Range of validity of the Rayleigh–Debye–Gans theory for optics of fractal aggregates

T. L. Farias, Ü. Ö. Köylü, and M. G. Carvalho

The range of validity of the Rayleigh–Debye–Gans approximation for the optical cross sections of fractal aggregates (RDG-FA) that are formed by uniform small particles was evaluated in comparison with the integral equation formulation for scattering (IEFS), which accounts for the effects of multiple scattering and self-interaction. Numerical simulations were performed to create aggregates that exhibit mass fractallike characteristics with a wide range of particle and aggregate sizes and morphologies, including $x_p = 0.01$–1.0, $|m - 1| = 0.1$–2.0, $N = 16$–256, and $D_f = 1.0$–3.0. The percent differences between both scattering theories were presented as error contour charts in the $|m - 1|x_p$ domains for various size aggregates, emphasizing fractal properties representative of diffusion-limited cluster–cluster aggregation. These charts conveniently identified the regions in which the differences were less than 10%, between 10% and 30%, and more than 30% for easy to use general guidelines for suitability of the RDG-FA theory in any scattering applications of interest, such as laser-based particulate diagnostics. Various types of aggregate geometry ranging from straight chains ($D_f \approx 1.0$) to compact clusters ($D_f \approx 3.0$) were also considered for generalization of the findings. For the present computational conditions, the RDG-FA theory yielded accurate predictions to within 10% for $|m - 1|$ to approximately 1 or more as long as the primary particles in aggregates were within the Rayleigh scattering limit ($x_p \leq 0.3$). Additionally, the effect of fractal dimension on the performance of the RDG-FA was generally found to be insignificant. The results suggested that the RDG-FA theory is a reasonable approximation for optics of a wide range of fractal aggregates, considerably extending its domain of applicability.

Key words: Rayleigh–Debye–Gans, optical cross sections, fractal aggregates, laser diagnostics.

1. Introduction

Aggregates composed of spherical particles are found in many engineering and natural environments because of unavoidable Brownian motion. Individual particles that form aggregates are generally small compared to the wavelength of light so that they satisfy the Rayleigh limit of the Mie scattering theory. On the other hand, optical cross sections of aggregates cannot be treated adequately by relatively simple electromagnetic theories for spheres or cylinders because of complex morphology and large dimensions involved.[1] In spite of these difficulties, however, recent developments in fractal concepts revealed that aggregates can be characterized as mass fractals,[2,3] which implies that two parameters, namely, fractal dimension D_f and prefactor k_f are sufficient to represent different shape and size clusters.[4,5] Fractal ideas along with certain assumptions regarding multiple scattering and primary particle properties within aggregates has led to the generalization of the classical Rayleigh–Debye–Gans scattering theory for fractal aggregates (RDG-FA).[3,6–9] The RDG-FA formulation yields analytical expressions, directly relating optical cross sections to aggregate/particle size and morphology. Therefore, this approximate scattering theory is of great practical importance, especially with respect to the interpretation of *in situ* laser diagnostic techniques in particulate-containing environments.[10,11]

Several investigators have explored the capability of the RDG-FA theory to estimate optical properties of aggregates, finding encouraging performance within uncertainties involved in scattering/extinction experiments[8–10] and more exact scattering computations.[12–14] However, these earlier studies have considered limited ranges of particle and aggre-

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gate properties, especially those corresponding to carbonaceous soot in flames. Thus, the objective of the present study is to establish the range of validity of the RDG approximate theory in comparison with the exact scattering solution for a wide range of fractal-like aggregate geometries. Specific parameters for particle and aggregate properties considered in this paper are $x_p = \pi d_p/\lambda = 0.01$–$1.0$, $|m - 1| = 0.1$–2.0, $N = 16 - 256$, and $D_f = 1.0$–3.0, where d_p is the primary particle diameter, λ is the wavelength of light, m is the complex refractive index, N is the number of primary particles in an aggregate, and D_f is the fractal dimension. RDG-FA predictions are compared to the integral equation formulation for scattering (IEFS), which accounts for multiple scattering and self-interaction contributions without any assumptions. We computed absorption, total scattering, and angular scattering cross sections using both RDG-FA and IEFS formulations. The results are presented conveniently by error (percent deviation) contour charts as general guidelines for the suitability of the RDG-FA theory in a particular scattering application, such as the analysis of light scattering/extinction measurements in particle-laden environments.

