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Light scattering in porous materials: Geometrical optics and stereological approach

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ABSTRACT

Porous material has been considered from the point of view of stereology (geometrical statistics), as a two-phase random mixture of solid material and air. Considered are the materials having the refractive index with the real part that differs notably from unit and the imaginary part much less than unit. Light scattering in such materials has been described using geometrical optics. These two – the geometrical optics laws and the stereological approach – allow one to obtain the inherent optical properties of such a porous material, which are basic in the radiative transfer theory: the photon survival probability, the scattering phase function, and the polarization properties (Mueller matrix). In this work these characteristics are expressed through the refractive index of the material and the random chord length distribution. The obtained results are compared with the traditional approach, modeling the porous material as a pack of particles of different shapes.

1. Introduction

Porous media are often met both in nature and in technology. Ceramics or pharmaceutical pills can be typical examples of manmade porous materials, while snowpacks, sea foams, or planetary regoliths are examples of natural porous media. Owing to their nature, porous media scatter light strongly. Snowpack, particularly, plays an important role in the Earth's radiance budget through the reflection of sunlight. Light scattering methods are also used to determine the inherent microphysical properties (e.g., porosity or grain size) of porous materials. That is why the description of scattering properties of such materials through their inherent optical and microphysical characteristics is of great importance.

One of the widely used approaches to model light scattering in a porous medium is to consider its grains as independent scatterers of different shapes. For example, hexagonal prisms (columns and plates) with smooth or rough surface [1,2], cylinders, cubes and cuboids [3], fractals [4], or even spheres [5] are often used in modeling light scattering in snow. Also spheres, cubes, and random irregular grains are used to model light scattering in planetary regoliths with the ray tracing method [6–8]. Different shapes with equivalent optical sizes (ratio of volume to mean geometric projection area) provide quite similar results for integral optical characteristics, such as layer albedo [5], but give strongly different results for angular characteristics of light scattering, such as bi-directional reflectance distribution function (BRDF) of a layer [1,2].

At the same time it is obvious that in most cases the porous material is a kind of stochastic medium with grains of different irregular shapes. From this point of view the approach based on the geometrical statistics (a.k.a. stereology) seems promising. In the framework of the stereological approach the porous material is a two-phase random mixture of solid material and air.

The key concept in stereology is random chord length distribution. Any random straight line inside the medium

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has a number of intercepts, lying either in the first (material) or in the second (air) component. These intercepts are called random chords. The chord length is a random value, characterized by some distribution function. We consider the statistically isotropic mixture, i.e., the distribution is independent of the direction. If the intersection of the material–air interface, when moving along the random straight line, is a Markov process (truly random mixture), the chord distribution is exponential.

In geometrical statistics, the two-dimensional (flat) random mosaic with Markov statistics was first, to our knowledge, supposed by Pielou [9]. Afterward, Switzer constructed the model of plane dissection with Markov statistic properties [10]. This model is easily generalized to the 3-dimensional (or, if desired, n-dimensional) case. However, Debye had considered scattering in porous media, using in fact the same model, several years earlier [11].

Debye's consideration is flawless; however it is restricted to the first Born (in optics more often called Rayleigh–Gans–Debye) approximation, i.e., is applicable to the materials with the refractive index close to unit, when the phase shift along the ray path is negligible. At the same time, there are plenty of materials with the refractive index that differs strongly from unit (which is the case of snow, for example, at least in the visible and the near IR range), in which case the method of Debye is inapplicable. On the other hand, the representative grain size in most porous materials is quite great, compared to the wavelength. If in addition the imaginary part of the refractive index is small compared with the real part, this allows us to apply the laws of geometrical optics to light scattering in such a medium. The combination of these two approaches – the geometrical optics and the stereology – allows one to obtain the analytical solution for the intrinsic scattering properties of a porous material.

2. General properties of the stochastic mixture

Let ξ be a random chord length, characterized by distribution functions: $f(\xi)$ with mean a for solid and $g(\xi)$ with mean h for gaps. For the Markov case (pure randomness) the chord distributions are

$$f(\xi)=\frac{1}{a}\exp(-\xi/a),\quad g(\xi)=\frac{1}{h}\exp(-\xi/h). \tag{1}$$

We will use these functions as an important particular case and in the examples of numerical calculations, giving however the general formulae for chord length distributions of any kind. A number of chord length distributions for particles of different shapes can be found in Refs. [12–14], as well as the relationships between the correlation function and the chord length distributions.

For exponential chord length distributions the correlation function of a mixture is also exponential. However, the following relationships hold true for any kind of distributions [11,15,16].

The volume fraction of solid β, the porosity of a mixture ϕ, the bulk density ρ_{bulk}, and the solid material density ρ_{solid} are related to the mean chords:

$$\beta=1-\phi=\frac{\rho_{bulk}}{\rho_{solid}}=\frac{a}{a+h}. \tag{2}$$

The correlation length l_c used by Debye is

$$l_c=\frac{ah}{a+h}. \tag{3}$$

The specific area s of the components interface per unit volume of the mixture is

$$s=\frac{4\phi(1-\phi)}{l_c}=\frac{4}{a+h}. \tag{4}$$

The very close characteristic – specific surface area (SSA) per sample mass – which is very often used to characterize the porous material, such as snow [17,18], equals

$$SSA=\frac{\text{interface area}}{\text{sample mass}}=\frac{s}{\rho_{bulk}}=\frac{4}{(a+h)\rho_{solid}\beta}=\frac{4}{\rho_{solid}a}. \tag{5}$$

Thus, SSA is determined by only one mixture characteristic – the mean chord of a grain. It is necessary to note that this relationship (Eq. (5)) is valid not only for the model of a stochastic medium, but also for any ensemble of independent convex particles. In that case mean chord a is related to a mean particle volume $\langle V\rangle$, a mean particle surface area $\langle S\rangle$, and a mean particle projection area $\langle S_\perp\rangle$ as

$$a=\frac{4\langle V\rangle}{\langle S\rangle}=\frac{\langle V\rangle}{\langle S_\perp\rangle}. \tag{6}$$

This means that the mean chord in the ensemble of convex particles coincides with the standard definition of the effective size (within a constant factor). It is the effective grain size, used in Refs. [1,2,4,5] for irregular snow grains, and effective radius r_{32}, widely used for spherical polydispersions in studies of aerosols and clouds [see, e.g., 19–22]. However, Eq. (5) is more general, as the stochastic mixture does not demand the grain convexity and can be dense-packed.

A number of useful information on geometrical probabilities can also be found in the fundamental monograph of Kendall and Moran [23].

Let us note some other features of the random mixture. First, the normals to the interface surface are distributed isotropically. Second, due to the stochastic nature of the mixture the distance the ray goes between two refractions/reflections does not depend on the refraction angle (conversely to the case of a regular shape, i.e. sphere, where the length of a ray inside a particle is uniquely determined by the refraction angle); so these two (angle and distance) are independent random values.

3. Extinction and absorption

To introduce the extinction coefficient we need to generalize the case of convex particles. This is the standard case of light scattering theory, with the extinction coefficient ε expressed through the extinction efficiency Q_{ext}, a mean particle projection area, a mean particle volume, and particles volume concentration β as follows:

$$\varepsilon=Q_{ext}\frac{\langle S_\perp\rangle}{\langle V\rangle}\beta=\frac{Q_{ext}}{a+h}. \tag{7}$$