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Passive Radiative Cooling Film Design via Transfer Matrix Modeling and Genetic Algorithm Optimization

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ABSTRACT

This paper proposes a unified modeling approach integrating optical modeling, thermal radiation calculations, and energy balance analysis for the passive radiative cooling performance of polydimethylsiloxane (PDMS) films. Based on film interference theory and the transfer matrix method, a quantitative relationship between film thickness and spectral emission characteristics is established, enabling systematic evaluation of emission performance under thermal equilibrium conditions. Model results reveal the critical influence of thickness modulation on mid-infrared radiation capability, providing explicit physical guidance for structural design. Building upon this, a genetic algorithm is introduced to perform global joint optimization of material combinations and layer thickness parameters in multilayer film structures, enabling synergistic control over solar reflection and atmospheric window radiation. Optimization results demonstrate that this approach significantly enhances net cooling performance at reduced structural thickness, exhibiting excellent versatility and scalability. It provides a unified modeling and optimization framework for designing radiative cooling films and related photothermal control structures.

KEYWORDS

Transfer Matrix Method; Multilayer Thin-Film Optimization Design; Genetic Algorithm

1. INTRODUCTION

With increasing energy consumption and worsening thermal environmental issues, achieving effective cooling without additional energy expenditure has become a critical research focus in materials and thermal management. Passive radiative cooling regulates a material's spectral response in the solar and mid-infrared bands to directly release heat as radiation into outer space, yet its performance heavily depends on material structure and parameter configuration [1].

Existing studies predominantly employ empirical design or local parameter scanning methods, making it challenging to systematically characterize the coupling relationships between film thickness, interference effects, and radiative behavior. This complexity is particularly pronounced in multilayer structures, where discrete material selection coexists with continuous thickness parameters, significantly complicating structural optimization [2]. To address this issue, this paper proposes a modeling and optimization framework for passive radiative cooling centered on spectral regulation. This method integrates thin-film interference theory with the transfer matrix approach to uniformly model the optical behavior of both single-layer and multilayer films. Coupled with thermal radiation and energy balance models, it enables quantitative evaluation of net cooling performance. Building upon this foundation, a genetic algorithm is employed for global joint optimization of material sequences and layer thickness parameters, enhancing the systematicity and efficiency of structural design [3]. Using PDMS-based thin-film structures as an example, the impact of thickness modulation and multi-layer co-design on cooling performance is analyzed. Experimental results demonstrate that this method can effectively suppress solar radiation and significantly enhance mid-infrared radiation capabilities at relatively thin structural thicknesses, thereby achieving stable and predictable net cooling performance.

2. MODEL CONSTRUCTION AND THICKNESS EFFECT ANALYSIS OF SPECTRAL EMISSIVITY FOR PDMS FILMS

2.1. Optical Property Description and Modeling Assumptions for Air-PDMS-Air System

To investigate the variation law of emissivity with wavelength for PDMS films under different thicknesses, an "Air-PDMS-Air" model is established.

Under normal incidence conditions, the reflection and transmission behaviors of s and p polarization components are identical. Additionally, PDMS is an absorptive medium, and its optical property is expressed by the complex refractive index:

$$\tilde{n}_1(\lambda) = n_1(\lambda) + ik_1(\lambda) \tag{1}$$

Here, $n_1(\lambda)$ denotes the refractive index, $k_1(\lambda)$ denotes the extinction coefficient, and λ denotes the wavelength in vacuum.

Air is a non-absorptive medium in the considered wavelength range, and its complex refractive index is expressed as $\tilde{n}_0 = \tilde{n}_2 = 1 + i0$.

Starting from the law of energy conservation, for a given wavelength λ, the energy incident on the film is divided into three parts in the entire "Air-PDMS-Air" system:

Reflection that returns to the incident side after multiple reflections at the upper surface and inside the film, denoted as $R(\lambda)$; Transmission that passes through the film and finally exits from the lower surface into the air below, denoted as $T(\lambda)$; Absorption that is absorbed by the medium inside the PDMS film, denoted as $A(\lambda)$.

