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Spectral Control and Structural Design of PDMS Radiative Cooling Films Based on Transfer Matrix Method and Particle Swarm Optimization

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Abstract: *Facing the challenges of surging global energy consumption and the intensifying urban heat island effect, passive daytime radiative cooling technology has garnered significant attention as a zero-energy cooling method. Polydimethylsiloxane (PDMS) has become a research hotspot for low-cost cooling materials due to its excellent intrinsic emissivity in the atmospheric window band; however, its high transmittance in the visible light spectrum limits its daytime cooling efficiency. This paper aims to overcome the performance bottlenecks of PDMS-based radiative cooling materials through a combination of computational optics and intelligent optimization algorithms. First, a high-precision calculation model for the spectral response of thin films was constructed based on the Transfer Matrix Method and cubic spline interpolation, quantitatively revealing the non-linear modulation laws of film thickness on spectral emissivity. Second, a steady-state thermal balance simulation model incorporating solar radiation and the atmospheric thermal environment was established to evaluate the cooling potential of different structural designs. On this basis, a multilayer film structure combining a metal substrate with a Distributed Bragg Reflector (DBR) was proposed, and the Particle Swarm Optimization algorithm was utilized to globally optimize the layer thicknesses, achieving efficient reflection of the solar spectrum and enhancement of infrared radiation. Furthermore, based on the Maxwell-Garnett Effective Medium Theory, the optical scattering enhancement mechanism of nanoporous structures was explored. Simulation results indicate that the optimized nanoporous structure, without requiring complex coating processes, can achieve broadband high reflection through strong Mie scattering effects, significantly enhancing net cooling power, and demonstrating distinct advantages in cost and engineering feasibility assessments.*

Keywords: *Passive Radiative Cooling, Polydimethylsiloxane (PDMS), Transfer Matrix Method (TMM), Particle Swarm Optimization (PSO), Spectral Selectivity, Nanoporous Structure*

1. Introduction

With the acceleration of global industrialization and continuous population growth, energy crises and environmental climate issues are becoming increasingly severe. The large-scale use of traditional cooling equipment such as air conditioners not only consumes enormous amounts of electricity but also further exacerbates global warming and the urban heat island effect[1], [2] through the emission of greenhouse gases and waste heat. Against this background, passive radiative cooling technology, which utilizes the "atmospheric window"[3], [4] existing between the Earth's surface and deep space, has emerged. This technology requires no external energy input and can directly dissipate surface heat in the form of infrared radiation through the atmospheric window into cold outer space, thereby achieving a passive cooling effect where the surface temperature is lower than the ambient temperature. In particular, achieving radiative cooling below ambient temperature during the day has significant engineering application value for reducing building energy consumption, improving photovoltaic power generation efficiency, and alleviating thermal environmental pressure.

Among numerous candidates for radiative cooling materials, photonic crystals and metamaterials can achieve near-ideal spectral selectivity—high reflectivity in the solar radiation band and high emissivity in the atmospheric window band—through precise nanostructure design. However, their high manufacturing costs and complex micro-nano processing techniques greatly limit their large-scale application. In contrast, polymer-based materials show immense application potential due to their low cost, ease of processing, and good scalability. Among them, Polydimethylsiloxane (PDMS)[5] is regarded as an ideal intrinsic radiative cooling material due to the strong phonon absorption characteristics of its molecular bonds in the 8 to 13 micron band. Nevertheless, pure PDMS films exhibit high transparency in the visible light band, resulting in an inability to effectively block the thermal load of solar radiation on the substrate, and simply increasing thickness makes it difficult to achieve complete reflection of the solar spectrum and ultimate optimization of infrared emission.