2. Theoretical Methods

A. Integral Equation Formulation for Scattering

One of the first versions of a scattering theory for aggregates formed by small particles was developed by Jones[15] based on the original integral equation formulation of Saxon.[16] Although several investigators have adopted different schemes for the aggregate scattering problem, Ku and Shim[17] recently unified most of these earlier approaches in the literature that involve the following set of $3N \times 3N$ linear equations for obtaining the internal electric field of each particle $\mathbf{E}_j$:

$$\mathbf{E}_j = \left(\frac{3}{m^2+2}\right)\mathbf{E}_{\text{inc},j} + i\left(\frac{m^2-1}{m^2+2}\right) \times x_p^2 j_1(x_p) \sum_{k=1,\neq j}^{N} \bar{\mathbf{T}}_{jk}\mathbf{E}_k + s_j\mathbf{E}_j; \quad j = 1, 2, \ldots, N, \tag{1}$$

where $m = \eta + i\kappa$ represents the complex refractive index ($i = \sqrt{-1}$), $\mathbf{E}_{\text{inc}} = \mathbf{E}_o \exp(ikz)$ represents the incident electric field that propagates along the z axis with a wave number of $k = 2\pi/\lambda$, $j_1(x_p)$ is a first-order spherical Bessel function of the first kind, and $\bar{\mathbf{T}}$ is the scattering matrix. The self-interaction coefficient s_j is insensitive to the locations of primary particles within an aggregate, and its value generally increases with x_p and m. Equation (1) divides an aggregate into sufficiently small particles so that the internal field within each particle is assumed to be uniform and considers not only the phase differences but also the multiple scattering and self-interaction contributions. Once the internal field of each spherical particle is known from Eq. (1), the following absorption, total scattering, and differential scattering cross sections can be obtained for an aggregate with N uniform size particles, respectively,

$$C_{\text{abs}} = \frac{4\pi}{k^2} x_p^2 j_1(x_p)\text{Im}(m^2 - 1) \sum_{j=1}^{N} |\mathbf{E}_j|^2, \tag{2}$$

$$C_{\text{sca}} = \frac{4\pi}{3k^2} x_p^4 j_1^2(x_p)|m^2 - 1|^2 \sum_{j=1}^{N}\sum_{k=1}^{N} \mathbf{E}_j \,\text{Re}(\bar{\mathbf{T}}_{jk})\mathbf{E}_k^*, \tag{3}$$

$$C_{pp}(\theta, \phi) = \frac{1}{k^2} x_p^4 j_1^2(x_p)|m^2 - 1|^2 \left|\sum_{j=1}^{N} \exp(-ikr_j \cos\beta_j)(E_{j,\theta}\hat{\theta} + E_{j,\phi}\hat{\phi})_{pp}\right|^2, \tag{4}$$

where the complex conjugate is represented by an asterisk, the real and imaginary parts of a complex quantity by Re and Im, respectively, the direction of the scattered field by spherical coordinates θ and ϕ (unit vectors by $\hat{\theta}$ and $\hat{\phi}$), the position of each individual unit by (r_j, θ_j, ϕ_j), the direction of polarization by subscript $_{pp}$, and $\cos\beta_j = \cos\theta_j \cos\theta + \sin\theta_j \sin\theta \cos(\phi_j - \phi)$. Additional details about the IEFS theory can be found in Ku and Shim[17] and Lou and Charalampopoulos.[18]

It should be emphasized that the above formulation is not restricted to aggregate geometries, i.e., one can apply it to any arbitrary shape scatterer by dividing it into small computational cells. Additionally, it is conceivable to treat the optics of aggregates with relatively large particle diameters and nonuniform particles.[19] Also note that one can reduce Eqs. (1)–(4) to other solutions in the literature by making appropriate assumptions.[20] However, the aggregate scattering formulation presented here is the most general approach without any approximation. As a result, the IEFS satisfies the optical theorem,[21] i.e., the sum of the total scattering and absorption cross sections is exactly equal to the following extinction cross section of an aggregate:

$$C_{\text{ext}} = \frac{4\pi}{k^2} x_p^2 j_1(x_p)\text{Im}\left[(m^2 - 1) \sum_{j=1}^{N} \mathbf{E}_{\text{inc}}^* \cdot \mathbf{E}_j\right]. \tag{5}$$

The IEFS is the most theoretically sound and elegant method, however, it is computationally intensive for treating relatively large and/or polydisperse aggregates that can be encountered in several applications such as the organic particulates produced during combustion of various hydrocarbon fuels. Moreover, it requires *a priori* knowledge of the primary particle positions within each aggregate in order to compute the optical cross sections.

