

Machine Learning Inverse Design Reveals a Double Narrow-Band Absorption Approach for Effective Colored Radiative Cooling Paints

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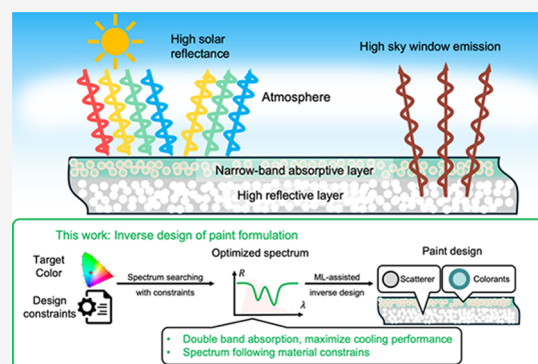
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Supporting Information

ABSTRACT: Colored radiative cooling paints offer immense potential for aesthetic thermal management. However, previous studies have been limited to either intuition-driven forward designs that are too expensive to yield non-intuitive optimal designs or to optimizations only at the level of idealized optical spectra. We present a machine-learning-enabled inverse design framework for generating paint formulations with optimal cooling performance and desired color. By integration of photon Monte Carlo simulations with surrogate modeling, this approach bridges the gap between theoretical spectra and physical realization. Our approach uncovers a non-intuitive “double narrow-band absorption” strategy that reduces solar heating power by up to 193 and 37 W/m² compared to conventional additive and single-band subtractive strategies, respectively. A systematic exploration of the HSL color space reveals that 24% of colors can achieve daytime subambient cooling when realistic material limitations are considered. This automated pipeline provides practical guidelines for designing high-performance, aesthetically tailored radiative cooling coatings.

KEYWORDS: colored radiative cooling paints, machine learning, inverse design, color science



With the growing need of reducing energy consumption and mitigating global warming, passive daytime radiative cooling (PDRC) technologies have gained significant attention as a passive, environmentally friendly cooling solution.^{1–3} PDRC materials function by reflecting incoming solar irradiation while emitting thermal radiation within the atmospheric transparency window (“sky window”, 8–13 μm) into deep space, enabling subambient cooling even under direct sunlight.^{4–6} It has been used for a variety of applications, such as building thermal management,⁷ energy harvesting,⁸ solar cell,⁹ agriculture,¹⁰ and personal thermal regulation.¹¹

The pursuit of radiative cooling began in the late 20th century. Daytime subambient cooling was reported on a winter day using a thin layer of commercial TiO₂/PVF paint coated on an aluminum substrate. However, not much detail was given, and the needed high solar reflectance was probably much boosted by the metal substrate rather than the intrinsic reflectance of the coating alone. Over the subsequent decades, the field has witnessed remarkable growth. Highly efficient daytime radiative cooling has been achieved through various approaches, including multilayer photonic structures,^{4,12,13} metal–dielectric composites,⁵ ultrawhite paint,^{14–18} textiles and fabrics,¹⁹ dual-layer thin films,²⁰ hierarchically porous polymers,²¹ aerogel,²² hydrogel,²³ non-reciprocal thermal radiation,^{24–26} etc. However, most high-performance radiative cooling materials appear white or silver, as they are designed to reflect a broad portion of the solar spectrum. While this maximizes cooling efficiency, it restricts the color options. In

many practical applications, color plays a critical role in aesthetics, visual harmony, and user experience.^{27,28} Moreover, the high reflectivity of white coatings can cause glare and visual discomfort in populated areas.

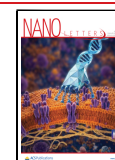
To address this need, researchers have begun developing colored radiative cooling materials that combine energy-saving capabilities with color need.^{29–34} One approach to achieve color in radiative cooling materials is through structural coloration with micro/nanostructured surfaces, including multilayer films,^{35–39} microcone arrays,⁴⁰ bilayer systems,⁴¹ and plasmonic structures.⁴² While these designs can offer vibrant colors, they often require complex and expensive fabrication, limiting scalability. In contrast, paint-like materials offer greater flexibility and scalability for applications.^{15,16,43,44} Since the broad band absorption (additive color) would lead to large thermal gain, researchers have proposed the subtractive color strategy that selectively absorbs a narrow band of light. Progress has been made using bilayer coatings,²⁹ quantum dots,^{30,31,45} phosphor dyes,³² and chemical absorbers.^{46,47} However, the development of paint formulations is largely

