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Efficiency Factors and Radiation Characteristics of Spherical Scatterers in an Absorbing Medium

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This paper investigates the radiative properties of bubbles or particles embedded in an absorbing medium. It aims first at determining the conditions under which the absorption by the surrounding medium must be accounted for in the calculation of the efficiency factors by comparing results from the (1) Mie theory, (2) far-field, and (3) near-field approximations. Then, it relates these approximations for a single particle to the effective radiation characteristics required for solving the radiative transfer in an ensemble of scatterers embedded in an absorbing medium. The results indicate that the efficiency factors for a spherical particle can differ significantly from one model to another, in particular for large particle size parameter and matrix absorption index. Moreover, the effective scattering coefficient should be expressed based on the far-field approximation. Also, the choice of the absorption efficiency factor depends on the model used for estimating the effective absorption coefficient. However, for small void fractions, absorption by the matrix dominates and models for the absorption coefficient and efficiency factor are unimportant. Finally, for bubbles in water, the conventional Mie theory can be used between 0.2 and 200 μm except at some wavelengths where absorption by water must be accounted for. © 2006 Optical Society of America

NOMENCLATURE

a	radius of particles or bubbles.
a_j, b_j, c_j, d_j	Mie coefficients.
a_j', b_j', c_j', d_j'	coefficients in Equations (12) and (13).
C	coefficient.
f_1	size distribution function of particles.
f_v	volume void fraction or porosity.
Im	imaginary part of a complex number.
j	index number.
k	absorption index of the continuous phase.
k'	absorption index of the scatterer.
m	complex refractive index of the continuous phase, $m = n - ik$.
m'	complex refractive index of the scatterer, $m' = n' - ik'$.
n	refractive index of the continuous phase.
n'	refractive index of the scatterer.
Q	efficiency factor.
r	distance to the particle center.
Re	real part of a complex number.
x	size parameter, $2\pi a/\lambda$.

Greek symbols

κ	absorption coefficient.
λ	wavelength.
σ	scattering coefficient.
ϕ	scattering phase function of a single bubble.
Φ	scattering phase function of the continuous phase containing polydispersed bubbles.
Θ	angle between the incident and scattered radiations.
ζ, φ	Riccati-Bessel functions.
ζ', φ'	derivatives of the Riccati-Bessel functions.

Subscripts

eff	refers to the effective properties.
sca	scattering.
abs	absorption.
ext	extinction.

Superscripts

M	refers to Mie theory.
FF	refers to far field approximation.
NF	refers to near field approximation.
NE	refers to the non-exponential decay model (Ref.[41]).

1. INTRODUCTION

Light and radiation transfer in *non-absorbing* media containing particles has long been a subject of study. Applications range from combustion systems and packed or fluidized beds to atmospheric science and astronomy. In all these cases, the conventional Mie theory[1] which ignores the absorption index of the continuous phase is used. However, when these scatterers are embedded in *semitransparent* media the conventional Mie theory is no longer valid.

Radiation transfer through semitransparent media containing bubbles or particles is of interest to many practical engineering applications ranging from remote sensing of the ocean surface and fire fighting to materials processing and colloidal systems in liquids or in the atmosphere. For example, thermal emission data from the ocean surface is used to retrieve wind speed and direction assuming a smoothly varying surface profile.[2,3] However, under high wind conditions, the presence of breaking waves, foam patches, and bubbles affect the emissivity of the ocean surface which can lead to errors in the retrieval of the wind speed and directions. Moreover, the cost and quality of nearly all commercial glass products are determined by the performance of the glass melting and delivery systems which strongly depend on thermal radiation transfer through glass foam layer covering part of the molten glass.[4] Light scattering by bubbles has also been used as a mean to non-invasively monitor the bubble dynamics in sonoluminescence.[5,6,7,8] Finally, the performance of bubble sparged photobioreactors can be strongly affected by light scattering and/or absorption by the bubbles and the bacteria or algae.[9]

Radiation transfer in heterogeneous medium containing bubbles or particles can be divided in four different regimes whether one considers a single scatterer or an ensemble of scatterers and whether the matrix is non-absorbing or absorbing at the wavelength of interest. A detailed discussion of each regime is provided in the following sections. Special emphasis is given to absorbing medium containing bubbles but unless otherwise mentioned the results can be applied to absorbing spherical particles.