Under thermal equilibrium conditions, according to Kirchhoff's law, the emissivity of the surface at each wavelength and direction is equal to the corresponding absorptivity. Therefore, $\varepsilon(\lambda) = A(\lambda)$.

Furthermore, bulk scattering is not considered inside the PDMS film, and only refraction and absorption are taken into account, meaning that electromagnetic waves decay exponentially in the medium. Meanwhile, the incident light is unpolarized, but under normal incidence, the optical properties of s and p polarizations are identical. Thus, the result of any single polarization can be calculated to represent the overall outcome [4].

For a given wavelength λ, the propagation constant in PDMS is:

$$\tilde{\beta}_1(\lambda) = \frac{2\pi}{\lambda}\tilde{n}_1(\lambda) = \frac{2\pi}{\lambda}\left[n_1(\lambda) + ik_1(\lambda)\right] \tag{2}$$

When the thickness of the PDMS film is d, the phase thickness of the electromagnetic wave propagating along the normal direction (i.e., under normal incidence) inside the film is:

$$\phi(\lambda, d) = \tilde{\beta}_1(\lambda)d = \frac{2\pi}{\lambda}\tilde{n}_1(\lambda)d \tag{3}$$

2.2. Emissivity Calculation Method and Solution Based on Thin Film Interference Theory

(1) Fresnel Coefficients and Multi-Beam Interference Model

Under normal incidence conditions, in the "Air-PDMS-Air" structure, the interface Fresnel coefficients are:

$$r_{01}(\lambda) = \frac{\tilde{n}_0(\lambda) - \tilde{n}_1(\lambda)}{\tilde{n}_0(\lambda) + \tilde{n}_1(\lambda)}, t_{01}(\lambda) = \frac{2\tilde{n}_0(\lambda)}{\tilde{n}_0(\lambda) + \tilde{n}_1(\lambda)}, r_{12}(\lambda) = \frac{\tilde{n}_1(\lambda) - \tilde{n}_2(\lambda)}{\tilde{n}_1(\lambda) + \tilde{n}_2(\lambda)}, t_{12}(\lambda) = \frac{2\tilde{n}_1(\lambda)}{\tilde{n}_1(\lambda) + \tilde{n}_2(\lambda)} \tag{4}$$

Since the air layers on both sides have the same optical properties, $\tilde{n}_0 = \tilde{n}_2 = 1$.

When the incident wave enters the PDMS film, a multi-beam interference phenomenon is formed. By superposing all reflected and transmitted beams, the equivalent reflection coefficient and equivalent transmission coefficient of the three-layer "Air-PDMS-Air" structure can be obtained as:

$$r(\lambda, d) = \frac{r_{01}(\lambda) + r_{12}(\lambda)e^{2i\phi(\lambda,d)}}{1 + r_{01}(\lambda)r_{12}(\lambda)e^{2i\phi(\lambda,d)}}, t(\lambda, d) = \frac{t_{01}(\lambda)t_{12}(\lambda)e^{i\phi(\lambda,d)}}{1 + r_{01}(\lambda)r_{12}(\lambda)e^{2i\phi(\lambda,d)}} \tag{5}$$

(2) Calculation of Reflectivity, Transmissivity and Emissivity

Under normal incidence with air on both sides, the reflectivity and transmissivity can be expressed by the squared modulus of the equivalent reflection coefficient and transmission coefficient:

$$= R(\lambda, d) = |r(\lambda, d)|^2, T(\lambda, d) = |t(\lambda, d)|^2 \tag{6}$$

From the energy conservation relationship, the absorptivity $A(\lambda, d)$ is given by $A(\lambda, d) = 1 - R(\lambda, d) - T(\lambda, d)$.

According to Kirchhoff's law, under thermal equilibrium conditions, the emissivity of the PDMS film at a given thickness and wavelength satisfies $\varepsilon(\lambda, d) = 1 - |r(\lambda, d)|^2 - |t(\lambda, d)|^2$.