B. Rayleigh–Debye–Gans Fractal Aggregate Theory

The fundamental requirement of the RDG scattering theory is to subdivide an arbitrary shape scatterer into small enough units (the Rayleigh limit) so that phase shift corresponding to any point in the scatterer is negligible ($2x_p|m - 1| \ll 1$).[22] Additionally, each subunit is assumed to be unperturbed by the presence of other particles in the aggregate, i.e., both the multiple scattering and self-interaction contributions are negligible. This means that the internal field within each particle is expressed simply as follows:

$$\mathbf{E}_j = \left(\frac{3}{m^2 + 2}\right)\mathbf{E}_{\text{inc},j}. \tag{6}$$

If $j_1(x_p) = (\sin x_p)/x_p^2 - (\cos x_p)/x_p$ is approximated by $x_p/3$, then it is trivial to show that Eqs. (2), (3) and (4) will yield the following optical cross sections within the RDG approximation:

$$C_{\text{abs}} = N\frac{4\pi x_p^3}{k^2}\,\text{Im}\left(\frac{m^2 - 1}{m^2 + 2}\right) = NC_{\text{abs}}^{\,p}, \tag{7}$$

$$C_{\text{sca}} = N^2\frac{8\pi x_p^6}{3k^2}\left|\frac{m^2 - 1}{m^2 + 2}\right|^2 g(x_a) = N^2C_{\text{sca}}^{\,p}g(x_a), \tag{8}$$

$$C_{vv} = N^2\frac{x_p^6}{k^2}\left|\frac{m^2 - 1}{m^2 + 2}\right|^2 f(w_a) = N^2C_{vv}^{\,p}f(w_a), \tag{9}$$

where the superscript p denotes particle properties given by the Rayleigh scattering theory.[21,22] The total scattering factor $g(x_a)$ and form factor $f(w_a)$ are generally dependent on the scatterer morphology. For randomly oriented fractallike aggregates, the average form factor is expressed as[3,6–10]

$$f(w_a) = \begin{cases} \exp\left(-\dfrac{w_a^2}{3}\right), & \text{Guinier regime} \\ w_a^{-D_f}, & \text{power-law regime} \end{cases}, \tag{10}$$

where $x_a = 2\pi R_g/\lambda$ with R_g being the radius of gyration of an aggregate, and $w_a = 2x_a \sin(\theta/2)$ with θ being the scattering angle. In Eq. (10), the boundary between the Guinier and power-law regimes is taken to be $w_a^2 = 1.5D_f$, following Dobbins and Megaridis.[7] On the other hand, $g(x_a)$ can be approximated by different forms depending on whether the power-law regime is reached for $\theta \le 180°$; see Köylü and Faeth[8] for these expressions.

Although the RDG-FA approximation does not satisfy the optical theorem, the above simple expressions make practical applications involving realistic morphology and size aggregates tractable. Moreover, this approximate formulation demonstrates that the optical properties of aggregates are substantially different from those of particles comprising them. For example, Eq. (7) suggests that the absorption is not affected by the aggregation process. On the other hand, Eqs. (8) and (9) imply that the aggregation enhances total scattering by $Ng(x_a)$ and angular scattering by $Nf(w_a)$ over the sum of the individual contributions of particles in the aggregate. Equation (10) also indicates that the radius of gyration, R_g, is the only determining aggregate parameter in the Guinier (small-angle) regime, whereas fractal dimension D_f, appears solely in the power-law regime, i.e., angular scattering patterns are sensitive to the morphology of the scatterer entirely at large angles. This implies that two different scatterers with different morphologies (D_f) but with the same size (R_g) can be distinguished only if one analyzes the large-angle scattering pattern because their scattering behavior is identical in the Guinier regime.

C. Simulation of Fractallike Aggregates

Aggregates were generated in order to obtain primary particle positions that are needed for computations of optical cross sections. Therefore, numerical simulations were performed to create aggregates that exhibit mass fractallike characteristics, i.e., the following statistical relationship between N, the number of particles in an aggregate, and R_g, the radius of gyration, applies:

$$N = k_f\left(\frac{R_g}{d_p}\right)^{D_f}. \tag{11}$$

A wide range of aggregate and particle characteristics together with various morphologies was covered during the present simulations in order to assess the range of applicability of RDG-FA theory within the computational limitations. Specifically, $x_p = 0.01$–1.0, $|m - 1| = 0.1$–2.0 ($\eta = 1 + \kappa$), $N = 16$–256, and $D_f = 1.0$–3.0, all of which correspond to several types of mass fractal objects subject to incident light with wavelength ranges from the ultraviolet to the far infrared. 32 different realizations of the same aggregate size, each sampled at 16 different orientations, were averaged to obtain statistically significant predictions with less than 10% numerical uncertainties (95% confidence interval). Additional details regarding the specific methods of aggregate simulations can be found elsewhere.[5,14,19]

3. Results and Discussion

We computed optical cross sections using the IEFS and RDG-FA theories on the same simulated aggregates by employing the particle positions directly for both scattering methods. This seemed to be necessary in order to minimize any potential artificial differences between aggregate simulations and Eq. (11) as well as between finite and random orientations, which considerably affect computations of the differential scattering cross section, especially in the power-law regime. However, these minor compatibility issues should be considered only when a comparison of methods is involved, i.e., the RDG-FA expressions given by Eqs. (7)–(10) can be employed together with the statistical fractal relationship of Eq. (11). The following discussion will include percent deviations of the RDG-FA from the IEFS for the absorption, total scattering, and differential scattering cross sections of fractallike aggregates that are