2. CURRENT STATE OF KNOWLEDGE

2.A. Single Scatterer and EM Wave Theory

2.A.1. Mie Theory

Mie theory[1] describes the absorption and scattering of radiation by a single spherical particle surrounded by a *non-absorbing* medium with arbitrary index of refraction n. Then, the absorption and scattering cross-sections or efficiency factors of a particle of radius a for radiation with wavelength λ depend on (i) the size parameter $x = 2\pi a/\lambda$ and on (ii) the complex index of refraction of the particle $m' = n' - ik'$ and (iii) of the non-absorbing surrounding medium $m = n$. The efficiency factors of scattering $Q_{sca}^M(a)$, absorption $Q_{abs}^M(a)$, and extinction $Q_{ext}^M(a)$ are expressed as[1,10]

$$Q_{sca}^M(a) = \frac{2}{n^2 x^2} \sum_{j=1}^{\infty} (2j+1)(|a_j|^2 + |b_j|^2), \tag{1}$$

$$Q_{ext}^M(a) = \frac{2}{n^2 x^2} \sum_{j=1}^{\infty} (2j+1) Re(a_j + b_j), \tag{2}$$

$$Q_{abs}^M(a) = Q_{ext}^M(a) - Q_{sca}^M(a). \tag{3}$$

Here, Re refers to the real part of the complex number while the superscript M refers to the Mie theory. The Mie coefficients a_j and b_j are expressed as[11]

$$a_j = \frac{m'\varphi_j'(nx)\varphi_j(m'x) - n\varphi_j(nx)\varphi_j'(m'x)}{m'\zeta_j'(nx)\varphi_j(m'x) - n\zeta_j(nx)\varphi_j'(m'x)}, \tag{4}$$

$$b_j = \frac{m'\varphi_j(nx)\varphi_j'(m'x) - n\varphi_j'(nx)\varphi_j(m'x)}{m'\zeta_j(nx)\varphi_j'(m'x) - n\zeta_j'(nx)\varphi_j(m'x)}, \tag{5}$$

where $\zeta(\rho)$, $\varphi(\rho)$, $\zeta'(\rho)$, and $\varphi'(\rho)$ are the Riccati-Bessel functions and their derivatives with respect to the argument ρ. Because the conventional Mie theory is valid only for a spherical particle embedded in a *non-absorbing* medium, attempts were made to expand the theory to *absorbing* matrix based on either the far-field approximation[12,13,14,15] or the near-field approximation.[16,17,18,19]

2.A.2. Far-Field Approximation

The far-field approximation is based on the asymptotic form of the electromagnetic (EM) field in the radiation zone far from the scatterer. Mundy *et al.*[12] obtained the particle's efficiency factors by integrating the radiative fluxes over a large sphere whose radius r is much larger than the particle radius a(i.e., $r \gg a$) and whose center coincides with that of the particle. Thus, the integrating sphere includes both the particle and the absorbing medium. The author showed that the formulae of the Mie theory [Equation (1) to (5)] and the associated computer program must be adapted for particles in a refracting and absorbing medium having an arbitrary complex index of refraction $m = n - ik$.[12] More precisely, the following changes must be made to the above equations,

(i) the variables m' must be replaced by the complex quantities $\tilde{m} = m'/m$.

(ii) the variables nx must be replaced by the complex quantities $\tilde{x} = mx$.

(iii) the coefficient $2/(n^2 x^2)$ in Equations (1) and (2) must be replaced by the coefficient

$$C^{FF} = \frac{4k^2 \exp[-2kx(r/a)]}{(n^2 + k^2)[1 + (2kx - 1)\exp(2kx)]}. \tag{6}$$

where the superscript FF refers to the far-field approximation. Thus, the scattering, absorption, and extinction efficiency factors are functions of the sphere radius r. They do not represent the efficiency factors of the particle alone.[17] Indeed, when the host medium is absorbing, the scattered wave has not only been attenuated in magnitude but it has also been modulated as it reaches the radiation zone.[20] Thus, for an observer in the radiation zone, the particle's inherent efficiency factors are coupled with the absorption by the medium in an inseparable manner. Note also that under certain conditions, the extinction efficiency factor can be smaller than the scattering efficiency factor. Thus, if $Q_{abs}^{FF}(a)$ can be defined as $Q_{abs}^{FF}(a) = Q_{ext}^{FF}(a) - Q_{sca}^{FF}(a)$ a negative absorption efficiency factor can be obtained.

Mundy et al.[12] also defined the so-called "unattenuated" scattering and extinction efficiency factors for a sphere in an absorbing medium by setting $r = a$ in Equation (6) making

the coefficient C independent of r and equal to

$$C^{FF} = \frac{4k^2 \exp(-2kx)}{(n^2 + k^2)[1 + (2kx - 1)\exp(2kx)]}. \tag{7}$$

On the other hand, when both $kx \ll 1$ and $k \ll n$, Equation (6) simplifies to[21]

$$C^{FF} = \frac{2}{n^2 x^2}. \tag{8}$$

Then, under these conditions, the coefficient C^{FF} is also independent of the radius r.

