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Spectral and angular light-scattering from silica fractal aggregates

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ABSTRACT

This work applies a new model based on the discrete dipole approximation to simulate the scattering of coherent broadband light from fixed and randomly oriented silica fractal aggregates. Two classes of aggregate morphology are considered; the standard aggregate with a single fractal dimension and superaggregates with two dimensions, both of which are generated from a Diffusion-Limited Cluster-Aggregation model. Light scattering from these aggregates is simulated over the short wave infrared to ultraviolet spectral range. We find features in the distribution of the spectral and angular scattered-intensities that do not disappear after orientation averaging and are seen for both types of aggregates. The important role played by the wavelength in q-space analysis is observed and we report the utility of a spectral q-space analysis for multi-dimensional objects like superaggregates with spectrally dependent refractive index.

1. Introduction

Ultrafine particles with a radius smaller than 100 nm such as silicon dioxide, titanium dioxide, and carbon soot are produced by a variety of processes [1]. Aggregates of these particles can form with a fractal morphology that [2,3] obeys a power law relationship between aggregate mass and size as given by Eq. (1) below. An understanding of the spectral and angular light scattering properties of ultrafine fractal aggregates is of interest in a range of scientific and industrial applications. This includes atmospheric studies [4], remote sensing [5], and public health [6]. For instance, man-made aerosols in the atmosphere can occur in the form of aggregates [7]. The influence of these can be measured by multispectral and hyperspectral remote sensing [8,9]. Thus, proper evaluation of the radiative impact of atmospheric aggregates in climate models requires an accurate knowledge of how light scatters from aggregates in the spectral domain [10].

The aim of this article is to study the spectral and angular light scattering behavior of aggregates using a new spectral scattering model based on the Discrete Dipole Approximation (DDA). We focus on silica aggregates as they have been previously studied and because their morphology is believed to contribute strongly to their light scattering properties due to their broadband low-absorption [11,12]. Silica aggregates present a fractal-like structure and can be found either as aerosols from combustion [13] or as colloids in solutions [14]. The observed fractal dimensions of these aggregates range from 1.55 to 1.85 for aerosols [15] and from 1.79 to 1.85 for the colloidal systems [16]. Using aggregates simulated by Diffusion-Limited Cluster-Aggregation (DLCA), we model the spectral and angular light scattering properties with our method. Scattered-light intensity curves are generated for coherent incident light with a wide spectrum ranging from the Short Wave Infrared (SWIR) to the ultraviolet (UV). We then compare our results to the commonly used Rayleigh–Debye–Gans Approximation (RDGA). A q-space analysis [17] is carried out and the role played by the wavelength in the analysis is discussed. The conclusions of our work will motivate the need for

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spectral q-space analysis, particularly in a broader range of spectral-based particle characterization applications.

2. Aggregate structure

Fractal aggregates display a highly complicated structure that is known to satisfy the scaling law:

$$N_m = k_0 \left(\frac{R_g}{R_m}\right)^{D_f} \quad (1)$$

where N_m is the number of monomers in an aggregate, k_0 is a constant called the fractal prefactor, R_m and R_g are the monomer radius and radius of gyration, respectively, and D_f is the fractal dimension. The latter quantity relates to the aggregate morphology and ranges from one for linear, chain-like aggregates to three for compact particles. The volume-equivalent-sphere radius is $R_v = R_m N_m^{1/3}$ and will be used in the following to quantify the aggregate size. This size may vary from a few nanometers to several micrometers. Various experiments employing static [18] or dynamic light scattering [19] are used to determine appropriate values for these characteristics. Although a single fractal dimension generally characterizes most aggregates, examples are known where an aggregate displays several dimensions. Such hybrids are called superaggregates [20]. Here, we use two types of DLCA-simulated aggregates [2,21] and call them the standard aggregate and superaggregate to delineate their different morphology. The standard aggregate has $k_0 = 1.3$, $D_f = 1.8$, $R_v = 89.6$ nm, $R_g = 219.4$ nm, $N_m = 90$ and $R_m = 20$ nm. The superaggregate, however, has a long-range structure described by an approximate fractal dimension of $D_f = 2.6$ and a short-range structure with $D_f = 1.8$ and $k_0 = 1.3$, $R_g = 148.2$ nm, $N_m = 4275$ and $R_m = 30$ nm. Fig. 1 presents images of these simulated aggregates.

3. Spectral scattering model

Simulation of light scattering from an arbitrary aggregate requires a versatile, yet accurate, computational method. Different numerical techniques are available, such as the superposition T-Matrix [22,23] method, the Method of Moments [24], and the DDA [25]. Most commonly used, however, is the RDGA due to its simplicity and agreement with laboratory measurements [26]. Nevertheless, the RDGA is technically valid for aggregates only when internal multiple scattering between the monomers is negligible. Given that the primary interest here is to study the scattering behavior over a wide spectrum, it is not clear when, or if, the RDGA may breakdown. Consequently, we use the DDA, which can simulate scattering from aggregates to essentially arbitrary numerical accuracy provided sufficient computational resources are available [27]. In the DDA model, the aggregate is discretized on a cubic lattice, with each lattice being assigned an electric dipole that responds to the incident wave and couples to the other dipoles in the aggregate. The accuracy of the DDA is then determined by the fineness of this discretization lattice [28].

We based our spectral scattering model on the DDA and call it Spectral-DDA or simply, SDDA. Applications often require computations for aggregates in random orientations. Consequently, we split our model into two operations: one is orientational averaging, and the other repeats the spectral calculations over the SWIR–UV wavelength range. The averaging operation consists of repeated calculations for the aggregate fixed in a set of equally probable random orientations following [29]. This results in a statistical ensemble-average of a single aggregate morphology [30]. However, rotating the aggregate position relative to the dipole lattice can produce so-called shape errors, which are orientation dependent [31]. It is preferable then to fix the DDA lattice to the aggregate and rotate the incident wave propagation and polarization directions while being careful to preserve the transverse nature of the wave. Other orientational-averaging schemes are possible such as analytical methods [32] and numerical evaluation using the T-Matrix that is retrieved from the DDA [33]. The spectral operation consists of calculating the scattered-intensities for wavelengths in the spectral range. To account for dispersion, we use the three-term Sellmeier dispersion equation [34] (Eq. (2)) to model the spectral properties of silica.

$$n = \left(1 + \frac{a\lambda^2}{\lambda^2 - b^2} + \frac{c\lambda^2}{\lambda^2 - d^2} + \frac{e\lambda^2}{\lambda^2 - f^2}\right) \quad (2)$$

The Sellmeier coefficients used in our study are given in Table 1 and are the dispersion coefficients taken from [35]. The imaginary part of the refractive index is neglected since its measured value is $< 10^{-7}$ across the spectral range considered [36]. The SDDA requires large repeated spectral and orientational calculations with the typical size of the dipole lattice being, e.g., $100 \times 100 \times 100$ sites for a single aggregate containing $\sim$4000 dipoles.

4. Results and discussion

The SDDA simulations are performed in the horizontal scattering plane, which is the plane perpendicular to the (linear) polarization of the incident wave. Scattered intensity curves are calculated with 1° angular resolution and averaged over the 200–2000 nm spectrum with 5 nm spectral resolution. Results from the SDDA model are presented for the standard aggregate and superaggregate previously detailed in Section 2. We perform these calculations for these aggregates in both fixed and averaged orientations. The spectral and angular scattered-light

Table 1
Sellmeier coefficients for silica.

	a	b	c	d	e	f
Sellmeier coefficients	0.6961663	0.0684043	0.4079426	0.1162414	0.8974794	9.896161