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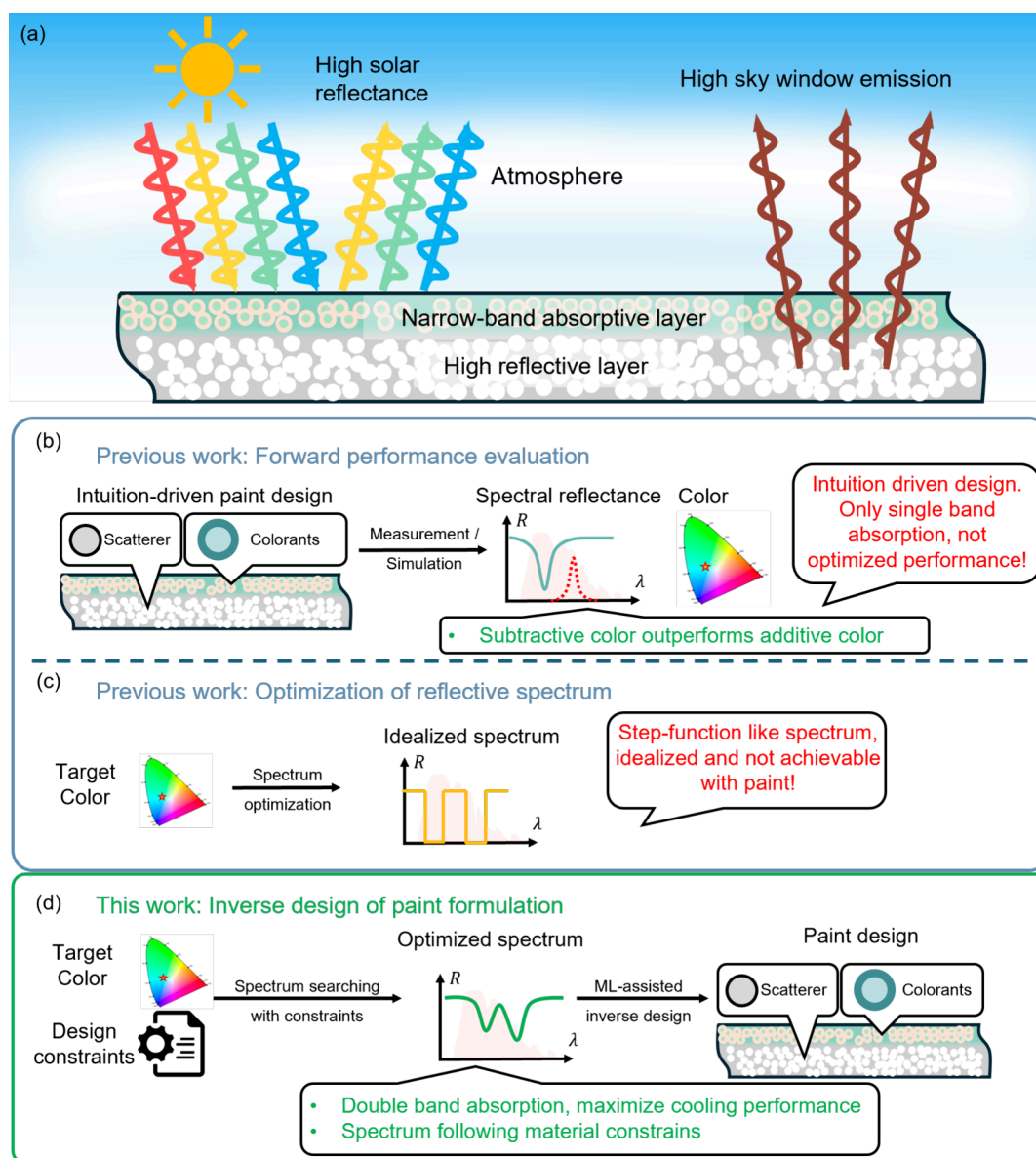


Figure 1. Overview of the inverse design of colored radiative cooling paint. (a) Principle of colored radiative cooling. (b) Forward experimental or theoretical performance evaluation of intuition-driven paint design. People noticed that subtractive color with narrow-band absorption would generate higher reflectance than additive color. However, the cooling performance is not optimized. (c) Theoretical optimization of the reflective spectrum. The optimization without considering materials' constraints would only yield an idealized spectrum but cannot be achieved. (d) Our inverse design framework bridges the gap between the spectrum optimization and the practical paint design.

based on an intuition-driven forward design process, where colorants with a single narrow absorption band in the visible spectrum are selected first, and the paints' color and cooling performance are subsequently evaluated. This approach is too expensive to yield optimal designs, particularly non-intuitive designs. Moreover, such trial-and-error methods are not suitable for practical applications, where customers request a target color and manufacturers develop formulations that meet both aesthetic and thermal performance requirements. On the other hand, some theoretical works consider metamerism, where different reflectance spectra can produce the same perceived color, resulting in a large and degenerate design space. While optimization of spectral optical reflectance under color constraints has been performed to maximize cooling power,^{35,48} these optical spectra are overly idealistic and cannot be achieved using realistic materials or paint systems. It

is evident that the inverse design of colored radiative cooling paints, in which a target color is specified and a realistic formulation is subsequently generated for optimal cooling performance, is highly desirable yet currently lacking.