Alternatively, the inherent scattering and absorption properties of the particle can be calculated when the local Poynting vector is integrated at the scattering particle's surface using the so-called near-field approximation.[16, 17, 18, 19]

2.A.3. Near-Field Approximation

This approach is based on the information of the EM field at the particle surface. Fu and Sun,[17] Sudiarta and Chylek,[18,19] and Lebedev[16] derived analytical expressions for the efficiency factors of absorbing spherical particle in an absorbing medium,[17]

$$Q_{sca}^{NF}(a) = \frac{8\pi k^2}{\lambda n[1 + (2kx - 1)\exp(2kx)]} \sum_{j=1}^{\infty} (2j+1)Im(B_j), \tag{9}$$

$$Q_{abs}^{NF}(a) = \frac{8\pi k^2}{\lambda n[1 + (2kx - 1)\exp(2kx)]} \sum_{j=1}^{\infty} (2j+1)Im(A_j), \tag{10}$$

$$Q_{ext}^{NF}(a) = Q_{abs}^{NF}(a) + Q_{sca}^{NF}(a). \tag{11}$$

where Im refers to the imaginary part of a complex value and the superscript NF refers to the near-field approximation. The complex coefficients A_j and B_j are expressed as[17]

$$A_j = \frac{|c_j'|^2 \varphi_j(m'x)\varphi_j'^*(m'x) - |d_j'|^2 \varphi_j'(m'x)\varphi_j^*(m'x)}{2\pi m'/\lambda}, \tag{12}$$

$$B_j = \frac{|a_j'|^2 \zeta_j'(mx)\zeta_j^*(mx) - |b_j'|^2 \zeta_j(mx)\zeta_j'^*(mx)}{2\pi m/\lambda}. \tag{13}$$

Here, the asterisk denotes the complex conjugate and the coefficients a_j', b_j', c_j' and d_j' are expressed as[17]

$$a_j' = \frac{m'\varphi_j'(mx)\varphi_j(m'x) - m\varphi_j(mx)\varphi_j'(m'x)}{m'\zeta_j'(mx)\varphi_j(m'x) - m\zeta_j(mx)\varphi_j'(m'x)}, \tag{14}$$

$$b'_j = \frac{m'\varphi_j(mx)\varphi'_j(m'x) - m\varphi'_j(mx)\varphi_j(m'x)}{m'\zeta_j(mx)\varphi'_j(m'x) - m\zeta'_j(mx)\varphi_j(m'x)}, \tag{15}$$

$$c'_j = \frac{m'\zeta_j(mx)\varphi'_j(mx) - m'\zeta'_j(mx)\varphi_j(mx)}{m'\zeta_j(mx)\varphi'_j(m'x) - m\zeta'_j(mx)\varphi_j(m'x)}, \tag{16}$$

$$d'_j = \frac{m'\zeta'_j(mx)\varphi_j(mx) - m'\zeta_j(mx)\varphi'_j(mx)}{m'\zeta'_j(mx)\varphi_j(m'x) - m\zeta_j(mx)\varphi'_j(m'x)}. \tag{17}$$

Note that when the matrix is non-absorbing, i.e., $m = n$, the above defined coefficients a'_j and b'_j are identical to a_j and b_j defined in Equations (4) to (5) for the conventional Mie theory.

Therefore, this approach eliminates the ambiguity in the definition of the extinction efficiency factor since the formulae depend only on the complex refraction indices and on the particle radius. The absorption, scattering, and extinction efficiency factors derived from the near-field approximation have been called "inherent" efficiency factors.[20] The adjectives "inherent", "true"[20] or "actual"[17] have been used interchangeably.

Finally, studies based on the near-field approximation[17,18,19] showed that, in the limiting case of spheres much larger than the wavelength of radiation and embedded in an absorbing host medium, the spectral extinction efficiency factor $Q^{NF}_{ext}(a)$ approaches unity as diffraction can be neglected.[19] These results contrast with the case of large spheres in a non-absorbing matrix where the Mie theory predicts that $Q^M_{ext}(a)$ approaches 2.[18,11] In addition, the scattering efficiency factor $Q^{NF}_{sca}(a)$ of a large sphere in an absorbing medium approaches the reflectivity of the flat interface at normal incidence.[18] The convergence to these asymptotic limits was found to be much faster for strongly absorbing matrices than for weakly absorbing ones.[17,18]

2.B. Multiple Scatterers and Radiation Transfer

Several studies have been concerned with photon transport in *non-absorbing* media containing an *ensemble* of bubbles. Common approaches include: (1) the diffusion approximation, (2) the Monte Carlo method, and (3) the radiation transfer equation (RTE).