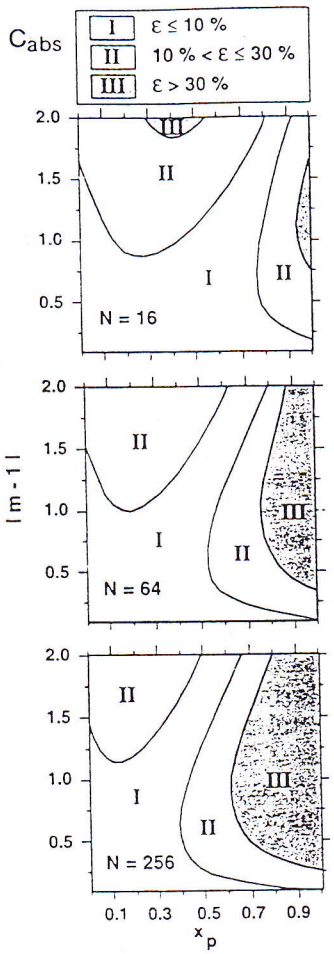

Fig. 1. Percent deviation contours for the accuracy domains of the RDG-FA approximation for predicting the absorption cross sections of various size DLCC aggregates ($D_f \approx 1.8$). In region I, the RDG-FA and IEFS theories agree to within 10%; the differences are between 10% and 30% in region II whereas they are greater than 30% in region III.

representative of diffusion-limited cluster–cluster aggregation (DLCCA). The results are conveniently presented by error contour charts for easy to use general guidelines in any aggregate scattering applications of interest. One can then evaluate the effect of fractal dimension on the validity of the RDG-FA scattering approximation by considering various other geometries with D_f in the range of 1.0–3.0.

A. Absorption Cross Section

The percent differences in absorption cross sections between the RDG-FA and IEFS solutions are illustrated in Fig. 1 for various refractive indices, particle size parameters, and number of particles in an aggregate. The results shown include DLCC aggregates that are typically given by $D_f \approx 1.8$ and $k_f \approx 8.0$.[5,23] The percent deviation contours are represented in the $|m - 1|x_p$ domain that involves three distinct regions. Region I is limited to the range of values of $|m - 1|$ and x_p for which the RDG-FA and IEFS theories agree to within 10%. The differences are between 10% and 30% in region II, whereas they exceeded 30% in region III. The RDG-FA theory can be used with good accuracy in region I and with caution in region II. However, it is clearly not a reliable approximation in region III in which a better optical treatment such as the IEFS formulation is necessary. Note also that the IEFS solution has been verified against the Mie theory for spherical aggregate geometries,[14,20] finding differences in the absorption and total scattering cross sections that never exceeded 1% for x_p to 0.5 and 2% for $x_p = 1.0$. Therefore, this exact scattering formulation is suitable for x_p ranges considered in this study without using multidipoles for the total optical properties.

As can be seen in Fig. 1, the RDG-FA approximation is best for small values of $|m - 1|$ and x_p as a consequence of the fundamental assumptions involved in its derivation. Similar observations were reported for spheres and cylinders by several investigators, as summarized by Kerker.[22] However, the requirements for the applicability of the RDG-FA theory are not as strict as expected because region I extends into domains in which $|m - 1|$ and x_p can be as large as 2 and 1, respectively, for $N = 16$. This is evidently a result of the canceling effects of large refractive indices and optical size parameters on the absorption cross section. In fact, for large refractive indices, the RDG-FA approximation tended to underpredict the IEFS results whereas for large primary particle sizes the opposite effect was observed. From a physical point of view, region I in Fig. 1 represents the domain in which the absorption cross sections of an aggregate and the individual particles composing them are identical, i.e., $C_{abs} = NC_{abs}^{P}$. As N increases from 16 to 256, it is obvious from Fig. 1 that region I shrinks. In other words, the RDG-FA approximation deviates more from the IEFS in estimating absorption cross section as the aggregation continues to increase the cluster size. Nevertheless, the RDG-FA theory seems to be reliable for x_p to 0.4 when $|m - 1|$ is approximately 1, which corresponds to $m = 1.7 + i0.7$.