Addressing the aforementioned issues, academia has conducted extensive research on material modification and structural design in recent years, attempting to improve the spectral characteristics of PDMS by doping nanoparticles or designing multilayer structures. However, traditional "trial-and-error" experimental research is often time-consuming and laborious, and it is difficult to traverse the massive structural parameter space to find the global optimum. With the development of computational science, material inverse design methods based on numerical simulation and intelligent algorithms provide new ideas for solving this problem. By establishing precise optical transmission models and thermodynamic models, combined with evolutionary algorithms for parameter optimization, the research and development cycle can be significantly shortened, and novel high-efficiency structures that are difficult to discover through traditional experience can be excavated.

This paper focuses on the computer-aided design and performance optimization of PDMS-based radiative cooling materials. First, using the Transfer Matrix Method to solve Maxwell's equations[6], [7], a numerical model for optical transmission suitable for layered media was constructed, and combined with the steady-state thermal balance equation, a complete cooling performance evaluation framework was formed. On this basis, the Particle Swarm Optimization algorithm was introduced to perform multi-parameter global optimization for a multilayer structure of "Metal Substrate + Dielectric Interference Layer + PDMS," achieving directional control of spectral characteristics. Furthermore, to overcome the defect of complex processes in multilayer films, this paper further introduced Effective Medium Theory to simulate the Mie scattering effect of nanoporous structures and conducted a comprehensive comparative analysis of different design schemes from dimensions such as cooling performance, manufacturing cost, and engineering feasibility. This study aims to provide scientific basis and technical support for the development of high-performance, low-cost, and scalable radiative cooling materials through algorithm-driven structural design.

2. Methods

This study aims to design and optimize an efficient radiative cooling film structure. To achieve precise calculation and performance evaluation of the spectral characteristics of Polydimethylsiloxane (PDMS) and its composite structures, this paper constructs a comprehensive computational framework integrating optical transmission simulation, thermodynamic balance calculation, and global optimization algorithms. The framework primarily includes a multilayer film optical model based on the Transfer Matrix Method, a steady-state radiative cooling thermal balance model, an effective medium model based on Maxwell-Garnett theory, and a Particle Swarm Optimization algorithm for structural parameter inversion.

2.1 Optical Property Calculation of Thin Films Based on Transfer Matrix Method

Since the thickness of the PDMS film and the subsequently designed multilayer structure is in the micrometer range, comparable to the wavelength of infrared radiation, significant interference and diffraction effects occur during the propagation of light waves inside the film. Traditional geometric optics models are difficult to accurately describe their spectral response; therefore, this study adopts the Transfer Matrix Method (TMM) to solve the boundary value problem of Maxwell's equations in layered media to quantitatively characterize the spectral emissivity and transmittance of the films.

For a layered structure composed of N layers of isotropic homogeneous media, assuming light waves are incident at a vertical angle. According to the boundary continuity conditions of the electromagnetic field, the electric field E and magnetic field H at the input and output interfaces of the j-th layer film satisfy a linear transformation relationship, and its characteristic matrix M_j can be expressed as:

$$M_j = \begin{bmatrix} \cos\delta_j & -\frac{i}{\eta_j}\sin\delta_j \\ -i\eta_j\sin\delta_j & \cos\delta_j \end{bmatrix} \tag{1}$$

Where δ_j is the phase thickness of the j-th layer film, characterizing the phase change produced by the light wave passing through that layer; η_j is the optical admittance of that layer. Their calculation formulas are respectively:

$$\delta_j = \frac{2\pi}{\lambda} N_j(\lambda) d_j \tag{2}$$

$$\eta_j = N_j(\lambda) \tag{3}$$

Where λ is the incident light wavelength, d_j is the physical thickness of the j-th layer, and $N_j(\lambda) = n_j(\lambda) + i\kappa_j(\lambda)$ is the complex refractive index of the material.