First, radiation transfer has often been treated as a diffusion process accounting for multiple scattering events.[22,23,24,25,26] Durian and co-workers[27,23,24,25,22] performed experimental,

theoretical, and numerical studies on the angular distribution of the diffusely transmitted and back-scattered light through various highly scattering media of thickness much larger than the photon transport mean free path. The authors found very good agreement between experimental data, the diffusion model, and random walk simulations. The diffusion approximation has also been used to simulate *transient* radiation transport in a non-absorbing foam layer.[26,28]

Moreover, when the diffusion approximation is not valid and both diffraction and interferences can be neglected then, photons can be treated as particles and Monte-Carlo simulations can be performed. For example, Wong and Mengüç[29] simulated depolarization of a collimated and polarized light through non-absorbing foams consisting of large spherical bubbles using a combined Monte Carlo/ray tracing approach as means to characterize the foam morphology. Finally, Tancrez and Taine[30] simulated radiation transfer in porous media consisting of overlapping (i) opaque particles embedded in a transparent fluid (e.g., packed beds) or (ii) transparent spheres in an opaque solid (e.g., open-cell foams) using Monte Carlo simulations. The authors proposed correlations for the effective radiation characteristics of such media.

An alternative approach consists of treating heterogeneous media as homogeneous and solving the RTE using some effective radiation characteristics. The latter can be modeled based on first principle and/or measured experimentally. Fedorov and Viskanta[31,32] proposed a model for the effective radiation characteristics of porous media with various bubble size distributions and porosities and solved the RTE to obtain the transmittance and reflectance of a layer of glass foams. The analysis was performed for bubbles much larger than the wavelength of radiation in the limiting case of anomalous diffraction.[10] Their model for the radiation characteristics was discussed in details by Pilon and Viskanta[33] for various porosities and bubble sizes. In brief, the following models for the effective absorption coefficient was proposed,[31]

$$\kappa_{eff} = \kappa - \pi \int\limits_0^\infty [Q_{abs,m}^M(a) - Q_{abs,m'}^M(a)] a^2 f_1(a) da, \tag{18}$$

where κ and κ_{eff} are the absorption coefficients of the matrix and of the two-phase medium, respectively. The bubble size distribution is denoted by $f_1(a)$ and defined as the number of

bubbles per unit volume having radius between a and $a + da$. The efficiency factors $Q_{abs,m}^M(a)$ and $Q_{abs,m'}^M(a)$ are computed for a sphere of continuous phase (m) or dispersed phase (m'), respectively. They are estimated using the asymptotic formulae (Ref.[34] p.35) for anomalous diffraction derived from the Mie theory for a sphere of radius a embedded in *vacuum*. On the other hand, the scattering coefficient and the scattering phase function were modeled following the conventional expressions used for particulate media with a non-participating matrix,[11,35]

$$\sigma_{eff} = \pi \int_0^\infty Q_{sca}^M(a) a^2 f_1(a) da, \qquad (19)$$

$$\Phi_{eff}(\Theta) = \frac{\pi}{\sigma_{eff}} \int_0^\infty Q_{sca}^M(a) \phi(a, \Theta) a^2 f_1(a) da, \qquad (20)$$

where ϕ and Φ_{eff} refer to the scattering phase functions of a single and of an ensemble of scatterers, respectively. The angle between the incident and scattered radiations is denoted by Θ. Here also, $Q_{sca}^M(a)$ is calculated based on the anomalous diffraction approximation.

More recently, Dombrovsky[21] questioned the validity of the above model on the basis that Equation (18) had not been validated and that $Q_{sca}^M(a)$ was estimated using the complex index of refraction of the dispersed phase m' instead of the ratio m'/m. To address this issue, Dombrovsky[21] suggested using the following model for the effective absorption and scattering coefficients using the far-field efficiency factors,

$$\kappa_{eff} = \kappa + \pi \int_0^\infty Q_{abs}^{FF}(a) a^2 f_1(a) da, \qquad (21)$$

$$\sigma_{eff} = \pi \int_0^\infty Q_{sca}^{FF}(a) a^2 f_1(a) da. \qquad (22)$$

Moreover, the second term on the right-hand side of Equation (21) is "an additional absorption of radiation by particle" that should be positive for particles absorbing more than the matrix (i.e., $k' > k$) and negative in the contrary (e.g., bubbles).[21] Therefore, the presence of the bubbles embedded in a semitransparent matrix reduces the effective absorption coefficient of the medium, i.e., $\kappa_{eff} \leq \kappa$.

Finally, two practical questions remain unanswered and are addressed in this manuscript:

(1) among all the above mentioned theory, which one should be used to estimate the efficiency factors of a spherical scatterer in an absorbing media? and (2) what would be the expressions of the associated radiation characteristics needed to solve the RTE? The present study aims first at determining the conditions under which the absorption by the surrounding medium must be accounted for in the calculation of the efficiency factors by comparing results from the Mie theory, the far-field, and the near-field approximations for specific absorbing media and particle/bubble size parameters. Then, it relates the far-field and near-field approximations for a single particle to models for the effective radiation characteristics required for solving the radiative transfer in an ensemble of scatterers embedded in an absorbing medium.