2.3. Numerical Characterization of PDMS Thin Film Emissivity as a Function of Thickness

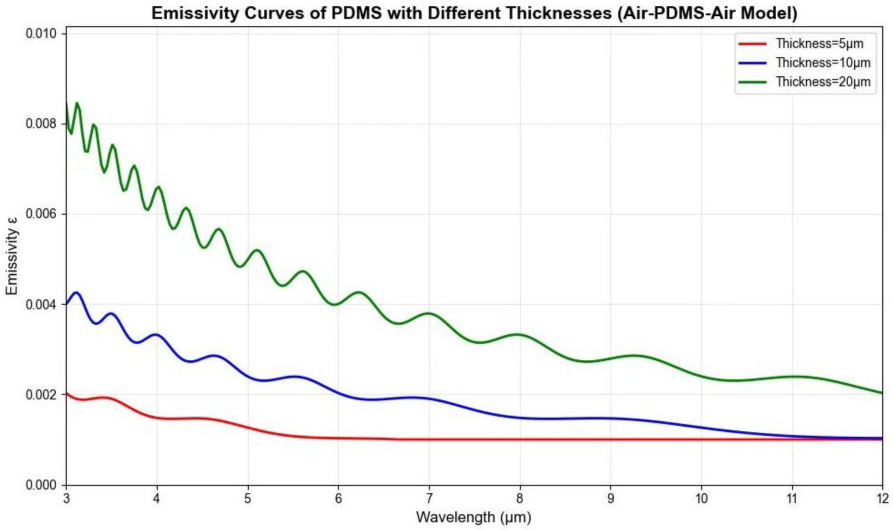

Figure 1. Emissivity curves of PDMS with different thicknesses

As shown in Figure 1, this section describes the variation law of spectral emissivity of PDMS films with wavelength under different thicknesses $d = 5, 10, 20\mu\text{m}$. It can be observed that the emissivity shows a monotonically increasing trend as the film thickness increases. Meanwhile, all curves gradually decrease with increasing wavelength. The periodic fluctuations in the curves are consistent with Fabry-Pérot type thin film interference. As the thickness increases, the variation amplitude of phase thickness enlarges, which makes the superposition effect of multiple reflection terms more significant: the 20 μm film exhibits the most obvious oscillatory structure, while the 5 μm film shows an approximately smooth attenuation. It can thus be concluded that under thick film conditions, the interference effect modulates the spectral structure more significantly, while under thin film conditions, the spectral structure is subjected to stronger absorption suppression.

3. THICKNESS-DRIVEN PERFORMANCE MODELING AND OPTIMIZATION OF PDMS FILMS FOR PASSIVE RADIATIVE COOLING

3.1. Multi-level Model Construction for Daytime Cooling Mechanism of PDMS Films

To analyze the passive radiative cooling capability of PDMS films with different thicknesses under solar irradiation conditions, a framework consisting of an optical model, a thermal radiation model, and an energy balance model is constructed, with the cooling function as the core indicator.

The first step in solving the model is to determine the spectral response of PDMS films at different wavelengths. The optical behavior of the film structure depends on its phase modulation of incident light and interface coupling. The light propagation characteristics inside the film are described by the propagation constant, which together with the film thickness determines the phase delay. The phase delay is introduced into the Transfer Matrix Method (TMM) through the characteristic matrix of the single-layer film, which allows accurate description of the multiple reflection and interference effects of the film [5]. The expression of the characteristic matrix is:

$$M = \begin{pmatrix} \cos\delta & \dfrac{j}{\eta_1}\sin\delta \\ j\eta_1 \sin\delta & \cos\delta \end{pmatrix} \tag{7}$$