The total transmission matrix M of the entire multilayer film system is obtained by the ordered product of the characteristic matrices of each sub-layer, i.e., $M = \prod_{j=1}^{N} M_j = \begin{bmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{bmatrix}$. Based on the matrix elements of the entire system, the complex amplitude reflection coefficient $r(\lambda)$ and transmission coefficient $t(\lambda)$ of the system can be derived:

$$r(\lambda) = \frac{\eta_0 m_{11} + \eta_0 \eta_{sub} m_{12} - m_{21} - \eta_{sub} m_{22}}{\eta_0 m_{11} + \eta_0 \eta_{sub} m_{12} + m_{21} + \eta_{sub} m_{22}} \tag{4}$$

$$t(\lambda) = \frac{2\eta_0}{\eta_0 m_{11} + \eta_0 \eta_{sub} m_{12} + m_{21} + \eta_{sub} m_{22}} \tag{5}$$

Where η_0 and η_{sub} are the optical admittances of the incident medium (air) and the substrate, respectively. Furthermore, the spectral energy reflectance $R(\lambda) = |r(\lambda)|^2$ and transmittance $T(\lambda) = \frac{\mathrm{Re}(\eta_{sub})}{\mathrm{Re}(\eta_0)} |t(\lambda)|^2$ can be obtained. According to Kirchhoff's law of thermal radiation, under thermal equilibrium, the spectral emissivity $\varepsilon(\lambda)$ of the film equals its spectral absorptivity $\alpha(\lambda)$, i.e.:

$$\varepsilon(\lambda) = \alpha(\lambda) = 1 - R(\lambda) - T(\lambda) \tag{6}$$

This calculation model provides core algorithmic support for the subsequent evaluation of the reflection performance of materials in the visible light band and emission performance in the atmospheric window band (8-13 μm).

2.2 Radiative Cooling Steady-State Thermal Balance Simulation

To evaluate the cooling efficiency of the film under actual working conditions, this study establishes a comprehensive thermal balance calculation model including solar radiation, atmospheric thermal radiation, surface radiation, and non-radiative heat exchange. The model is based on the law of conservation of energy, treating the film as a node in dynamic heat exchange. The net cooling power per unit area $P_{net}(T)$ is defined as the difference between the energy radiated outward by the film and the energy absorbed from the outside:

$$P_{net}(T) = P_{rad}(T) - P_{atm}(T_{air}) - P_{sun} - P_g(T_{air}) - P_{non}(T, T_{air}) \tag{7}$$

Where T is the film surface temperature and T_{air} is the ambient temperature. The terms on the right side of the equation represent the film's own radiation power P_{rad}, the absorbed atmospheric thermal radiation power P_{atm}, the absorbed solar radiation power P_{sun}, the absorbed ground radiation power P_g, and the non-radiative heat exchange power P_{non}, respectively.

Specifically, the film's own radiation power is determined by Planck's law of blackbody radiation and spectral emissivity:

$$P_{rad}(T) = \int_0^{\infty} P_B(\lambda, T) \varepsilon(\lambda, T) d\lambda \tag{8}$$

Where $P_B(\lambda, T) = \frac{2hc^2}{\lambda^5} \frac{1}{e^{hc/(\lambda k_B T)} - 1}$ is the blackbody spectral radiance. The absorbed solar radiation power P_{sun} depends on the AM1.5G standard solar spectral irradiance $I_{AM1.5}(\lambda)$ and the film's spectral absorptivity:

$$P_{sun} = \int_0^{\infty} I_{AM1.5}(\lambda) \varepsilon(\lambda) d\lambda \tag{9}$$

The atmospheric thermal radiation absorption power P_{atm} involves atmospheric transmittance $t_{atm}(\lambda)$, calculated as:

$$P_{atm}(T_{air}) = \int_0^{\infty} P_B(\lambda, T_{air}) [1 - t_{atm}(\lambda)] \varepsilon(\lambda) d\lambda \tag{10}$$

In addition, considering air convection and heat conduction in the actual environment, the non-radiative heat exchange power can be described by the combined heat transfer coefficient U: $P_{non} = U(T_{air} - T)$. By solving for the temperature T when $P_{net}(T) = 0$, the steady-state equilibrium temperature T_{eq} of the system can be obtained, thereby quantifying the ultimate cooling capacity of the film.