3. ANALYSIS

The assumptions used in this study include (1) all particles or bubbles are spherical, (2) the scattering behavior of a single particle or bubble is not affected by the presence of its neighbors (independent scattering),[36] (3) the radiation field within the continuous phase is incoherent (i.e., scattering centers are randomly distributed with zero-phase correlation), and(4) each phase is homogeneous and has uniform optical properties. Practically the assumption of independent scattering by wavelength-sized and larger particles is satisfied when the particles are randomly positioned and separated by distances larger than 4 times their radius.[37,38]

3.A.   *Difference Between Far-Field and Near-Field Approximations*

This section compares the results for the different efficiency factors obtained by (1) the Mie theory, (2) the far-field, and (3) the near-field approximations. Results for the Mie theory were computed based on the code provided by Bohren and Hoffman.[39] The same code was adapted for the far-field approximation following the suggestions by Mundy *et al.*[12] reviewed previously and using $r = a$. The code was successfully validated against the efficiency factors reported by Mundy *et al.*[12] and by Dombrovsky.[21] Similarly, the code for the near-field approximation was kindly provided by W. Sun and was validated against Fu and Sun's

results.[17] Here, the same situations as those explored by Fu and Sun[17] are investigated. In all cases, the series in Equations (1), (2), (9), and (10) are truncated and terminated when the summation index j is equal to the integer closest to $x + 4x^{1/3} + 2$.[39]

Figure 1 shows the scattering, extinction, and absorption efficiency factors as functions of the size parameter x for a non-absorbing bubble ($m' = 1$) embedded in an absorbing medium of refractive index $m = 1.34 - ik$ with k equals to 0, 0.001, 0.01, and 0.05. First, for non-absorbing matrix ($k = 0$), the Mie theory, the far-field, and near-field approximations give identical results with $Q_{abs}(a) = 0$ and $Q_{sca}(a) = Q_{ext}(a)$. In addition, the extinction efficiency factor converges to 2 as the size parameter tends to infinity corresponding to the well know diffraction paradox.[11]

Moreover, Figure 1 indicates that for bubbles in an absorbing matrix, the far-field efficiency factors are always smaller than their near-field counterparts and the difference increases as the matrix absorption index k increases. Then, $Q_{abs}^{NF}(a)$ is equal to zero while $Q_{abs}^{FF}(a)$ is negative for all values of k and, as a result, $Q_{ext}^{FF}(a)$ is smaller than $Q_{sca}^{FF}(a)$ and sometimes even negative. Using either approximation, both $Q_{sca}(a)$ and $Q_{ext}(a)$ decrease as k increases. In addition, as the size parameter tends to infinity, both $Q_{sca}^{NF}(a)$ and $Q_{ext}^{NF}(a)$ converge to 1. On the contrary, $Q_{sca}^{FF}(a)$ and $Q_{ext}^{FF}(a)$ converge to 0.5 and 0, respectively. Finally, as k increases, the asymptotic values are reached for smaller size parameters.

The same comparison was performed for absorbing particles. Figure 2 shows the scattering, extinction, and absorption efficiency factors as functions of size parameter for an absorbing particle having $m' = 1.34 - 0.01i$ embedded in an absorbing medium such that $m = 1.0 - ik$. Similarly, Figure 3 shows results for different particle and matrix featuring $m' = 1.4 - 0.05i$ and $m = 1.2 - ik$. In both cases, k takes the values of 0.0, 0.001, 0.01, and 0.05. The same conclusions as above can be drawn except for the scattering efficiency factors. Indeed, as the size parameter tends to infinity, $Q_{sca}^{NF}(a)$ and $Q_{sca}^{FF}(a)$ converge to 1 for an absorbing particle in a non-absorbing matrix while they both converge to zero when the matrix is absorbing. In addition, one can note that the efficiency factors are always positive for the near-field approximation. On the contrary, the absorption and extinction efficiency factors obtained by

the far-field approximation can be negative if k is larger than k' as shown in Figures 1 and 2 when $k = 0.05$ and $k' = 0.01$. Note also that $Q_{abs}^{NF}(a)$ is found to be nearly independent of the matrix absorption index k.

