

Inversion of Optical Properties of Porous Membrane Materials Based on Monte Carlo Method

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Abstract: Absorption and scattering are important parameters that reflect the radiation performance of porous materials but cannot be directly measured. By solving the radiation transfer equation by Monte Carlo method, the relationship between the material's absorptivity, scattering, reflectivity and transmittance can be established. Then, when the reflectivity and transmittance have been measured, the absorptivity and scattering can be obtained by genetic algorithm inversion. The absorptivity and scattering of cellulose acetate porous membrane samples were obtained by the above method, which proved the feasibility of the method.

Keywords: Monte Carlo method, genetic algorithm, cellulose acetate porous membrane.

1. Introduction

In recent years, with the intensification of climate change, reflective materials with the advantages of thermal comfort and energy saving have attracted more and more attention [1][2]. Due to the inherent properties of porous materials, high porosity and special microstructure, they still have good reflective performance at a small thickness. Under the same lighting conditions, compared with other materials, reflective materials absorb less heat, which can indirectly produce cooling and heat dissipation effects.

The radiation transfer equation describes the process of electromagnetic waves attenuating when propagating in a medium due to absorption, scattering and other effects. By inverting the radiation transfer equation, the radiation characteristic parameters of the medium can be obtained. Due to the special properties of the radiation transfer equation, it is impossible to obtain the above parameters. There are some studies in this area: J. Hunt [3] conducted a detailed study on the scattering in silica aerogels and proposed a formula for the relationship between transmittance and scattering based on the Beer-Lambert (B-L) law, but the study ignored the absorptivity and the accuracy was not high; L. Zhao, S. Yang [4] et al. used the discrete coordinate method based on the radiation transfer equation to invert the absorptivity and scattering of silica aerogels. In addition, multi-flux method [5][6], finite volume method [7], etc. are also used to simulate radiation transmission. However, these methods discretize the solid angle and medium space when simulating the transmission of light, which will produce certain errors.

Monte Carlo Methods (MC) [8] is a method that can simulate the transmission of radiation in a medium. It can provide a method to calculate the reflectivity and scattering rate through the absorptivity and scattering rate. This process of inferring the radiation transmission path and energy change through radiation and medium properties is also called "forward modeling". The problem faced at this time is to find the best solution from all possible absorptivity and scattering rate solution spaces when the reflectivity and transmittance are known. This process of inferring medium properties through radiation reflection and transmission can also be

called "inversion". The randomness and non-differentiability of the forward modeling results of the Monte Carlo method means that traditional inversion methods such as Newton iteration method that require gradient information cannot be used. In view of this, we choose to use genetic algorithms (GA) to achieve the inversion of the above problems. Genetic algorithms, as a random search and optimization method based on biological evolution theory, are a powerful tool for solving complex problems without relying on gradient information.

Cellulose acetate porous membrane was prepared as a sample, and the spectral hemispherical reflectivity and spectral hemispherical transmittance of the sample were measured. The absorptivity and scattering of the sample were inverted. The results were consistent with the Rayleigh-Gans theory, and the effective scattering diameter was calculated to be about 325nm, indicating that the method has a certain reference significance for studying the optical properties and structure of porous materials.

2. Radiation Transmission in Porous Materials and Monte Carlo Simulation

2.1. Monte Carlo method

The Monte Carlo method repeatedly simulates the transmission of a single photon, decomposes the transmission process into a series of independent sub-processes such as emission, transmission, reflection, absorption and scattering, and counts the destination of each photon, thereby simulating the entire radiation transmission process. In the process of single photon propagation, photons are emitted with the same initial weight, and their movement step length and movement direction are random: the movement step length of the photon is determined according to the attenuation coefficient of the medium, and the change in the propagation direction of the photon at each step is determined by the scattering characteristics of the medium system. The total weight of all photons escaping from the same side of the tissue phantom as the incident light is counted as the reflectivity; the fraction of the total weight of photons that pass through the medium and

escape from the other side of the tissue phantom is counted as the transmittance.

2.1.1. Assumptions of the Monte Carlo method

In order to accurately simulate the transmission of radiation in porous materials and simplify the program and calculation, the following assumptions are made:

(1) Photons are regarded as classical particles, and polarization and wave phenomena are ignored.