In this work, we introduce a machine learning (ML)-enabled inverse design framework for colored radiative cooling paints. ML has recently demonstrated immense potential in accelerating the design and understanding of complex materials and physical systems.^{39,49–51} By integration of ML surrogate models with photonic domain knowledge, our framework effectively reduces the prohibitive computational costs and bridges the gap between optimized optical spectra and material realization. Subsequently, we develop a ML model that can accurately map these optimized spectra to realistic paint formulations. In contrast to conventional forward design methods, our approach enables the discovery of optimal and

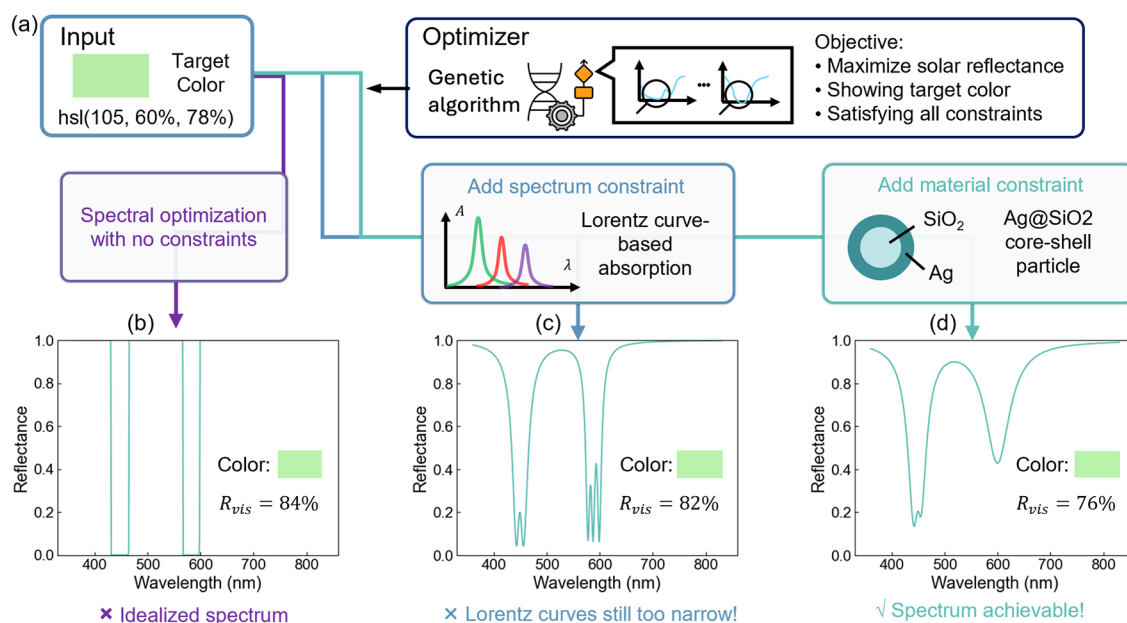


Figure 2. Optimizing spectral reflection with the given color. Panel a shows the workflow of the spectrum optimization. We used a generic algorithm to find the target spectrum that satisfies the constraints. Panel b shows the optimized spectrum with only the color constraint, which is idealized and cannot be achieved. After the addition of the Lorentzian curve constraint, the optimized spectrum becomes panel c. Finally, when the material constraints are added, the optimized spectrum is shown in panel d, which can be achieved with given materials.

often non-intuitive designs. Moreover, unlike prior efforts that yield only overly idealized optical spectra, our approach produces practical paint formulations with realistic optical spectra that are attainable using real materials. Notably, our framework has uncovered a non-intuitive double narrow-band absorption approach, which significantly reduces the heating power by up to 193 W/m² over the conventional additive color approach and 37 W/m² over the intuitive subtractive color strategy with single narrow-band absorption, respectively. We further conducted a comprehensive exploration of the entire hue–saturation–lightness (HSL) color space and provided a “coolability map”. We find that, while daytime subambient cooling is theoretically possible for 46% of colors using ideal optical spectra, this fraction drops to 24% when realistic materials and optical spectra are considered, establishing practical guidelines for colored radiative cooling paint development. Ultimately, our framework enables an inverse design of high-performance colored radiative cooling paints.