B. Total Scattering Cross Section

Figure 2 shows the percent deviations of the RDG-FA from the IEFS predictions for total scattering cross sections of DLCC aggregates, similar to those in Fig. 1. For $N = 16$, region I, which represents the 10% deviation regime, covers almost half of the chart whereas the agreement becomes poorer as N increases, with trends identical to the absorption cross section. A comparison of Figs. 1 and 2 indicates that the RDG-FA performs better in predicting C_{abs} than C_{sca} of DLCC aggregates. However, the results shown in Fig. 2 still confirm the applicability of this approximate scattering theory as long as the diameter of particles that form the aggregates is small compared to the wavelength of light, e.g., $x_p \leq 0.3$. On the other hand, it is evident that the refractive index of particles does not have to be close to 1, which is one of the classical requirements for the validity of the RDG theory.[22] In fact, the results in Fig. 2 suggest that the RDG-FA is in good agreement with the IEFS to $|m - 1| = 2$ for $x_p \leq 0.3$. This is apparently a

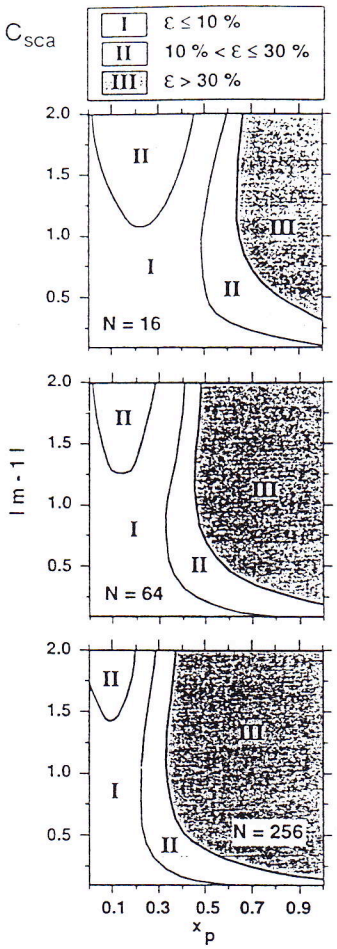

Fig. 2. Contour charts for the accuracy domains of the RDG-FA theory for predicting the total scattering cross sections of various size DLCC aggregates. Definitions of the regions are similar to Fig. 1.

result of the RDG formulation presented in this study [see Eqs. (7)–(9)], which involves the refractive-index function in full form, i.e., $G(m) = (m^2 - 1)/(m^2 + 2)$. Therefore, by not making the approximation that $G(m) \approx 2(m - 1)/3$, the restriction of near-unity refractive index for applicability of the RDG-FA theory can be relaxed, extending its range of validity considerably for absorption and total scattering cross sections of aggregates.

C. Angular Scattering Cross Sections

The contour charts for the differential scattering cross sections (vertically polarized incident and scattered light) at three different angles of 0, 45, and 90 deg are illustrated in Figs. 3, 4, and 5, respectively. Region I in all three figures represents the $|m - 1|$ and x_p values for which the multiple scattering and self-interaction effects are negligible. Specifically, region I in Fig. 3 is the domain in which $C_{vv}(0°) \approx N^2 C_{vv}^{\,p}$ to within 10% accuracy. It is interesting to note that the limits of the percent deviation contours for forward scattering in Fig. 3 are similar to the ones for total scattering in Fig. 2. However, as the scattering angle increases from 0° to 90°, the agreement between the RDG-FA and IEFS theories becomes poorer. For example, for $|m - 1| = 1.0$ and $N = 64$, the difference between the approximate and exact scattering theories is less than 10% for $x_p = 0.4$ at 0°, whereas the same domain of region I is recovered for x_p to approximately 0.3 and 0.25 at 45° and 90°, respectively. At this point, it should be emphasized that the IEFS predictions for 90° scattering angle are questionable for $x_p \geq 0.5$ after which it is necessary to subdivide each primary particle further into smaller computational cells. Nevertheless, Fig. 5 still reflects the trends of the applicability domains of the RDG-FA theory, i.e., it should not be used as a good theoretical scattering tool at large angles after x_p reaches approximately 0.25 for moderate size aggregates.

The results presented so far for DLCCA that are characterized by $D_f \approx 1.8$ suggest that the RDG-FA theory is an excellent approximation for small values of particle diameter, which implies that the multiple scattering and self-interaction terms in the aggregate scattering formulation are generally negligible as long as x_p is less than approximately 0.3. This fact extends the criteria given by Berry and Percival[24] for this kind of fractal aggregates. Apparently, the optical predictions of the RDG-FA formulation become more satisfactory as the refractive index, number of primary particles within an aggregate and scattering angle decrease. Moreover, the differences between the RDG-FA and IEFS predictions of optical cross sections increase from absorption to total scattering, reaching their maximum values for differential scattering cross sections at large angles. On the other hand, the present findings suggest that the following classical requirements can be relaxed somewhat for the validity of the RDG-FA theory: $|m - 1| \ll 1$ and $2x_p|m - 1| \ll 1$. Generally, the error contour charts indicate that the RDG-FA agrees with the IEFS to within 10% for $|m - 1|$ to 1 and $2x_p|m - 1|$ to 0.6. In fact, the error contour charts presented for DLCCA indicate that the deviation between scattering theories is relatively less sensitive to the complex refractive index than the optical particle size. In Subsection 3.D we discuss the effect of the fractal dimension on the applicability of the approximate theory in order to generalize further this study for any aggregate geometry.