(2) It is assumed that an infinitely narrow photon beam acts vertically on a medium with infinite lateral dimensions.

(3) It is assumed that the refractive index of the medium is the same as that of air, that is, the radiation does not attenuate energy or change the propagation direction at the interface between the porous membrane medium and the air. Since the interior and surface of the material are porous and the porosity is high, it can be assumed from reference [5] that the refractive index of the material is close to that of air, and the reflection of light on the surface is extremely small. In addition, according to reference [11], the small uncertainty of the refractive index has little effect on the calculated absorption and scattering rate of the material. In order to simplify the Monte Carlo calculation, it can be assumed that the refractive index of the material is the same as that of air, that is, there is no reflection and refraction of radiation on the surface of the membrane.

(4) It is assumed that the porous membrane material is an optically homogeneous medium, that is, different parts of the material have the same optical parameters.

(5) The thermal radiation of the material itself is ignored.

(6) Assume that the material scatters isotropically. That is, $\forall \lambda, \forall \mu, \forall \mu', \phi_\lambda(\mu, \mu') = 1$, where $g_\lambda = 0$, where g_λ is the asymmetry coefficient of the medium, defined as the average cosine of the scattering angle. This assumption is made because g_λ is difficult to derive from measurements or calculations. In this way, what we get is not necessarily the true absorptivity and scattering rate of the material, but the absorptivity and scattering rate of the equivalent isotropic medium. This method is called isotropic scaling or transport approximation [12]. The relationship between the equivalent extinction coefficient and albedo and the true extinction coefficient and albedo is:

$$\beta_\lambda^* = \beta_\lambda(1 - \omega_\lambda g_\lambda) \quad (5)$$

$$\omega_\lambda^* = \frac{\omega_\lambda(1-g_\lambda)}{(1-\omega_\lambda g_\lambda)} \quad (6)$$

2.1.2. Monte Carlo method to simulate the process of radiation transmission

The simulation process of the Monte Carlo method to simulate the radiation transmission process can be divided into six parts: photon emission, photon transmission, photon absorption, photon scattering, reflection and transmission on the boundary, and photon termination. For details, see the literature [13].

3. Genetic Algorithm Design

In this study, after measuring the hemispherical spectral reflectance and hemispherical spectral scattering through experiments, we hope to invert the absorption and scattering through the Monte Carlo method. Due to the randomness of the Monte Carlo method, it is impossible to use traditional mathematical methods (such as Newton iteration method) to

solve it. When using Monte Carlo to solve the inverse problem, heuristic algorithms including genetic algorithms and annealing algorithms are very effective [14]. Here we choose the genetic algorithm. Genetic algorithm is a commonly used optimization algorithm for solving NP-hard problems. It was first proposed by John Holland of the United States [15] in the 1970s. It is a method for searching for the optimal solution by simulating the natural evolution process. It has the advantages of strong global search ability, strong parallel processing ability, strong adaptability, etc., and can solve complex optimization problems.

In the genetic algorithm, a feasible solution to the problem is called an individual. A specific feature or attribute of an individual is called a "gene", which is the most basic unit in the genetic algorithm. The sum of the genetic information of an individual is called a "chromosome", which forms a complete genetic code through the arrangement and combination of genes. During the execution of the genetic algorithm, chromosomes are manipulated, crossed, and mutated to generate new individuals. In the problem of this paper, the inversion target is the two values of absorption rate a and reflectivity s , so the chromosome of each individual consists of two genes, such as $[a, s]$, where a and s represent absorption rate and scattering rate respectively. In genetic algorithms, fitness is used to evaluate the quality of individuals or solutions in a given problem. In this paper, the fitness of an individual $[a, s]$ is defined as the Euclidean distance between the reflectance $R(a, s)$ and transmittance $T(a, s)$ obtained by the Monte Carlo method and the true values T_0, R_0 : $\sqrt{(T(a, s) - T_0)^2 + (R(a, s) - R_0)^2}$.

The smaller the difference between the calculated reflectivity and transmittance and the true value, the better, so the smaller the fitness, the better. We set a fitness threshold. When the fitness of an individual is less than this value, the individual is considered good enough, and the corresponding absorption rate and scattering rate of this individual are used as the inversion result.