In this work, we are focusing on a double-layer paint where the bottom layer is a white paint layer that provides high solar reflectance and high sky window emission. The top thin layer contains dyes that provide solar absorption and produce color (Figure 1a). To produce a certain color, the most straightforward way is to reflect the irradiation at the corresponding wavelength based on the dispersion of visible light while absorbing the rest (Figure 1b). However, such an “additive color” would lead to a broadband absorption across the visible range and excessive solar heating. In contrast, the subtractive color approach is favorable for a thermal purpose, where the narrow-band absorption at a selective wavelength can reduce solar heating while maintaining the desired color (Figure 1c). In addition to the visible region, the paint should have high reflectivity in the ultraviolet (UV) and near-infrared (NIR) regions, which contribute to 5 and 43% of the AM1.5 global horizontal solar irradiance. Furthermore, strong emissivity in the mid-infrared (mid-IR) region is critical for

radiating absorbed heat through the atmospheric transparency window, thereby enabling passive cooling under sunlight.

Our ML-assisted inverse design framework, shown in Figure 1d, comprises two important steps. First, given the target color, we search for the reflectance spectra that maximize the cooling power. Physical constraints related to both the spectral response and material properties, which were not considered in previous studies, are introduced here to ensure that the generated spectra are physically achievable. This inverse design problem is formulated as an optimization task and is solved using evolutionary algorithms. Second, we translated the optimized spectra into realizable paint formulations. To deal with the challenges brought by the high dimensionality of the spectral data and the large design space of core–shell nanoparticles, we adopt a Lorentzian kernel decomposition strategy that isolates the relationship between individual particles and their corresponding Lorentzian components. This approach enables one-on-one mapping between particles and curves, substantially reducing the computational cost and simplifying the design process.

The design process began with the visible reflectance spectrum based on the target color, with the workflow shown in Figure 2a. The objective was to identify a spectrum that minimized solar absorption while maintaining the color difference within an acceptable tolerance. Given that multiple spectral profiles can satisfy a single color constraint, the optimization landscape is inherently non-convex. To robustly search for a global optimum and avoid local minima, we utilized a genetic algorithm (GA) that evaluates a diverse population of candidates with each optimization repeated across five different random initializations (details in the Method section of the Supporting Information). We pick a green color with HSL (hue, saturation, and lightness) values of 105, 60, and 78% for demonstration. In the conventional approach, no constraints were imposed on the spectral shape, allowing the optimization algorithm to explore the ideal