Furthermore, the relative differences between the far-field and near-field approximations for the efficiency factors are shown in Figure 4. It indicates that the relative difference increases as the absorption index k increases. For a weakly absorbing matrix ($k < 0.001$) and $1 < x < 100$, the predictions of the scattering efficiency factor from the far-field approximation fall within 10% of that from the near-field approximation under the conditions tested. However, for small size parameters ($x < 1$), the relative difference in the scattering efficiency factor can be significant. Similar trend was observed by Yang and co-workers (see Fig. 3 in Ref.[20]). This can be attributed to the fact that (1) the scattering efficiency factor is small (less than 0.1 for $x < 1$) and therefore sensitive to numerical uncertainty and how the summations are performed and/or (2) the computation of the Riccati-Bessel functions by forward recurrence is unstable.[39] In addition, when the absorption index of the medium is larger than that of particles, the extinction and absorption efficiency factors predicted by the far-field approximation can be negative while those predicted by the near-field approximation are always greater than zero. Note that (i) the relative differences in the extinction and absorption efficiency factors can be larger than 100% when $Q_{abs}^{FF}(a)$ and $Q_{ext}^{FF}(a)$ are negative and (ii) the relative differences of the absorption efficiency factor for bubbles ($m' = 1.0$) are always unity since $Q_{abs}^{NF}(a)$ is always zero.

Finally, the relative differences between the Mie theory and the near-field approximation for the efficiency factors are shown in Figure 5. It indicates that the relative difference in the absorption efficiency factors between the Mie theory and the near-field approximation is relatively small and less than 16%. However, there are large relative differences in the scattering and extinction efficiency factors for matrix with large absorption index and/or for large size parameters. Since the efficiency factors predicted by the far-field approximation are always smaller than those predicted by the near-field approximation and sometimes can be negative, the relative differences between the Mie theory and the far-field approximation

are much larger than those shown in Figure 5. Thus, one can see that the Mie theory deviates significantly from the near-field and far-field approximations for matrix with large absorption index and/or for large size parameter. Under these conditions, the matrix absorption index cannot be ignored in computing the efficiency factors. On the other hand, for small values of x, the large relative difference is due to numerical error but unimportant for all practical purposes.

3.B.   *Application To Radiation Transfer*

One of the main motivations in determining single particle efficiency factors is for radiative transfer calculations which require both the cross sections and an accurate description of the phase matrix. Moreover, predicting radiation transfer through heterogeneous media requires the efficiency factors of the particle in the far field.[40,20] Thus, the inherent scattering efficiency factor obtained from the near-field approximation by considering the EM field at the particle surface cannot be used for modeling the effective scattering coefficient.[20] Indeed, it does not have the conventional meanings in that the corresponding cross sections are not simply the products of these factors and the projected area of the particle.[20]

Moreover, Fu and Sun[17] derived the scattering, absorption, and extinction efficiency factors based on the near-field approximation while they obtained the scattering phase function using the far-field approximation. This approach appears to be conceptually inconsistent. To address this inconsistency, Yang *et al.*[20] used (i) the unattenuated scattering efficiency factor $Q_{sca}^{FF}(a)$ using Equation (7), (ii) the near-field inherent absorption efficiency factor $Q_{abs}^{NF}(a)$ since it represents absorption by the particle alone, and (iii) the apparent extinction efficiency factor defined as $Q_{ext}(a) = Q_{sca}^{FF}(a) + Q_{abs}^{NF}(a)$. Then, $Q_{ext}(a)$ is larger than the scattering efficiency factor $Q_{sca}^{FF}(a)$ since $Q_{abs}^{NF}(a)$ is always non-negative. This definition is consistent with the scattering phase function and asymmetry factor derived by Fu and Sun[17] based on the far-field scattered waves. It also overcomes the shortcoming of the far-field approximation where $Q_{ext}^{FF}(a)$ and/or $Q_{abs}^{FF}(a)$ could be negative.[12,13,14,15]

Recently, Fu and Sun[41] extended this approach by suggesting that an apparent absorption

efficiency factor needs to be introduced to take into account the non-exponential decay of the near-field scattered radiation in the absorbing matrix. This approach is referred by the superscript NE. The non-exponential absorption can be quantified by the difference between the actual and apparent scattering efficiency factors. Thus, they defined an apparent absorption efficiency factor given by,[41]

$$Q_{abs}^{NE} = Q_{abs}^{NF} + (Q_{sca}^{NF} - Q_{sca}^{FF}). \tag{23}$$

The apparent extinction efficiency factor is then $Q_{ext}^{NE} = Q_{abs}^{NE} + Q_{sca}^{FF} = Q_{ext}^{NF}$. Thus, the extinction of incident radiation remains the same as Q_{ext}^{NF} defined by Fu and Sun.[17]

Consequently, the unattenuated (i.e., $r = a$) far-field scattering efficiency factor and the far-field phase function seem to be the preferred approach for radiation transfer calculations. However, there are three alternatives for the apparent absorption efficiency factor namely, (i) the unattenuated absorption efficiency factor $Q_{abs}^{FF}(a)$ defined by Mundy *et al.*[12] with the constant C^{FF} given by Equation (7), (ii) the near-field absorption efficiency factor $Q_{abs}^{NF}(a)$ given by Equation (10), and (iii) the absorption efficiency factor $Q_{abs}^{NE}(a)$ given by Equation (23).