Where $\eta_j(j=0,1,2)$ denotes the optical admittance of the medium. Since the overall optical response of the material is determined by the coupled admittance of the film and the substrate, the equivalent optical admittance of the system is:

$$\eta_{eq}=\frac{M_{11}\eta_2+M_{12}}{M_{21}\eta_2+M_{22}} \tag{8}$$

Meanwhile, the relationship between the reflection coefficient, transmission coefficient, spectral reflectivity, transmissivity, and absorptivity can be derived as:

$$A=1-R-T=1-|r|^2-\left|\frac{\eta_2}{\eta_0}t\right|^2=1-\left|\frac{\eta_0-\eta_{eq}}{\eta_0+\eta_{eq}}\right|^2-\left|\frac{\eta_2}{\eta_0}\cdot\frac{2\eta_0}{\eta_0+\eta_{eq}}\right|^2 \tag{9}$$

The above optical calculations yield the absorptivity of PDMS films at different wavelengths, as well as the emissivity obtained based on Kirchhoff's law. Using these spectral absorption and emission data, the optical behavior is mapped to actual radiative energy flux to evaluate the heat exchange process between the film and the environment. To this end, a thermal radiation model is further established to quantify three types of energy contributions: the material's own radiation, atmospheric downward radiation, and solar absorption.

Since the thermal radiation behavior of the material is governed by Planck's blackbody radiation law, its spectral radiance (radiation intensity per unit wavelength) is given by:

$$I_{BB}(\lambda,T)=\frac{2\pi hc^2}{\lambda^5}\left(e^{\frac{hc}{\lambda k_B T}}-1\right)^{-1} \tag{10}$$

To fully describe the radiation process of the film, two additional energy components—atmospheric downward radiation and solar absorption—must be considered. By incorporating the emissivity, the radiative heat dissipation power of the material at temperature T can be determined; the contribution in the atmospheric window (8−13 μm) band is particularly critical for the cooling effect. In this case:

$$P_{rad}=\int_{8\mu\mathrm{m}}^{13\mu\mathrm{m}}\varepsilon(\lambda,d)I_{BB}(\lambda,T)d\lambda \tag{11}$$

The atmospheric downward radiance depends on atmospheric transmittance, and the expression for its absorption power is:

$$P_{atm}=\int_0^{\infty}\varepsilon(\lambda,d)\varepsilon_{atm}(\lambda)I_{BB}(\lambda,T_{amb})d\lambda \tag{12}$$

Since solar absorption is the main loss term for daytime radiative cooling, the expression for its solar absorption power is:

$$P_{sun}(d)=\int_{0.3\mu\mathrm{m}}^{2.5\mu\mathrm{m}}A(\lambda,d)I_{sun}(\lambda)d\lambda \tag{13}$$

The actual cooling effect of PDMS films depends on the net energy flux (per unit area) that the material transmits to outer space. Therefore, three radiative energy components (the material’s own radiation, atmospheric downward radiation, and solar radiation absorption) and non-radiative heat transfer processes need to be integrated into a single energy framework to represent the net cooling capacity of the material at a given temperature and thickness. The expression is:

$$P_{net}(T,d) = P_{rad} - P_{atm} - P_{sun} - h_c\left(T - T_{amb}\right) \tag{14}$$

When $P_{net} > 0$, the film has active cooling potential; When $P_{net} = 0$, the system reaches a steady-state temperature; When P_{net} reaches its maximum value with respect to d, the corresponding d can be regarded as the optimal thickness.

3.2. Influence of Thickness on Net Cooling Power and Optimal Design

(1) Influence of Thickness on Net Cooling Power

When the film thickness varies in the range of 5–50 μm, the variation trend of the net cooling power (calculated by the system) is obtained. The calculation shows that the cooling power exhibits a distinct characteristic of "first rising rapidly, then tending to plateau" as the thickness increases. The results indicate: $d^* \approx 25\mu\text{m}, P_{net,\max} = 30.32\text{W/m}^2$; as the thickness continues to increase, the mid-infrared absorption of the film approaches saturation, causing the cooling performance to enter a plateau region.