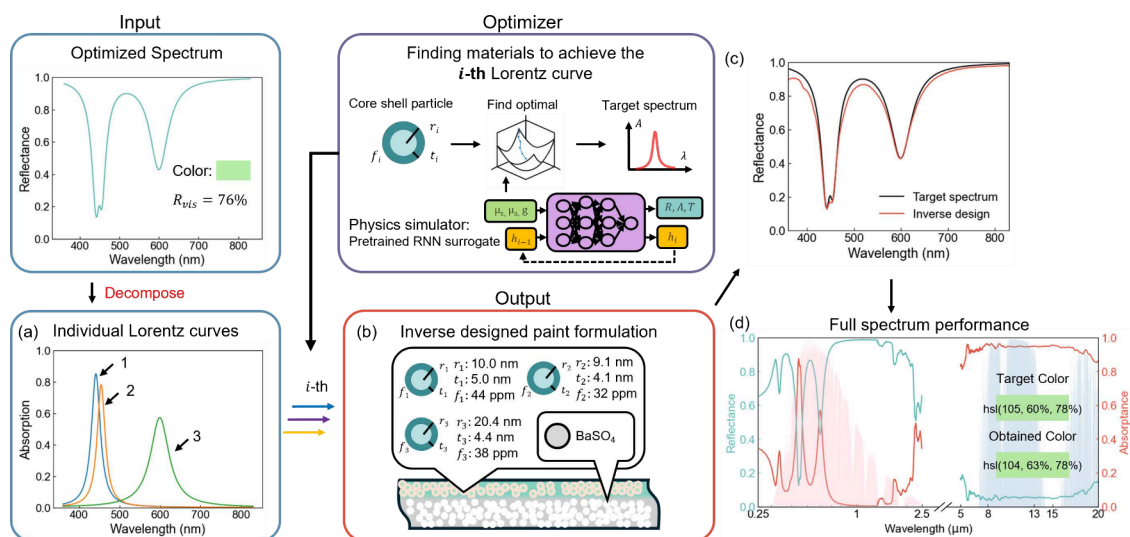


Figure 3. Designing the practical paint formulation from the optimized spectrum. The figure shows the workflow of the inverse design. (a) Lorentzian curves decomposed from the optimized spectrum. (b) Inverse designed paint formulation. Here, the concentration of core-shell nanoparticles is defined by their volume fraction within the paint matrix (ppm). (c) Comparison between the target spectrum and the spectrum of the obtained paint formulation. (d) Full reflectance spectrum of our designed paint formulation.

spectrum solutions (Figure 2a, purple lines). The resulting optimized spectrum is shown in Figure 2b, with a visible reflectance of 84%. Interestingly, instead of a single narrow-band absorption as commonly believed, the optimized spectrum shows two narrow absorption bands. The optimization algorithm avoided significant absorption near the peak irradiation to minimize the overall solar energy absorption. The optimized spectrum is mostly binary, with reflectance values of either 0 or 1, resulting in a step-like appearance.

However, such a spectrum is not feasible for real-world paint applications since the absorption spectra are usually smooth for real materials. Besides, perfect absorption and reflection are not achievable in practical systems. To develop a more realistic spectral design, we consider the mechanisms of color formation in paint. Common methods for creating color, such as the use of plasmonic particles,^{52,53} organic dyes,⁵⁴ and phosphor particles,³² rely on different mechanisms including plasmonic effects, molecular vibrations, and electronic transitions, respectively. Despite their differences, these methods share a fundamental characteristic: the resonant oscillation at specific frequencies with damping. For paints composed of multiple particles with different resonance frequencies, the total spectral absorption is the superposition of individual Lorentzian functions, which could be described by a Lorentzian kernel regression (LKR) model.

$$A(\lambda) = \sum_i h_i \frac{\gamma_i^2}{(\lambda - \lambda_i^*)^2 + \gamma_i^2} \quad (1)$$

We add the spectrum constraint by forcing the absorption spectrum to be described by the LKR model (Figure 2a, blue lines). We also restricted the maximum absorbance to account for the specular reflectance. The optimized spectral absorbance for the given color is shown in Figure 2c with a total visible reflectance of 82%. The spectral curve retains the double narrow-band absorption feature, with each dip now composed of several Lorentzian components. The visible reflectance was slightly reduced due to the addition of

constraints, but the spectrum became more physically realizable.

While the reflective spectrum described by the LKR model meets the physics principles of resonance, there will be a constraint on the choices of Lorentzian kernel functions related to the material choices. We demonstrate this by choosing Ag@SiO₂ core-shell particles as a filler for getting color. Due to the plasmonic effects, such core-shell particles provide selective narrow-band absorption, and the spectral absorption coefficients are Lorentzian curves.^{31,45,53} Also, the continuously tunable mapping between metallic shell thickness and localized surface plasmon resonance frequency provides a rich design space of the Lorentzian curve. However, it should be noted that the same inverse design framework is applicable to any alternative coloration approaches that offer tunable material parameters linked to the absorption profile and not just a limit to this material system. We precomputed the design space of Lorentzian kernel functions $\mathcal{D}$ based on a random sample of the particle parameter space and then used it as the material constraint on the Lorentzian kernel functions (Figure 2a, green lines, with details in the Method section of the Supporting Information). These narrow-band Lorentzian absorption peaks typically have a full width at half maximum (fwhm) ranging from 20 to 60 nm. The final optimized spectral reflectance is presented in Figure 2d. The visible reflectance is now 76%, further reduced compared to the unconstrained design, while the color difference remains within the acceptable tolerance. With the assurance that each Lorentzian kernel adheres to the constraints of the design space, the resulting spectrum minimizes solar heating while remaining achievable.