2.3 Global Optimization of Multilayer Structure Based on Particle Swarm Optimization

To maximize infrared emission while suppressing solar absorption, this study proposes a multilayer film structure of "Metal Substrate + Dielectric Interference Layer + PDMS Functional Layer." This is a typical high-dimensional non-linear optimization problem, where the goal is to find a set of optimal layer thickness combinations that maximize the net cooling power. For this purpose, the Particle Swarm Optimization (PSO) algorithm[8], [9] is introduced for global optimization.

Define the decision variable vector D as the thickness of each dielectric layer and the top PDMS layer:

$$D = [d_1, d_2, \dots, d_N, d_{PDMS}]^T \tag{11}$$

Establish the objective function $f(D)$, aiming to maximize the net cooling power per unit area:

$$\max f(D) = P_{rad}(D) - P_{atm}(D) - P_{sun}(D) \tag{12}$$

During the optimization process, the algorithm initializes a swarm of random particles, each representing a potential thickness combination in the solution space. The velocity v_{id} and position x_{id} of the particles are updated iteratively based on the individual historical best solution p_{id} and the global historical best solution p_{gd}, with the update formulas as follows:

$$v_{id}^{k+1} = w v_{id}^{k} + c_1 r_1 \left(p_{id}^{k} - x_{id}^{k}\right) + c_2 r_2 \left(p_{gd}^{k} - x_{id}^{k}\right) \tag{13}$$

$$x_{id}^{k+1} = x_{id}^{k} + v_{id}^{k+1} \tag{14}$$

Where w is the inertia weight, c_1, c_2 are learning factors, and r_1, r_2 are random numbers. To ensure process feasibility, boundary constraints are applied to the thickness of each layer. Through iterative search, the PSO algorithm can effectively escape local extrema and converge to the global optimal thickness configuration.

2.4 Microstructure Optical Modeling Based on Effective Medium Theory

For the nanoporous scattering structure and doping modification scheme, the material is no longer a homogeneous medium. This study adopts the Maxwell-Garnett Effective Medium Theory (EMT)[10], [11] to calculate the effective optical constants of the composite material. This theory is applicable where the characteristic size of the dispersed phase is much smaller than the light wavelength and is discretely distributed.

Let the complex permittivity of the host material (PDMS) be ε_{host}, the complex permittivity of the inclusion phase (such as air pores or SiO_2 microspheres) be εinc, and the volume fraction of the inclusion phase be V. The effective permittivity ε_{eff} of the mixed medium satisfies the following equation:

$$\frac{\varepsilon_{eff} - \varepsilon_{host}}{\varepsilon_{eff} + 2\varepsilon_{host}} = V \frac{\varepsilon_{inc} - \varepsilon_{host}}{\varepsilon_{inc} + 2\varepsilon_{host}} \tag{15}$$

Rearranging gives the explicit expression for the effective permittivity:

$$\varepsilon_{eff}(\lambda, V) = \varepsilon_{host}(\lambda) \frac{2\varepsilon_{host}(\lambda) + \varepsilon_{inc}(\lambda) + 2V[\varepsilon_{inc}(\lambda) - \varepsilon_{host}(\lambda)]}{2\varepsilon_{host}(\lambda) + \varepsilon_{inc}(\lambda) - V[\varepsilon_{inc}(\lambda) - \varepsilon_{host}(\lambda)]} \tag{16}$$

Furthermore, the effective complex refractive index of the composite material $N_{eff}(\lambda, V) = \sqrt{\varepsilon_{eff}(\lambda, V)}$ can be derived. The obtained effective complex refractive index is used as an input parameter into the transfer matrix model described in Section 2.1, thereby achieving rapid and accurate prediction of the spectral characteristics of micro-nano structured composite films.