(2) Variation Law of Isothermal Cooling Power with Thickness

To further evaluate the theoretical limit cooling capacity of the material at a fixed surface temperature, the net cooling power of films with different thicknesses is calculated under the isothermal condition (i.e., $T_s = T_{amb}$). As the thickness increases from 20 μm to approximately 50 μm, the maximum cooling capacity rises rapidly, reaching a peak of over 90 W/m^2; subsequently, the cooling power shows a slow downward trend with increasing thickness.

(3) Net Cooling Power–Temperature Relationship for Films of Different Thicknesses

To further reveal the influence of thickness on cooling dynamics, this study calculates the net cooling power of PDMS films with different thicknesses as the surface temperature changes. All curves exhibit a linear downward trend as temperature increases. In the relatively low temperature range of 280–300K, PDMS films with thicknesses of 50–200 μm show higher cooling power due to their greater window emissivity; in contrast, films with thicknesses of 10–20 μm have significantly lower cooling capacity across the entire temperature range.

4. MATERIAL SELECTION AND THICKNESS OPTIMIZATION MODEL FOR MULTI-LAYER FILM STRUCTURES

4.1. Material Selection Basis and Design Requirements for Multi-Layer Film Structures

(1) Multi-Layer Film Radiative Cooling Structure and Design

To enhance the passive radiative cooling performance of PDMS films, a multi-layer film structure is introduced. By combining the refractive indices and extinction coefficients of different materials, "spectral selective regulation" can be achieved in the target waveband.

Thus, candidate materials to be paired with PDMS include SiO_2, Ag, Al_2O_3, etc. Multi-layer film schemes with three-layer and four-layer structures are designed, and their optimal net cooling powers are compared; the scheme with better performance is finally selected.

(2) Optical Modeling of Multi-Layer Films: Transfer Matrix Method (TMM)

The spectral reflectivity and transmissivity of multi-layer films are calculated using the transfer matrix method. Assume the structure consists of L layers from top to bottom; the complex refractive index of the j-th layer material is:

$$\tilde{n}_j(\lambda) = n_j(\lambda) + ik_j(\lambda) \tag{15}$$

The thickness of the j-th layer is d_j. For normal incidence, the characteristic matrix of the j-th layer is:

$$M_j = \begin{pmatrix} \cos\delta_j & \dfrac{i}{\eta_j}\sin\delta_j \\ i\eta_j\sin\delta_j & \cos\delta_j \end{pmatrix}, \delta_j = \frac{2\pi\tilde{n}_j d_j}{\lambda} \tag{16}$$

Where η_j is the equivalent optical admittance of this layer. The total transfer matrix of the multi-layer structure is:

The total transfer matrix of the structure is the product of the characteristic matrices of each layer:

$$M = \prod_{j=1}^{L} M_j = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} \tag{17}$$

From the total matrix, the complex reflection coefficient $r(\lambda)$ and transmission coefficient $t(\lambda)$ of the system at wavelength λ can be obtained, further leading to:

$$R(\lambda) = |r(\lambda)|^2, T(\lambda) = \frac{n_2}{n_0}|t(\lambda)|^2 \tag{18}$$

Where n_0 and n_2 denote the refractive indices of air and the substrate, respectively. Since the film contains no active gain medium, and according to Kirchhoff's law, the spectral emissivity of the system equals the absorptivity, i.e.:

$$\varepsilon(\lambda) = A(\lambda) = 1 - R(\lambda) - T(\lambda) \approx 1 - R(\lambda) \tag{19}$$

(3) Optimization Variables and Constraints

In the spectral regulation design of multi-layer films, the material type and thickness of each layer significantly affect the overall distribution of reflectivity, transmissivity, and infrared emissivity. Thus, this study integrates the material selection and layer thickness design of the multi-layer structure into the optimization framework, constructing a mixed decision system that includes discrete and continuous variables.

To maintain model consistency and material feasibility, a unified candidate material pool is first defined: $\mathrm{M} = \{\mathrm{Ag}, \mathrm{Al}, \mathrm{TiO_2}, \mathrm{SiO_2}, \mathrm{PDMS}\}$. Regardless of whether the number of structure layers L is 3 or 4, the material type of each layer is selected from this pool as $m_j \in \mathrm{M}, j = 1, 2, ..., L$.