The process of the genetic algorithm is as follows:

(1) Determine the reflectivity r and transmittance t as the target to be inverted; set the fitness threshold when stopping iteration.

(2) Randomly generate an initial population with a population size of $nPop$. Each individual represents a combination of absorptivity and scattering rate.

(3) Evaluation: Use the fitness function to evaluate the fitness of each individual. If the fitness of an individual is less than the threshold, output the individual as the inversion result, otherwise go to step (4).

(4) Selection: Select excellent individuals for reproduction based on the fitness value. Generally speaking, the individuals with higher fitness are more likely to be selected. In this paper, the selection method uses the "roulette selection method".

(5) Select two parent individuals for crossover. Suppose the chromosomes of the two parents are $[a_1, s_1]$, $[a_2, s_2]$, and the chromosomes of the two offspring after crossover are $[a_1, s_2]$, $[a_2, s_1]$.

(6) Mutate the new individuals generated by crossover to introduce new genetic diversity. Mutate with probability p : randomly generate a random number r_1 between (0,1).

When $r_1 \geq p$, do not change the genetic code of the offspring. If $r_1 < \frac{p}{2}$, randomly mutate the absorption rate a of the offspring individual. If $\frac{p}{2} < r_1 < p$, randomly mutate the scattering rate s of the offspring individual.

(7) Calculate the fitness of each individual of the offspring. Sort the offspring individuals and parents according to fitness from low to high, and use the first nPop individuals as the parents of the new generation.

(8) Repeat steps (3)-(7) until the fitness of an individual is less than the set fitness threshold. At this time, terminate the iteration and output the absorption rate and scattering rate values of the individual with the highest fitness as the inversion result.

4. Inversion and Verification of The Absorptivity and Scattering Rate of Cellulose Acetate Porous Membrane

In order to verify the feasibility of the Monte Carlo method and inversion, we prepared a cellulose acetate porous membrane with a thickness of 0.35 mm, measured the

hemispherical spectral reflectance and hemispherical spectral transmittance in the radiation wavelength range of 650 nm to 2500 nm, inverted the absorptivity and reflectivity of the material, and used the Rayleigh-Gans theory to estimate the effective scattering diameter of the material.

5. Experimental Results and Analysis

A cellulose acetate porous membrane sample with a porosity of 84% and a thickness of 0.35 mm was prepared using the evaporation-induced phase separation method assisted by solvent template pore preparation, and the spectral hemispherical reflectance and spectral hemispherical transmittance in the wavelength range of 650 nm to 2500 nm were measured using a UV-visible near-infrared spectrophotometer. The results are shown in Figure 1.

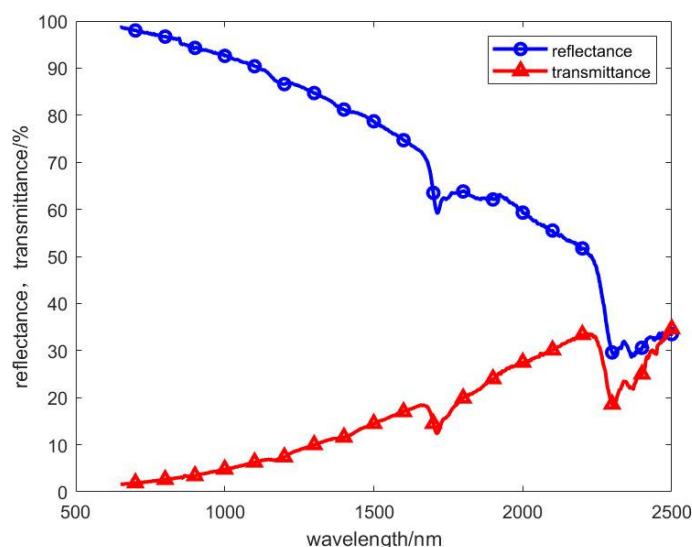


Figure 1. Hemispherical spectral reflectance and transmittance of cellulose acetate porous membrane.

The inversion results are shown in Figure 2.