D. Effect of Fractal Dimension

Figure 6 illustrates the percent deviations of the RDG-FA from the IEFS formulations for the predictions of various optical cross sections as a function of the fractal dimension for typical values of $|m - 1| = 0.75$, $x_p = 0.3$ and $N = 64$. The simulated aggregate was approximately a straight chain in the case of $D_f \approx 1.0$ whereas a compact cube was adopted to represent $D_f \approx 3.0$. The first thing to realize in Fig. 6 is that the differences between the RDG-FA and IEFS generally increase with increasing fractal dimension, i.e., with the compactness of aggregates. However, for the parameters considered in Fig. 6, this behavior is less significant for the absorption and forward-scattering cross sections, which seem to be relatively

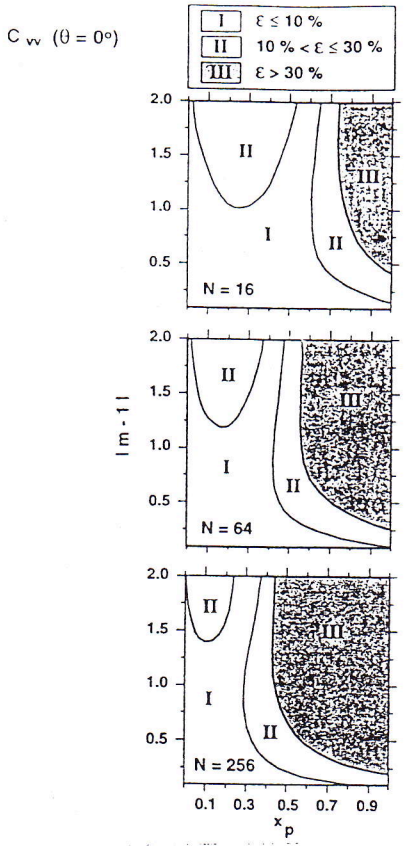

Fig. 3. Contour charts for the accuracy domains of the RDG-FA theory for predicting the vertically polarized angular scattering cross sections at $\theta = 0°$ of various size DLCC aggregates. Definitions of the regions are similar to Fig. 1.

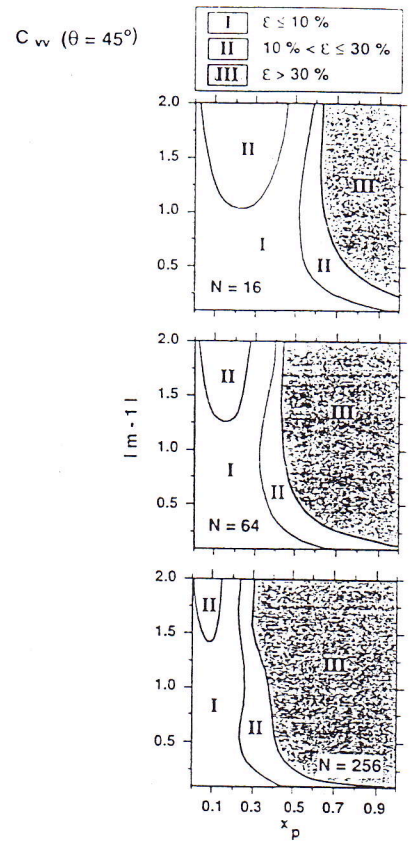

Fig. 4. Same as Fig. 3 except for $\theta = 45°$.

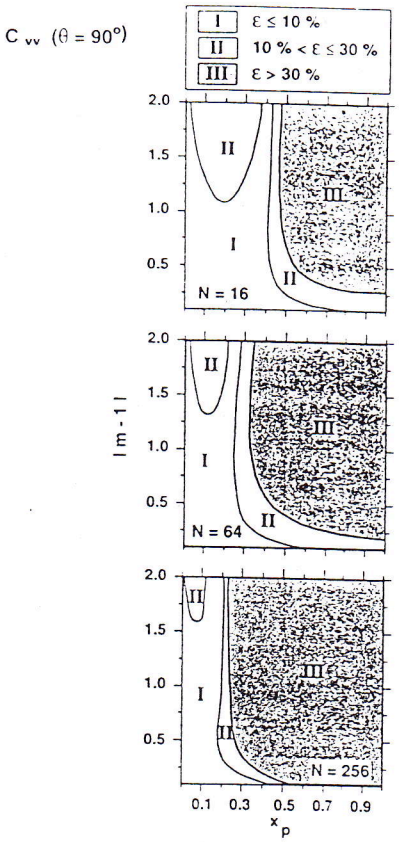

Fig. 5. Same as Fig. 3 except for $\theta = 90°$.