After the optimized spectrum was obtained, the next step was to translate the spectral design into a paint formulation. Color paint generally has two components: a colorant, which selectively absorbs light at specific wavelengths, and a scatterer, which reflects light across other solar wavelengths. The paint matrix and the scatterer can also provide high emissivity in the mid-IR range. There are mainly two configurations for the color paint.²⁹ One is to disperse the colorant within the scatterer matrix, creating a single-layer structure.⁵⁵ The second

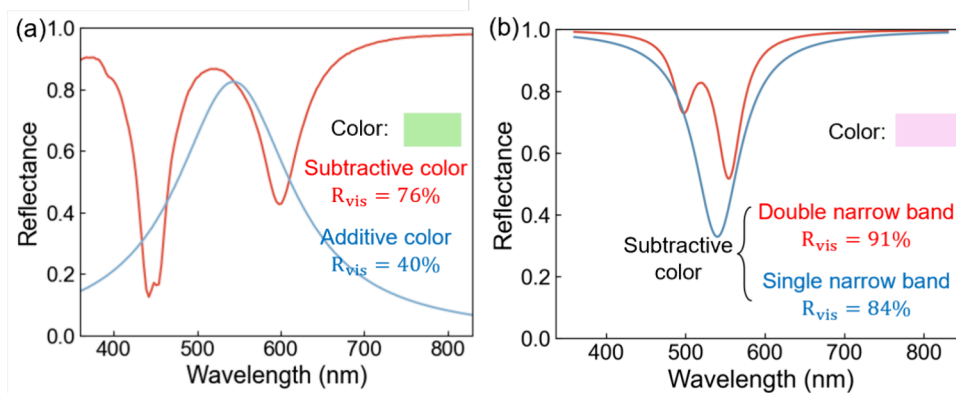


Figure 4. Comparison of the intuition-driven designed spectrum and optimized double-band absorption. (a) Additive color spectrum and optimized subtractive color spectrum. (b) Single narrow-band absorption and optimized double narrow-band absorption. Our double narrow-band absorption spectrum shows higher reflectance while keeping the same color in both cases.

approach employs a bilayer structure,^{30,31,53,56} where the colorant is in the upper layer, ensuring narrow-band absorption, while the lower layer consists of scatterers to maximize reflectivity and mid-IR emissivity. Based on previous studies^{29,48} and our own study (see [Supplementary Figure 2](#)), the bilayer structure would lead to better performance, with narrower absorption bands and less absorption in the NIR region. Accordingly, we adopted a bilayer design with a bottom layer composed of BaSO₄ acrylic, chosen for its high solar reflectance, and a top layer composed of multiple Ag@SiO₂ core-shell nanoparticles, which provide the desired color. The coating thickness plays an important role in the paint performance.^{57,58} For our benchmarks, we used a 400 μm bottom layer to ensure that the coating is “optically thick”,¹⁵ maximizing solar reflectance and decoupling the colorant optimization from substrate interference. The top layer was set to 10 μm to balance fabrication feasibility with the need to minimize solar absorption from the polymer matrix. While reducing the total thickness would decrease the net cooling power due to lower NIR reflectance, the inverse design framework remains effective at identifying the optimal formulation under any given thickness constraint.

The spectrum-to-formulation workflow is shown in [Figure 3](#). Instead of matching the whole spectrum at once, we first decompose the target spectrum into a set of individual Lorentzian curves, each representing an individual resonant absorption feature. This decomposition significantly reduces the design complexity by allowing each spectral component to be addressed independently, as opposed to mapping the entire spectrum to material parameters in one step.^{59,60} Subsequently, we developed a gradient descent (GD)-based optimization framework to minimize the difference between the target Lorentzian curve and the optical response of the core-shell structure. Photon Monte Carlo simulations⁶¹ were used to evaluate the particle’s optical response. Such physics simulation can be highly computationally expensive, and various of models have been developed to improve computational efficiency.^{49,61–64} Here, we integrated a fine-tuned recurrent neural network (RNN)-based surrogate model to replace the computationally expensive ray-tracing simulations. As detailed in a recent methodological study,⁶² this RNN architecture predicts the spectral response of multilayer radiative cooling paints with a mean absolute error (MAE) of less than 0.01 while providing a speedup of up to 2 orders of magnitude

compared to standard Monte Carlo simulations. Further details of the surrogate model implementation for this work are provided in the [Method](#) section of the Supporting Information. It should be noted that, while our modeling provides a robust physical basis, intrinsic discrepancies can exist between simulated and experimental spectra due to real-world factors, like particle aggregation and non-spherical geometries. Nonetheless, our framework remains a powerful tool for identifying optimal design strategies, and its flexible architecture allows for the integration of more complex scattering models or electromagnetic solvers as they become available to further improve prediction accuracy.