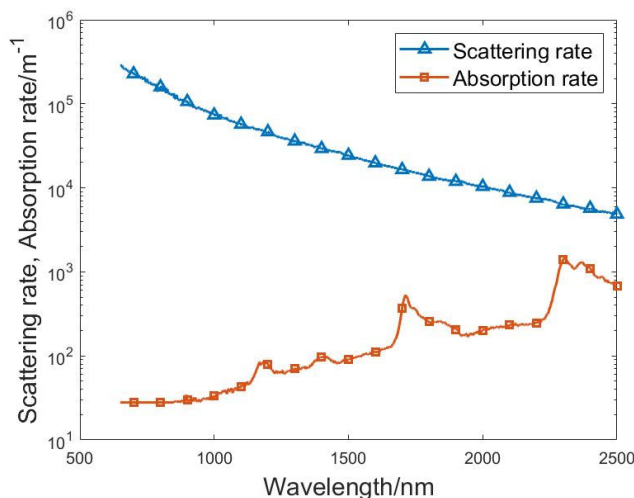


Figure 2. Scattering rate and absorption rate of cellulose acetate porous membrane.

From Figure 1, it can be seen that from 650nm to 2500nm, as the wavelength increases, the reflectivity decreases from nearly 100% to about 30%, and the transmittance increases from 0 to about 30%; absorption peaks at wavelengths around

1700nm and 2300nm can be observed.

From Figure 2, it can be seen that the scattering rate is a continuous smooth function of the radiation wavelength, which decreases with increasing wavelength, and the

decreasing trend is gradually gentle. The absorbance tends to increase with wavelength, and obvious absorption peaks can be observed around 1700nm and 2300nm, corresponding to

the absorption of reflectance and transmittance in the same band in Figure 3.

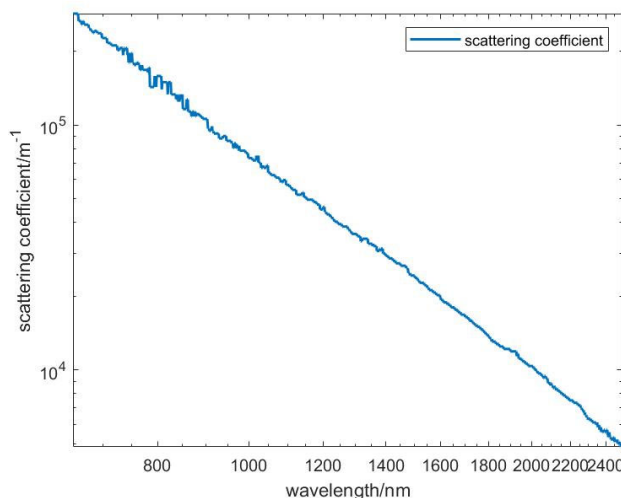


Figure 3. Log-log plot of scattering rate vs. wavelength.

The scattering rate of the material from 650nm to 2500nm is much greater than the absorbance, and it can be concluded that the high scattering rate leads to a larger reflectivity of the porous film.

In order to further study the relationship between wavelength and scattering rate, a log-log plot of scattering rate and wavelength is drawn as shown in Figure 3, and a linear fit is performed in the range of 650nm to 2500nm. According to the Rayleigh-Gans theory, the scattering coefficient can be calculated as follows [16]:

$$S = 4\pi^4 \frac{\rho_1}{\rho_2} \frac{d^3}{\lambda^4} \left(\frac{n^2 - 1}{n^2 + 2} \right)^2 \quad (7)$$

Where S represents the scattering rate, d is the scattering

center diameter, λ is the wavelength, ρ_1 represents the density of the porous membrane, and ρ_2 represents the density of the porous membrane cellulose acetate. n is the relative refractive index of cellulose acetate to air.

Let the porosity be k, and the value of (1-k) is used to replace $\frac{\rho_1}{\rho_2}$, and the refractive index of cellulose acetate is measured. The effective scattering diameter can be calculated by the above formula and the known data, which is about 325nm. According to the pictures taken by the electron microscope (see Figure 4), the skeleton diameter of the cellulose acetate porous membrane ranges from about 100nm to 1000nm, and it can be considered that the calculated effective scattering diameter is basically reliable.