3. Experiments and Results

This chapter mainly presents the simulation experiment results based on the aforementioned calculation models and optimization algorithms. First, the optical constants and environmental spectral data used for simulation are described, followed by a detailed analysis of the spectral response characteristics and baseline cooling performance of PDMS films. On this basis, the design results of the multilayer film structure based on Particle Swarm Optimization (PSO) are further elaborated, and the comprehensive performance of the improved scheme incorporating nanoporous structures is comparatively analyzed in terms of performance and engineering feasibility.

3.1 Data Description and Preprocessing

The accuracy of the simulation experiments relies heavily on the precision of material optical constants and environmental spectral data. This study constructed a comprehensive database containing matrix materials, functional media, and environmental parameters.

Material Optical Constants: The complex refractive index of PDMS $N(\lambda) = n(\lambda) + i\kappa(\lambda)$ covers the 0.4-20 μm band, capable of accurately characterizing its transparency in the visible region and phonon absorption features within the 8-13 μm atmospheric window. The optical constants for metal substrates (Ag, Al) and dielectric layers (SiO, TiO, Al_2O_3, HfO_2) are all taken from authoritative literature.

Environmental Spectral Data: Solar radiation adopts the AM1.5G global tilt spectrum under the ASTM G173-03 standard, with total irradiance normalized to 1000 W/m^2. The atmospheric thermal radiation background is based on the US Standard Atmosphere 1976 model, focusing on the characteristic absorption of water vapor, CO_2, and O_3 in the infrared band.

Data Cleaning and Interpolation: Given the non-uniformity of wavelength sampling in the original experimental data, this study employs Cubic Spline Interpolation to resample all optical constants, mapping them uniformly to a computational grid of [0.3, 25] μm with a step size of 0.01 μm, to meet the high-resolution spectral calculation requirements of the Transfer Matrix Method and ensure the stability of numerical simulation.

3.2 Results Analysis

3.2.1 Numerical Simulation of PDMS Film Spectral Characteristics

Using the established thin-film optical transmission model, we first calculated the spectral emissivity $\varepsilon(\lambda, d)$ of pure PDMS films under different thicknesses (15-200 μm). The simulation results shown in Figure 1 indicate that the spectral response of PDMS exhibits significant wavelength selectivity and thickness dependence.

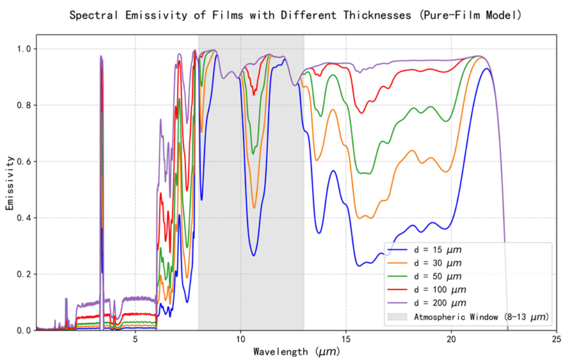

Fig. 1 Spectral Emissivity Variation Curves with Wavelength at Different Thicknesses

Table 1 Average Emissivity within the Atmospheric Window

Film Thickness (μm)	Average Emissivity	Increase Relative to Baseline
15	75.82%	--
30	85.16%	12.32%
50	89.93%	18.61%
100	93.48%	23.29%
200	94.49%	24.62%

In the visible light band (0.3-0.7 μm), due to the extremely low extinction coefficient κ (order of magnitude 10^{-6}), the emissivity of films of all thicknesses approaches 0, demonstrating high transparency. In the 8-13 μm atmospheric window band, emissivity shows a non-linear growth trend with increasing thickness. As shown in Table 1, when the thickness increases from 15 μm to 100 μm, the average emissivity in the atmospheric window significantly increases from 75.82% to 93.48%; however, further increasing the thickness to 200 μm results in an increase of only 1.33%, indicating that radiative performance has tended towards saturation. This calculation result reveals the limitations of simply

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increasing thickness in terms of cost-effectiveness and points out that pure PDMS films still have weak transmission in the infrared band and cannot fully achieve ideal blackbody radiation.