For the target spectrum obtained in the previous section, the decomposition yielded three Lorentzian curves ([Figure 3a](#)), each corresponding to a distinct absorption peak in [Figure 2d](#). The inverse-designed paint structure was shown in [Figure 3b](#), consisting of three different types of Ag@SiO₂ core-shell nanoparticles. The reconstructed spectrum closely approximates the target, as shown in [Figure 3c](#). Minor discrepancies arise due to the inherent ultraviolet absorption of silver and the omission of scattering effects in the decomposition model. Nonetheless, the reconstructed spectrum is sufficiently accurate to reproduce the desired optical properties. The full spectrum optical response was shown in [Figure 3d](#), with a high solar reflectance of 83% and a sky-window emissivity of 0.95. Additional absorption in the UV and NIR regions is due to Ag and the acrylic matrix. The resulting net cooling power for this green color paint is 22 W/m² under AM1.5 global horizontal solar irradiance, predicting the capability of the paint to achieve daytime below-ambient cooling for the given color.

Furthermore, to demonstrate the effectiveness of our inverse design, we chose two colors to compare our designed spectrum to other intuition-driven designs. In comparison to an additive color design, our inverse design improves the visible reflectance from 40 to 76%, reducing the heat load by 193 W/m², as shown in [Figure 4a](#). Even in comparison to another subtractive color with a single narrow-band absorption, our optimized design improves the reflectance from 84 to 91%, reducing the heat load by 37 W/m² ([Figure 4b](#)). These results demonstrate the advantage of double narrow-band absorption and our inverse design approach.

Beyond this specific example, our framework is capable of generating radiative cooling paint formulations across a wide range of colors. [Figure 5](#) presents the optimized solar spectra,

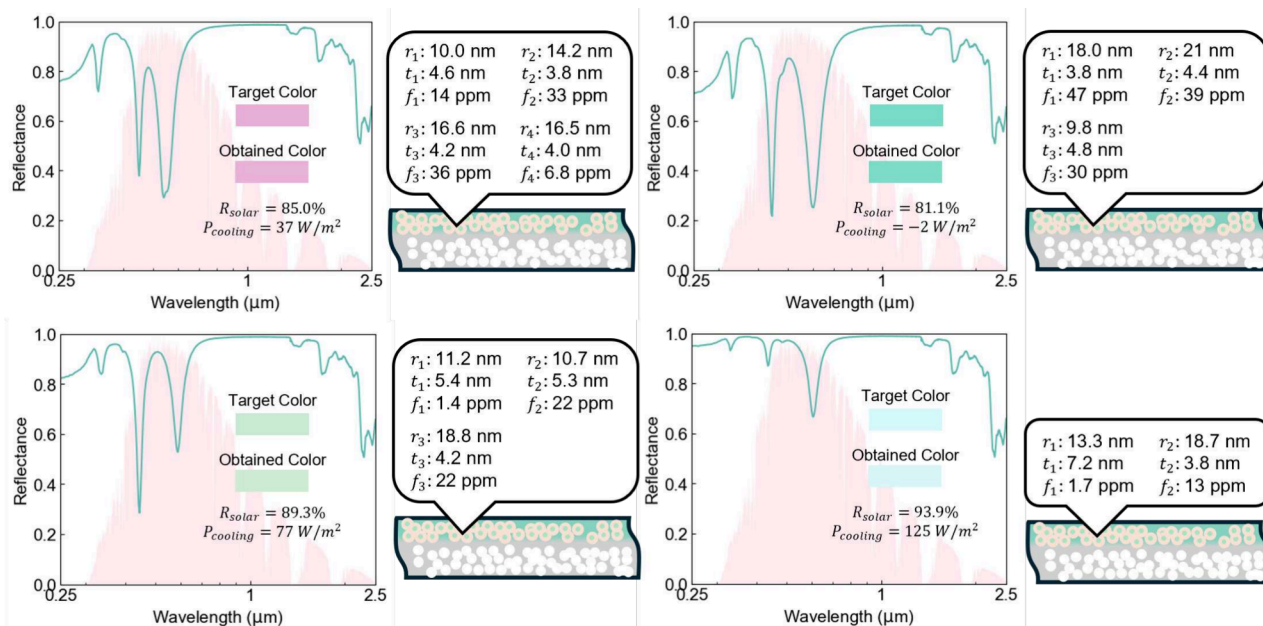


Figure 5. Paint formulation and the spectra of various colors. Comparison of target and obtained colors alongside their corresponding solar reflectance spectra and paint formulation.

the corresponding paint formulations, and the cooling power achieved for different colors. Note that, even for color with a negative cooling power, our approach can still give a corresponding paint formulation that minimizes solar absorption. Several additional designed spectra and the comparison to intuition-driven designs are provided in the [Supporting Information](#). The results illustrate the versatility and effectiveness of the proposed inverse design approach, which successfully balances color requirements with the functional demands of the radiative cooling.