The absorption efficiency factor to be used to estimate the effective absorption coefficient will depend on the model selected [e.g., Equations (18) or (21)]. In Fedorov and Viskanta's model,[31] $Q_{abs}^{M}(a)$ and $Q_{abs,m}^{M}(a)$ were calculated using the Mie theory in the anomalous diffraction limit. This should be reconsidered and among the above three alternatives, $Q_{abs}^{NF}(a)$ should be used since it is always positive and nearly independent of the absorption of the medium. Thus, when the absorption index of the medium is greater than that of the scatterer (i.e., $k > k'$) then, $Q_{abs,m}^{NF}(a) > Q_{abs}^{NF}(a)$ and $\kappa_{eff} < \kappa$ and vice-versa. Moreover, in Dombrovsky's model,[21] the absorption efficiency factor is calculated using $Q_{abs}^{FF}(a)$[21] as it depends on the medium properties and can be negative. Thus, when the absorption index of the medium is greater than that of the scatterer, $Q_{abs}^{FF}(a) < 0$ and $\kappa_{eff} < \kappa$ otherwise $Q_{abs}^{FF}(a) > 0$ and $\kappa_{eff} > \kappa$.

Finally, $Q_{abs}^{NE}(a)$ is always positive since $Q_{sca}^{NF} - Q_{sca}^{FF}(a) \geq 0$ and $Q_{abs}^{NF}(a) \geq 0$ even for bubbles. Thus, it cannot be used in combination with Equation (21). In addition, the term

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$Q_{abs,m}^{NE}(a) - Q_{abs,m'}^{NE}(a)$ can be negative or positive depending not only on the absorption index of both phases but also on the difference in scattering efficiency factors. For example, it could be negative for bubbles and thus, appears also incompatible with Equation (18). Then, a new model for κ_{eff} conceptually compatible with the definition of $Q_{abs}^{NE}(a)$ should be developed.

3.C.   Application: Radiation Characteristics of Water Containing Bubbles

This section discusses the effective radiation characteristics of water containing bubbles predicted by the above models. The complex index of refraction of air bubbles is equal to unity ($m' = 1$). The refractive and absorption indices of water n and k over the spectral range from 0.2 to 200 μm are given in the literature.[42]

3.C.1.   Effective Scattering Coefficient

First, note that the expression for the effective scattering coefficient proposed by Fedorov and Viskanta[31] [Equations (19)] and by Dombrovsky[21] [Equation (22)] differ only by the choice of the model for the scattering efficiency factor. As previously discussed, $Q_{sca}^{FF}(a)$ should be used. To simplify the problem, we further assume that the air bubbles have the same radius. Thus, the effective scattering coefficient simplifies as,

$$\sigma_{eff} = \frac{3f_v}{4a}Q_{sca}^{FF}(a). \tag{24}$$

Figure 6 compares the effective scattering coefficient predicted by Equation (24) (solid line) with that predicted by the Mie theory (assuming $k = 0$) (dotted line) as a function of wavelength for different void fractions and bubble radii. For given wavelength and bubble radius, the effective scattering coefficient increases with increasing void fraction. For large wavelengths ($> 7\mu$m), the effective scattering coefficient increases with increasing bubble diameter. In addition, the relative error between these two approaches is independent of void fraction. For wavelengths smaller than 2 μm, the relative error is less than 10% while it can be larger than 50% for wavelengths around 3, 6, 13, and 20 μm corresponding to peaks

in the optical properties n and/or k of water. Thus, for wavelengths beyond 2 μm, neglecting absorption by water can cause large errors on the effective scattering coefficient.

3.C.2. Effective Absorption Coefficient

Moreover, based on the two different models for the effective absorption coefficients proposed by Fedorov and Viskanta[31] and Dombrovsky[21] and different expressions of $Q_{abs}(a)$, the effective absorption coefficient accounting for the absorption by the matrix with monodispersed bubbles can be calculated by two alternate ways,

$$\kappa_{eff,1} = \kappa - \frac{3f_v}{4a}[Q_{abs,m}^{NF}(a) - Q_{abs,m'}^{NF}(a)], \tag{25}$$

$$\kappa_{eff,2} = \kappa + \frac{3f_v}{4a}Q_{abs}^{FF}(a), \tag{26}$$

where $\kappa = 4\pi k/\lambda$ is the absorption coefficient of water. Four different bubble radii, a=0.01, 0.1, 1.0, and 10 μm, and three different void fractions f_v=0.05, 0.4, and 0.74 covering the range from bubbly flow to maximum packing of spheres of uniform size are investigated over the spectral range from 0.2 to 200 μm.