In addition to material type, the thickness of each layer also has a decisive impact on the film's spectral characteristics. Thickness not only affects visible light interference intensity but also directly controls the Fabry-Pérot interference effect in the mid-infrared window, thereby influencing the radiation coupling efficiency of PDMS. The constraint on thickness is therefore set as $d_j \in [0.1, 20]\mu\text{m}, j = 1, 2, ..., L$.

Under this variable system, the design objective of the multi-layer structure is to maximize its net cooling capacity at a specified ambient temperature. This is essentially a non-convex mixed integer-continuous optimization problem, which requires solution via global optimization methods such as genetic algorithms.

4.2. Global optimization algorithm and result analysis for multi-layer film structures

To automatically search for the optimal material sequence and thickness combination in the unified material candidate pool, this study adopts a genetic algorithm suitable for mixed variables. To enable the genetic algorithm to handle both material types and layer thicknesses simultaneously, each design scheme is represented as a unified decision vector: $\vec{x} = [m_1, ..., m_L, d_1, ..., d_L]$ [6].

For each candidate vector, the corresponding multi-layer film structure is first constructed based on its material sequence and thickness sequence. Then, the TMM model is used to calculate its spectral reflectivity, transmissivity, and emissivity, which are substituted into the net cooling power model; the result is taken as the fitness value of the genetic algorithm.

To make individuals with high fitness more likely to influence the next generation, a three-way tournament selection strategy is adopted. A mutation mechanism with a 5% probability is set to ensure that the finally obtained structure has global optimization potential. The algorithm terminates when the population fitness no longer improves significantly or the iteration upper limit is reached.

The performance of six three-layer structures output by the genetic algorithm is shown in Table 1.

Table 1. Summary of Optimised Performance Results for Six Three-Layer Composite Structures

Serial number	Material composition	Optimum cooling capacity	R_{solar}	ε_{IR}	Note
1	Ag + TiO_2 + PDMS	233.86	0.988	0.535	optimal
2	Al + Ag + PDMS	162.94	0.993	0.368	
3	Al + TiO_2 + PDMS	114.07	0.718	0.864	
4	SiO_2 + Ag + PDMS	59.91	0.981	0.169	Infrared radiation is weak
5	SiO_2 + TiO_2 + PDMS	47.21	0.702	0.756	Insufficient solar reflection
6	Al + SiO_2 + PDMS	16.02	0.930	0.102	Lowest overall performance

Analysis of the six groups of results shows that the Ag-TiO_2-PDMS combination achieves the best trade-off between high solar reflection and moderate infrared emission. The Ag+SiO_2+PDMS combination is the optimal three-layer material structure, with the optimal thicknesses: 0.52µm for Ag, 0.86µm for TiO_2, and 7.17µm for PDMS. Its optimal performance is $P_{\max} = 233.86\text{W/m}^2$ $R_{\text{solar}} = 0.988, \varepsilon_{\text{IR}} = 0.535$. This three-layer structure represents the optimal balance point between solar reflection and mid-infrared emission, and is supported by a clear physical mechanism.

For the four-layer structure, the optimal configuration is the Ag+SiO_2+Al_2O_3+PDMS structure. Its spectral transmittance and reflectance are shown in Figure 2.

