ambient air temperature even while facing the warm ground. However, the ambigels maintain an average temperature 0.5°C cooler than the reference broadband emitter over the 5 h measurement window. These observed temperature differences under controlled conditions indicate a fundamental variance in heat flow, which serves as a proxy for potential real-world energy savings. We compare this temperature difference against radiative cooling simulations (see Supporting Information for details) in Figure 5f. Selectivity at varying angles of incidence confirms that the observed radiative cooling performance originates from the intrinsic selective emission of the ambigel rather than from angular emission effects (Figure S9). These simulations, which consider weather conditions, including humidity and wind speed (Table S1), predict a temperature difference at steady state (Figure 5f) which is in good agreement with experimentally measured data. These simulations also confirm that low absorption of heat from the ground and atmosphere is the driving force behind selectivity in ambigels (Figure S16).

For practical applications, the mechanical durability of the radiative cooler is essential. To evaluate durability, a thicker ambigel slab was attached to soda-lime glass and exposed to simultaneous cycles of temperature fluctuations, UV irradiation, and humidity [73] (Figure S3). The ambigel-coated glass showed negligible visual changes throughout the accelerated aging test, with the transmittance decreasing slightly from $90 \pm 1\%$ to $89 \pm 1\%$ after 30 days (Figure S3). The spectral selectivity of the ambigel is also preserved when adhered to an FTO-coated glass substrate, showing that it is capable of providing an optically transparent and IR-selective window coating (Figure S19). The cooling performance of ambigels under more extreme weather conditions are also considered, with simulated cooling performance in both winter (Figure S17) and extreme summer heat (Figure S18), showing the practicality of radiative cooler use in real-world conditions.

3 | Conclusion

In this work, we have used sol-gel chemistry to design an organosilica ambigel with high porosity and controlled thickness that can minimize absorbances outside the 8–13 μm atmospheric transparency window while ensuring saturation of absorption within the window. With careful control over the synthetic parameters of the ambigel, an optically clear gel with a thickness of ~90 μm could be obtained, representing a novel length scale in the synthesis of ambigels. Optical characterization reveals that these gels exhibit properties consistent with a mix of amorphous silica and organosilica, with phonons shifted due to modified bond angles. A genetic algorithm was used to map the dielectric function of the as-synthesized ambigels, leading to accurate fitting of the features seen in spectroscopy. Given the selectivity

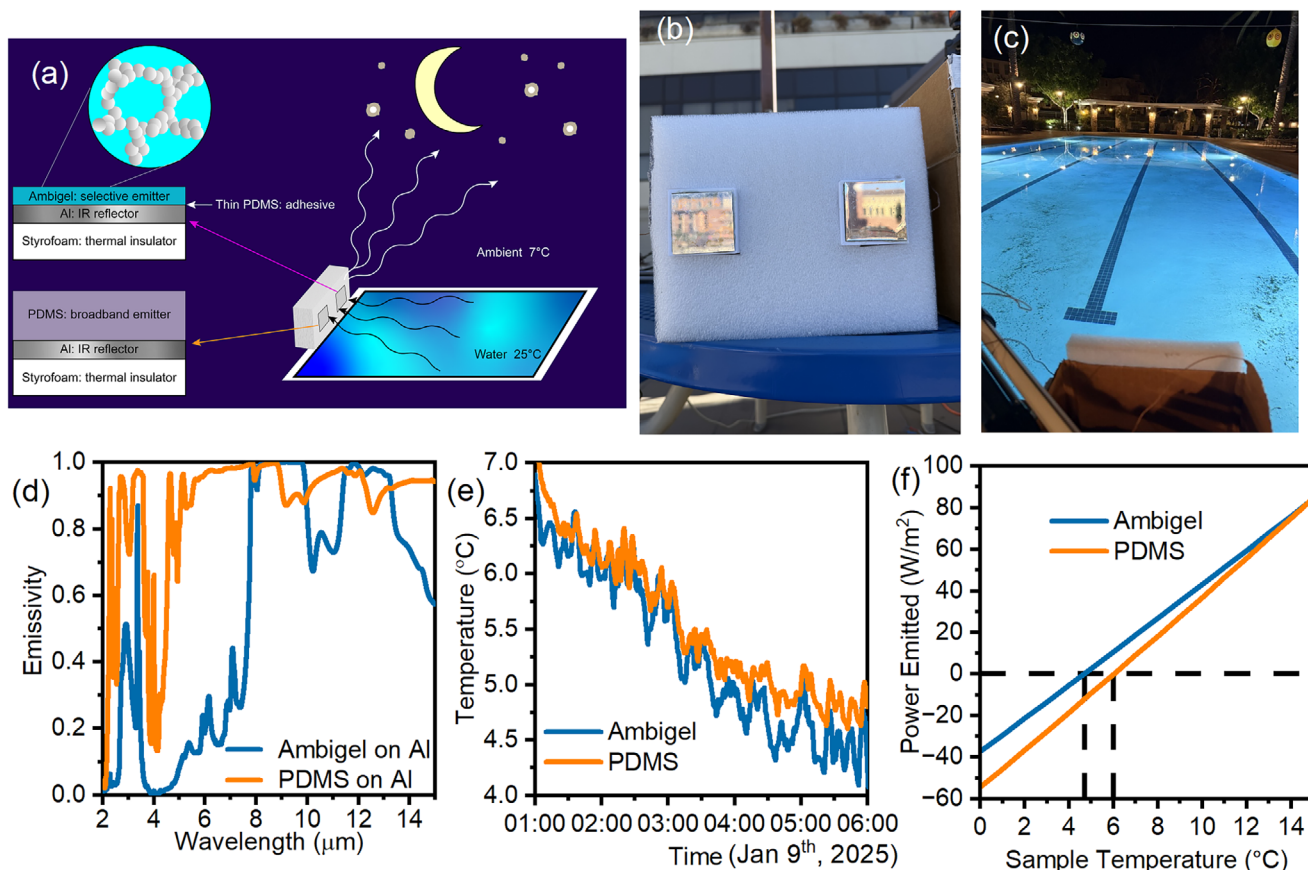


FIGURE 5 | (a). Schematic of the radiative cooling experimental setup, illustrating the vertically oriented samples facing the warm pool. (b) Both samples were mounted in a 3D-printed holder embedded in thermally insulating foam. (c) A photograph from the perspective of the samples shows their orientation toward the warm pool. (d) Infrared reflectance spectra of the test ambigel and the PDMS reference sample after being adhered to an aluminized silicon substrate. (e) Temperature measurements of both samples over a 5 h window. (f) Radiative heat transfer simulation demonstrating excellent agreement with experimental temperature data.

of the as-synthesized ambigels in the atmospheric transparency window, they were used in an outdoor cooling test. When facing a heated pool at nighttime, the ambigels were able to cool 0.5°C below a broadband emitter in a vertical configuration. These temperature differences reflect a fundamental difference in radiative heat flow between the two emitters and serve as an indicator of potential real-world energy savings. One use case for this technology is as a window coating, as the high transparency and low haze of the ambigel should not affect visibility of windows, yet the cooling effect achieved through selective emission could help reduce temperatures of buildings passively, allowing for reduced energy usage. Future work, including full building-integrated assessments as well as realistic room thermal mass, airflow, and indoor–outdoor temperature differentials represent an important next step in demonstrating the potential of this class of materials for window applications.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting File: sml175152-sup-0001-SuppMat.docx.