3.2.2 Baseline Cooling Performance Evaluation

Based on the steady-state thermal balance equation, the cooling power of suspended PDMS films under standard environment ($T_{air} = 300K, U = 10W/(m^2K)$ was numerically solved. The calculation results in Figure 2 show that with increasing thickness, the net cooling power exhibits a saturation growth trend similar to emissivity. At a thickness of 200 μm, the maximum net cooling power achievable by single-layer PDMS is 50.94 W/m ^{2}, and the equilibrium temperature is about 4 °C lower than the ambient temperature.

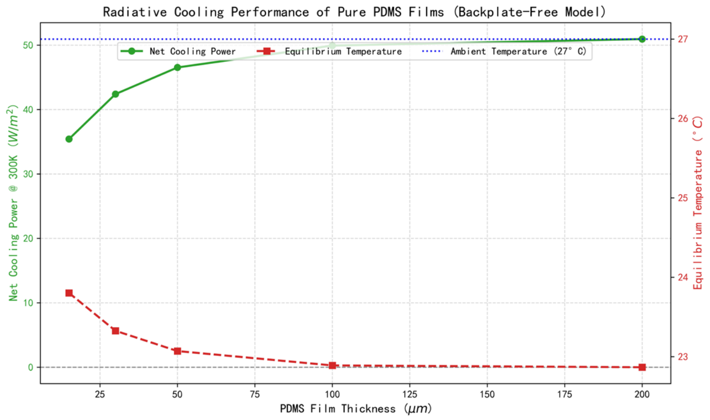

Fig.2 Radiative Cooling Performance of Pure Films

Although PDMS possesses cooling capabilities, thermal balance analysis points out that its performance is limited by two physical mechanisms: first, its high transmittance in visible light leads to an inability to block the thermal load of solar radiation on the substrate; second, a single material is difficult to achieve perfect reflection of the solar spectrum across the entire band. This confirms the necessity of introducing spectral control structures.

3.2.3 Multilayer Structure Optimization Design Based on PSO

To break through the performance bottleneck of single-layer materials, this study applied the Particle Swarm Optimization (PSO) algorithm to globally optimize the multilayer structure. The algorithm set a population size of 4 and a maximum number of iterations of 80.

The iteration convergence curve shown in Figure 3 displays that the objective function value tends to stabilize after about 40 iterations, indicating that the global optimal solution has been searched. The optimized best structural parameters are: SiO_2 and TiO_2 alternating layer thicknesses vary between 30-800 nm, and the top PDMS thickness is 100 μm.

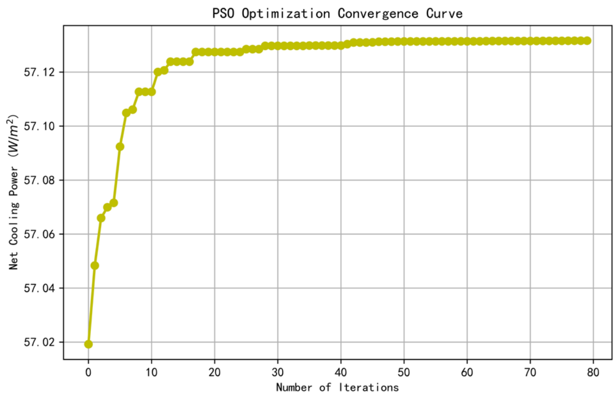

Fig.3 PSO Optimization Convergence Curve

As shown in Figure 4, the spectral characteristics of the optimized structure exhibit ideal "selective reflection-emission" features: in the 0.3-2.5 μm solar radiation band, thanks to the interference effect of the Distributed Bragg Reflector (DBR), spectral absorptivity is suppressed below 0.15 (i.e., reflectivity > 0.85); in the 8-13 μm band, utilizing the phonon resonance of PDMS and constructive interference of the dielectric layers, the average emissivity is increased to over 0.95. Ultimately, the net cooling power of this structure reaches 57.11 $\mathrm{W/m^2}$, an increase of about 12% compared to pure PDMS films.