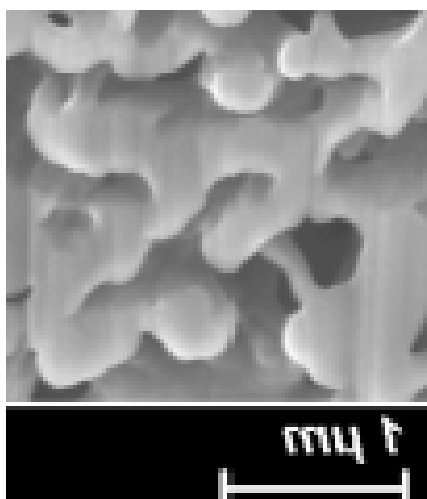


Figure 4. Electron microscopic image of cellulose acetate porous membrane.

6. Conclusion

In this study, the Monte Carlo method was used to simulate the transmission of radiation in porous materials, and the genetic algorithm was used to accurately invert the absorptivity and scattering rate of high-porosity porous materials. A 0.35mm thick cellulose acetate porous membrane

was prepared and the reflectivity and transmittance were measured. The absorptivity and scattering rate were inverted, and the effective scattering diameter of the porous membrane material was calculated to be about 325nm using the Rayleigh-Gans theory. The results show that the method in this paper can be used to invert the absorptivity and scattering rate of porous materials, which has a certain reference

significance for the calculation of optical properties of reflective materials including porous materials and the improvement of optical performance.

References

- [1] Raman A P, Anoma M A, Zhu L, et al. Passive radiative cooling below ambient air temperature under direct sunlight[J]. *Nature*, 2014, 515(7528): 540-544.
- [2] Hernández-Pérez I, Álvarez G, Xamán J, et al. Thermal performance of reflective materials applied to exterior building components—A review[J]. *Energy and Buildings*, 2014, 80: 81-105.
- [3] Hunt A J. Light scattering for aerogel characterization[J]. *Journal of non-crystalline solids*, 1998, 225: 303-306.
- [4] Zhao L, Yang S, Bhatia B, et al. Modeling silica aerogel optical performance by determining its radiative properties[J]. *AIP Advances*, 2016, 6(2).
- [5] Eldridge J I, Spuckler C M, Markham J R. Determination of scattering and absorption coefficients for plasma-sprayed yttria-stabilized zirconia thermal barrier coatings at elevated temperatures[J]. *Journal of the American Ceramic Society*, 2009, 92(10): 2276-2285.
- [6] P. Mudgett and L. Richards, Multiple scattering calculations for technology, *Appliedoptics*, vol. 10, no. 7, pp. 1485–1502, 1971.
- [7] Eymard R, Gallouët T, Herbin R. Finite volume methods[J]. *Handbook of numerical analysis*, 2000, 7: 713-1018.
- [8] Metropolis N, Ulam S. The monte carlo method[J]. *Journal of the American statistical association*, 1949, 44(247): 335-341.
- [9] Zhu C, Liu Q. Review of Monte Carlo modeling of light transport in tissues[J]. *Journal of biomedical optics*, 2013, 18(5): 050902-050902.
- [10] Wilson B C, Adam G. A Monte Carlo model for the absorption and flux distributions of light in tissue[J]. *Medical physics*, 1983, 10(6): 824-830.
- [11] Dombrovsky L A. Radiation heat transfer in disperse systems[M]. Begell House, 1996.
- [12] Lallich S, Enguehard F, Baillis D. Experimental determination and modeling of the radiative properties of silica nanoporous matrices[J]. 2009.
- [13] Wang L, Jacques S L, Zheng L. MCML—Monte Carlo modeling of light transport in multi-layered tissues[J]. *Computer methods and programs in biomedicine*, 1995,
- [14] Mosegaard K, Tarantola A. Monte Carlo sampling of solutions to inverse problems[J]. *Journal of Geophysical Research: Solid Earth*, 1995, 100(B7): 12431-12447.
- [15] Holland J H. Genetic algorithms[J]. *Scientific american*, 1992, 267(1): 66-73.
- [16] Bohren C F, Huffman D R. Absorption and scattering of light by small particles[M]. John Wiley & Sons, 2008.