insensitive to the changes in fractal dimension. This immediately suggests that the effect of aggregation on absorption cross section is negligible even for compact aggregate geometries. Additionally, the forward-scattering cross section of an aggregate with any fractal dimension can be accurately predicted by $N^2C_{vv}{}^P$, extending the applicability range of RDG-FA theory for other fractallike shapes with D_f to 3.0. Similar behavior was also observed for C_{vv} at 45°. On the other hand, the total scattering together with the angular scattering cross sections at 90° are the most affected optical properties by variations in D_f. As can be seen from Fig. 6, the maximum differences between scattering theories of interest for $D_f = 3.0$ are approximately 15% for C_{sca} and 30% for $C_{vv}(90°)$.

To evaluate the influence of fractal dimension on the performance of the RDG-FA approximation for a broader range of parameters, we reproduced error contour charts in the $|m - 1|$ and D_f domains for absorption and total scattering cross sections in Figs. 7 and 8, respectively. These two figures, with the definitions of regions being similar to Figs. 1–5, illustrate additional combinations of $|m - 1|$, x_p, and N. Figure 7 generally supports the conclusions drawn from Fig. 6, i.e., the 10% reliability domain of the RDG-FA scattering approximation is almost independent of fractal dimension, except possibly for the cases of $x_p = 0.3$ and $N = 256$. This verifies that the absorption cross section of an aggregate with D_f as large as 3.0 can be represented as $NC_{abs}{}^P$. The total scattering results shown in Fig. 8 also indicate that D_f insignificantly affects the performance of the RDG-FA approximation as long as the primary particle size is small ($x_p \approx 0.1$). On the other hand, for $x_p = 0.3$ [Fig. 8(b)], the RDG-FA predictions of C_{sca} become poorer as D_f increases. Apparently, a bal-

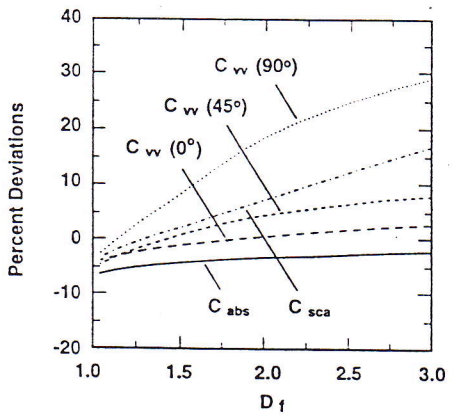

Fig. 6. Effect of fractal dimension D_f on the accuracy of the RDG-FA theory for predicting the various optical cross sections of fractallike aggregates with $|m - 1| = 0.75$, $x_p = 0.3$, and $N = 64$. The simulated aggregate is a straight chain for $D_f = 1.0$ and a compact cube for $D_f = 3.0$.

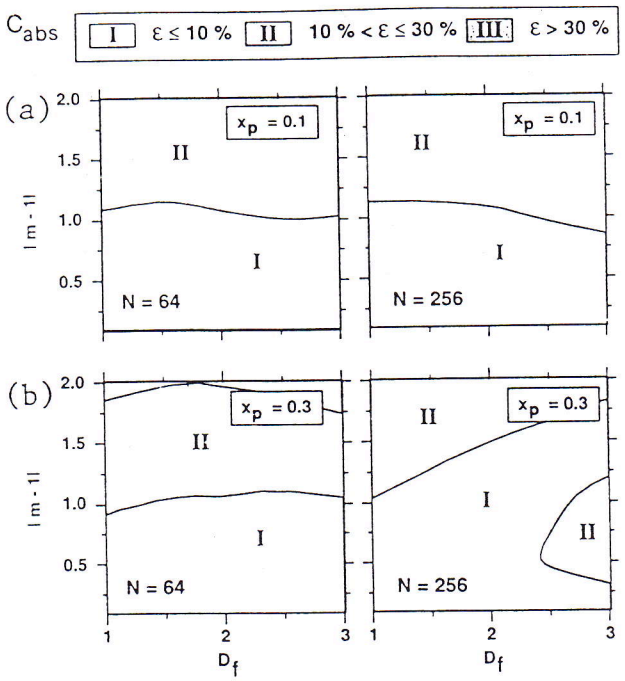

Fig. 7. Error contour charts for the accuracy domains of the RDG-FA theory for predicting the absorption cross sections of various aggregate morphologies with $1 \le D_f \le 3$. Definitions of the regions are similar to Fig. 1.

ancing effect between m and D_f appears to exist for the total scattering cross sections, similar to the canceling behavior between m and x_p as shown in Figs. 1–5. Subsequently, region I in Fig. 8 extends to larger refractive indices and fractal dimensions.