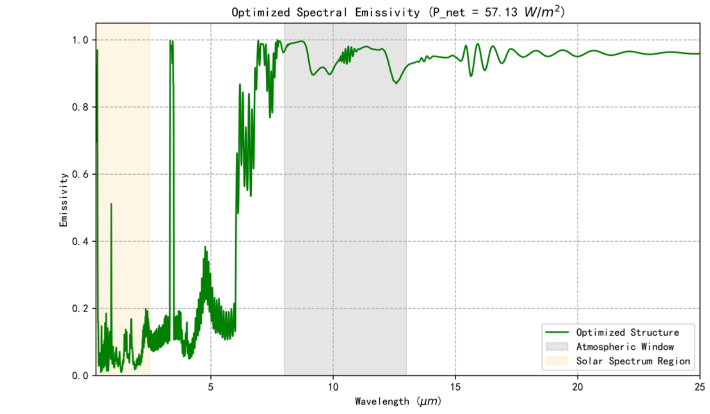

Fig.4 Optimized Spectral Emissivity

3.2.4 Nanostructure Modification and Comprehensive Performance Comparison

Addressing the problems of complex processes and high costs of multilayer film structures, the study further introduced Maxwell-Garnett Effective Medium Theory to simulate the optical behavior of introducing nanopores into the PDMS matrix (Scheme B) and compared it with the multilayer film improvement scheme (Scheme A).

Calculation results show that Scheme B (Al substrate + Nanoporous PDMS) utilizes the strong Mie scattering induced by the refractive index difference between the pores and the matrix to achieve broadband high reflection (reflectivity > 0.9) and enhanced infrared emission without the need for expensive DBR stacks. Its net cooling power reaches the highest in the field at 60.71 $\mathrm{W/m^2}$.

To evaluate engineering feasibility, we constructed a "Cost-Performance" two-dimensional evaluation matrix as shown in Figure 5.

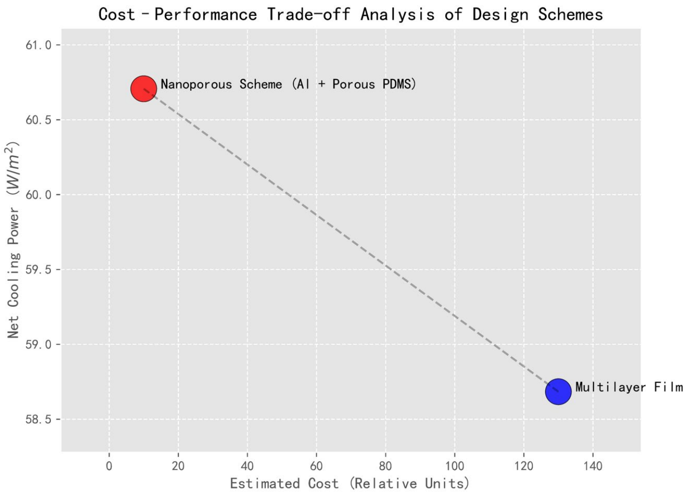

Fig.5 Cost-Performance Trade-off Analysis of Design Schemes

The results clearly show that Scheme B is located at a better position on the Pareto frontier: compared to the multilayer film scheme, while improving performance by 3.5%, the estimated manufacturing cost is reduced by about 92.3%. The radar chart analysis shown in Figure 6 indicates that Scheme B is significantly superior to Scheme A in dimensions of processing simplicity, lightweighting, and cost advantage. Although slightly inferior in weather resistance, overall, the nanoporous structure is the best design scheme combining high-performance computational indicators with potential for engineering implementation.