After studying a few certain colors, we want to scan for multiple colors in the color space to build a “coolability map”. A total of 2000 colors were randomly sampled from the HSL color space, and the corresponding radiative cooling power under AM1.5 global horizontal solar irradiance was calculated for each point to provide a robust statistical representation of the achievable performance within the color space.

In the ideal case in which we assume perfect spectral reflectance and absorptance in the visible range and unity emissivity in the mid-IR, 46% of the sampled colors were found to show net cooling power over zero, as shown in [Figure 6a](#), implying potential for below-ambient cooling. This serves as an absolute theoretical bound in colored radiative cooling. However, when practical constraints were introduced, including limitations on the spectral shape, design constraints of Ag@SiO₂, additional absorptance in the UV and NIR regions, and non-ideal mid-IR emissivity, the fraction of colors capable of achieving radiative cooling was significantly reduced to 24% ([Figure 6b](#)). These results provide insights into the impact of material constraints on the viability of colored radiative cooling paints and guidelines for the design of colored radiative cooling paint. There is also potential to combine the inverse design framework with atomic or nanoscale simulations to further discover new materials or nanostructures with more ideal optical and thermal properties and expand the color bound.

Looking forward, there is significant potential to expand these “coolability” bounds by coupling our inverse design

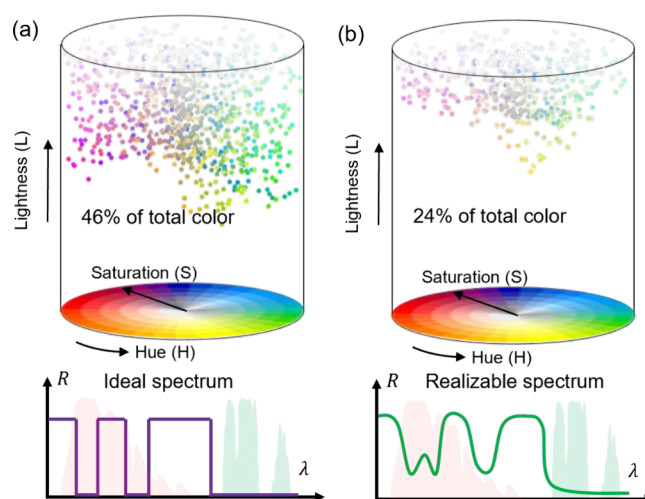


Figure 6. Analyzing daytime radiative cooling performance across the HSL color space. All the dots can have a positive net cooling power. (a) Ideal spectrum case. (b) Considered practical constraint cases.

framework with atomic and nanoscale simulations.^{65–68} Such a multiscale approach could accelerate the discovery of new materials or nanostructures with optimized properties, potentially pushing the performance of colored coatings closer to their theoretical ideals.

In conclusion, we present a ML-enabled inverse design framework of colored radiative cooling paints, and it identifies a realizable double narrow-band absorption strategy, which is non-trivial and significantly outperforms the conventional single narrow-band absorption strategy. By integration of optimization algorithms, physics-based simulations, and ML models, our approach outperforms the trial and error searching approach and bridges the critical gap between theoretical spectral design and practical paint formulation. With the rigorous accounting of both material and spectral constraints, our framework yields realizable reflective spectra that directly

guide experimental verification. These designed paints maximize the cooling performance while maintaining a similar color. Furthermore, by systematically exploring the entire HSL color space, we find that 24% of colors can achieve daytime subambient cooling when realistic material constraints are applied. This framework offers insights into the color paint design and would inspire further experimental confirmation.

■ ASSOCIATED CONTENT

Data Availability Statement

The original results of the study are available from the corresponding authors upon reasonable request.

■ Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.6c01941>.

Additional modeling and optimization methods (including details of the genetic algorithm for spectrum optimization, core-shell nanoparticle scattering calculations, and RNN surrogate model architecture) and supplementary figures (showing paint absorption analysis, comparison of single- and double-layer designs, comparison of machine learning inverse design and manual design, designed spectra and color coordinates for various hues, and influence of core-shell parameters on Lorentzian absorption curves) (PDF)

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Author Contributions

Z.G., D.F., G.L., and X.R. conceived the study. Z.G. and D.F. designed and implemented the models, analyzed the results, and wrote the manuscript. D.C. helped with the Monte Carlo simulation and RNN surrogate code. G.L. and X.R. supervised the project. All authors contributed to discussions and revisions of the manuscript.

Notes

The authors declare no competing financial interest.

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