The overall results discussed with respect to Figs.

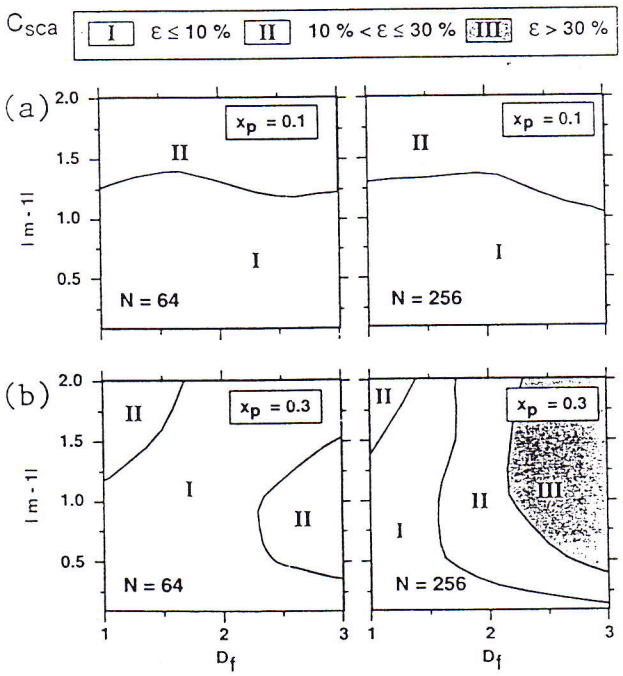

Fig. 8. Error contour charts for the accuracy domains of the RDG-FA theory for predicting the total scattering cross sections of various aggregate morphologies with $1 \le D_f \le 3$. Definitions of the regions are similar to Fig. 1.

6–8 generally suggest that the RDG-FA theory is a reasonable approximation for optics of aggregates with $D_f \le 2$. Moreover, for more compact geometries with D_f as large as 3.0, the RDG-FA approximation continues to be appropriate as long as the size of the primary particles is small. However, the value of $x_p = 0.3$, suggested in this study as an upper limit for the applicability of the RDG-FA theory for aerosols with $D_f \approx 1.8$, decreases with the compactness of aggregates. Indeed, Singham and Bohren[12] also concluded that the interactions among particles are negligible even for a fractal dimension as high as 2.5, which is consistent with the findings of this study.

4. Conclusions

The range of validity of the Rayleigh–Debye–Gans approximation for the optical cross sections of fractal aggregates that are formed by uniform small particles has been evaluated in comparison with the integral equation formulation for scattering, which accounts for the effects of multiple scattering and self-interaction. We performed numerical simulations to create aggregates that exhibit mass fractal-like characteristics with a wide range of particle and aggregate sizes and morphologies, including x_p = 0.01–1.0, $|m - 1|$ = 0.1–2.0, N = 16–256, and D_f = 1.0–3.0. The percent differences between both scattering theories were presented as error contour charts in the $|m - 1|$ and x_p domains emphasizing various size diffusion-limited cluster–cluster aggregates. These charts conveniently identified the regions in which the differences were less than 10%, between 10% and 30%, and more than 30% for easy to use general guidelines for suitability of the RDG-FA theory in any scattering applications of interest. The effect of the fractal dimension on the performance of the RDG-FA scattering theory was also assessed to generalize further the findings for any type of aggregate geometries ranging from approximate straight chains ($D_f \approx 1.0$) to compact clusters ($D_f \approx 3.0$). The major conclusions of this study can be summarized as follows:

(1) For the current computational conditions, the RDG-FA theory yields accurate predictions to within 10% for $|m - 1|$ to approximately 1 or more as long as the primary particles in aggregates are within the Rayleigh scattering limit. This wide range of $|m - 1|$ observed for the applicability of the RDG-FA is apparently a consequence of not approximating the refractive-index function $(m^2 - 1)/(m^2 + 2)$ by $2(m - 1)/3$.

(2) The RDG-FA approximation is in good agreement with the IEFS solution generally for $x_p \le 0.3$ for various DLCC aggregates, although it may still be accurate for larger primary particles within moderate size aggregates. This extends the criteria of negligible multiple scattering and self-interaction contributions to the scattering field for a broader range of x_p's.

(3) The effect of fractal dimension on the range of validity of the RDG-FA approximation for predictions

of absorption and small-angle differential scattering cross sections is found to be relatively insignificant. Although its performance for C_{sca} and $C_{vv}(90°)$ becomes poorer as D_f increases, the RDG-FA theory still appears to be a reasonable approximation for optics of fractal aggregates even for some compact geometries with $D_f \geq 2$.

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