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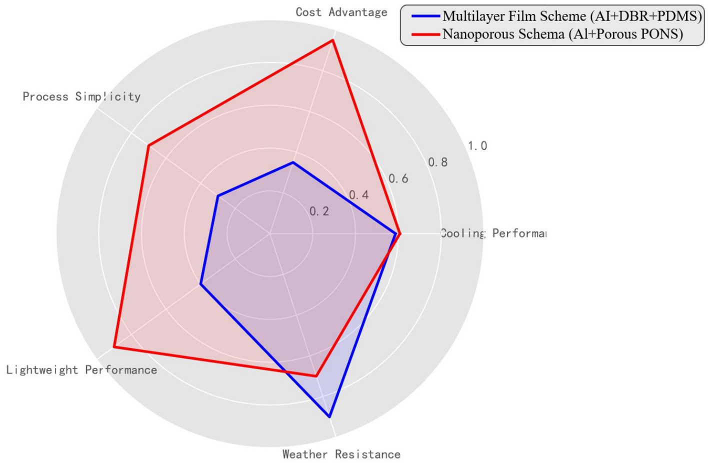

Fig.6 Comprehensive Feasibility valuation of Radiative Cooling Designs

4. Conclusion

This paper addressed the issues of insufficient spectral selectivity and limited cooling efficiency of Polydimethylsiloxane (PDMS) radiative cooling films by constructing a comprehensive computational framework integrating optical simulation, thermodynamic simulation, and intelligent optimization. The mechanisms of film thickness, multilayer interference structures, and micro-nano scattering units on regulating radiative cooling performance were systematically studied.

The research first established a high-precision optical model based on the Transfer Matrix Method, quantitatively elucidating the spectral response laws of pure PDMS films. Simulation results reveal that although increasing film thickness can improve the average emissivity within the atmospheric window by enhancing optical path absorption, this gain effect presents a significant marginal diminishing characteristic with increasing thickness. More critically, the high transmission characteristic of pure PDMS material in the visible light band makes it unable to independently shield the solar radiation thermal load, leading to a theoretical upper limit on the net cooling power of single-layer structures, which is difficult to meet the needs of efficient daytime cooling.

To break through this performance bottleneck, this paper proposed and optimized a multilayer film structure design scheme based on spectral control. By introducing the Particle Swarm Optimization algorithm, global optimization of dielectric layer thicknesses was achieved. Optimization results show that the alternating film structure constructed using the Distributed Bragg Reflector principle can utilize light interference effects to form a high reflection band in the visible light range, while cooperating with the PDMS top layer to achieve efficient emission in the infrared band. This algorithm-driven structural design significantly suppressed solar radiation absorption, resulting in a marked increase in net cooling power compared to pure films, verifying the effectiveness of multilayer interference structures in precise spectral control.

Furthermore, addressing the cost and process challenges faced by multilayer film structures in engineering applications, this paper explored a modification scheme based on Effective Medium Theory using nanoporous structures. The study found that introducing nanopores into the PDMS matrix can induce strong Mie scattering effects, which not only significantly reduces the material's effective refractive index to match impedance but also significantly enhances the reflection capability of sunlight and the absorption path of infrared light through multiple scattering. Comprehensive comparative analysis shows that this scheme, while achieving the highest net cooling power in the entire field, possesses overwhelming advantages in manufacturing cost, lightweighting, and process simplicity, locating it on the Pareto frontier of the cost-performance evaluation system.

In summary, this paper not only proved the significant role of structural optimization in improving radiative cooling performance through numerical calculation but also demonstrated the core value of computer algorithms in material inverse design. The nanoporous PDMS structure scheme derived from the research combines high performance with low cost advantages, offering a highly potential solution for passive radiative cooling technology to move from the laboratory to large-scale engineering applications.

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