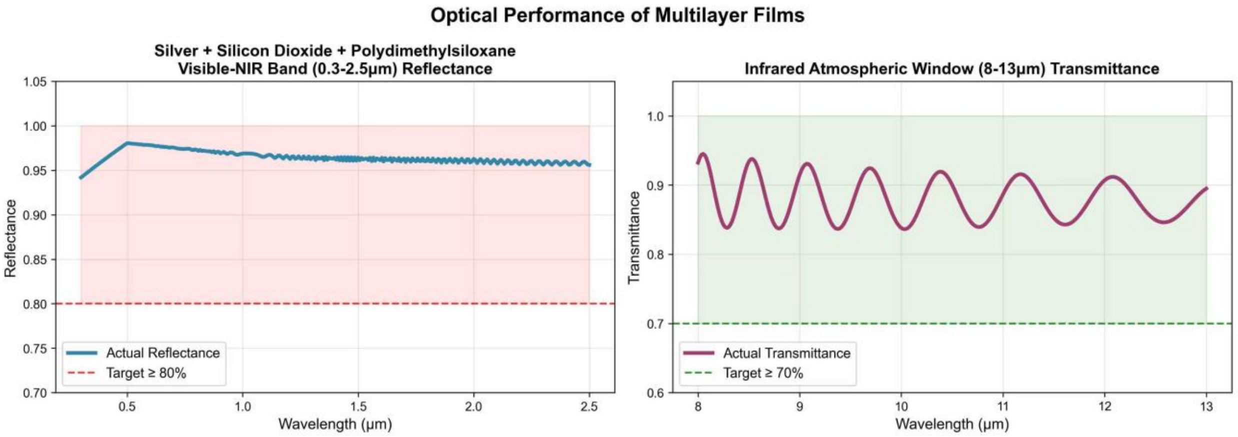

Figure 2. Spectral transmittance and reflectance of the Ag+SiO_2+Al_2O_3+PDMS four-layer structure

The four-layer structure maintains a high reflectance of 0.95-0.98 in the solar band, possessing the same solar absorption suppression capability as the three-layer structure. Comprehensive spectral results show that the four-layer structure improves spectral tunability without significantly increasing absorption loss, and thus has potential for further optimization.

4.3. Comprehensive Optimization Design of Radiative Cooling Materials and Structures

On the basis of multi-layer film combination and thickness optimization, the genetic algorithm is further used for global search of material sequences and layer thicknesses, with "maximization of daytime net cooling power" as the objective function. Considering the dual requirements of high reflectance in the solar band and high emissivity in the atmospheric window, Ag+TiO_2+PDMS is finally determined as the optimal multi-layer film structure; the material combination and structure sequence are silver reflection layer, titanium dioxide functional layer, and polydimethylsiloxane radiation layer.

The thickness optimization results are: 0.52μm for the Ag layer, 0.86μm for the TiO_2 layer, and 7.17μm for the PDMS layer, with a total thickness of only 8.55μm. Combining the optimized thickness and material functions, the final design can be described as follows: The substrate is a continuous Ag mirror reflection layer to ensure the reflection limit; A TiO_2 dielectric layer is deposited on it to fix the interference enhancement channel; The surface is covered with a PDMS radiation film; a micro-textured surface can be used if necessary to further improve scattering reflection and infrared coupling efficiency. Compared with the daytime net cooling power of the traditional single-layer PDMS structure (31.2W/m^2), its core indicator, the net cooling power, is 233.86W/m^2. The solar reflectance is 0.988, which significantly suppresses solar heat absorption; the average emissivity in the 8-13μm atmospheric window is 0.535, achieving the optimal trade-off between "enhancing radiative heat dissipation" and "suppressing solar parasitic absorption."

5. CONCLUSIONS

This paper proposes a unified modeling and optimization method for passive radiative cooling thin-film structures. By coupling optical interference modeling, thermal radiation calculations, and energy balance analysis, it achieves a systematic description of the structural performance. This model can simultaneously handle both single-layer and multi-layer structures within the same framework, reducing uncertainties arising from empirical assumptions and providing a reliable basis for performance prediction. Combined with genetic algorithms for joint optimization of material type and layer thickness parameters, the model demonstrates strong global search capabilities within complex design spaces, enabling effective synergistic control of solar reflectance and mid-infrared radiation. Using PDMS-based films as an example, the optimized structure achieves a solar reflectance close to 0.99 and an atmospheric window emissivity exceeding 0.5 with a total thickness under 10 μm. Its daytime net cooling performance significantly outperforms monolayer structures, validating the model's effectiveness and practical value. Future research may further extend and refine this methodology by integrating real-world operating conditions and fabrication factors.

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