Figure 7 compares the effective absorption coefficient predicted by Equations (25) and (26) for monodispersed bubbles of radius a=0.01 μm for different void fractions. For small void fractions, such as $f_v = 0.05$, the differences between these two models are small and the predicted effective absorption coefficient is close to the absorption coefficient of water. Thus, even though $Q_{abs}^{NF}(a)$ and $Q_{abs}^{FF}(a)$ are significantly different, absorption by the matrix represented by κ dominates the overall absorption of the composite medium. In other words, $\kappa_{eff} \approx \kappa$ for small void fraction and the model chosen for $Q_{abs}(a)$ is unimportant. This was the case of the experimental measurements reported by Baillis and co-workers[43,44] for fused quartz containing bubbles of average radius 0.64 mm and void fraction of 4% in the spectral region from 2.6 to 4 μm where quartz is weakly absorbing ($k < 10^{-4}$) and $\kappa x << 1$.

However, when the void fraction increases the differences become large (Figure 7). Then, the second term on the right-hand side of Equations (25) and (26) dominates. Also, $\kappa_{eff,2}$ is much smaller than $\kappa_{eff,1}$. In addition, $\kappa_{eff,2}$ is negative for wavelengths around 3 and 11

to 200 μm and void fraction of 0.74, which is physically unacceptable. Note that similar results have been found for other bubble radii. Therefore, for large void fractions, the model proposed by Dombrovsky[21] may give unphysical results. On the other hand, $\kappa_{eff,1}$ was found to be always positive even for large void fractions and various bubble radii as illustrated in Figure 8. Thus, the model proposed by Fedorov and Viskanta[31] using $Q_{abs}^{NF}(a)$, tends to give more physically acceptable results.

Moreover, the original model proposed by Fedorov and Viskanta[31] using $Q_{abs}^{M}(a)$ for monodispersed bubbles was expressed as

$$\kappa_{eff,3} = \kappa - \frac{3f_v}{4a}[Q_{abs,m}^{M}(a) - Q_{abs,m'}^{M}(a)]. \tag{27}$$

Considering the relative error between $\kappa_{eff,1}$ and $\kappa_{eff,3}$ defined as $|\kappa_{eff,3} - \kappa_{eff,1}|/\kappa_{eff,3}$ as a function of wavelength for different void fractions and bubble radii establishes that for $f_v = 0.05$ the relative error is less than 2% for all wavelengths. Increasing the void fraction results in larger relative errors. For example, for $f_v = 0.4$, the relative error reaches up to 20% at some wavelengths and for $f_v = 0.74$, it may exceed 80% for some combinations of wavelengths and bubble radius. In practice, when the wavelength is less than 1 μm, the effective absorption coefficient is small and does not significantly affect the radiation transfer calculations.

4. CONCLUSIONS

This paper has investigated (i) the efficiency factors of particles and bubbles embedded in an absorbing medium and (ii) the effective radiation characteristics of two-phase mixture consisting of bubbles in an absorbing medium. The efficiency factors predicted by the conventional Mie theory, the far-field, and the near-field approximations were compared. The best approach for the radiation characteristics to be used in the radiation transfer equation was clarified and the following conclusions can be drawn:

1. Ignoring the absorption index of the matrix can results in significant error on the scattering and extinction efficiency factors predicted by the conventional Mie theory

except when the host medium is non-absorbing or weakly absorbing. This is particularly true if the size parameter is large. Then, the near-field or far-field approximations offer alternatives that should be used.

2. The efficiency factors for a spherical particle predicted by the far-field and near-field approximations can be significantly different. This difference increases as the matrix absorption index increases.

3. The effective scattering coefficient σ_{eff} should be expressed as a function of the far-field scattering efficiency factor $Q_{sca}^{FF}(a)$.

4. The choice of the absorption efficiency factor depends on the model used for estimating the effective absorption coefficient.

5. For small void fractions, absorption by the continuous phase dominates and the choices of the model for absorption coefficient and the associated absorption efficiency factor are unimportant.

6. For large void fractions, the models by Fedorov and Viskanta[31] and by Dombrovsky[21] differ significantly from one another. The model proposed by Fedorov and Viskanta[31] gives physically acceptable results while that by Dombrovsky[21] can yield negative absorption coefficient.

7. For most wavelengths between 0.2 and 200 μm, the absorption index of water can be neglected and the conventional Mie theory for non-absorbing medium can be used. However, at some wavelengths, neglecting the absorption of medium results in large errors in the efficiency factors and in the associated radiation characteristics.

Finally, note that experimental data for medium with large volume fractions of scatterers and/or for matrices with a relatively large absorption index is still needed to validate the above effective radiation characteristic models. Alternatively, the rigorous approach developed by Mishchenko[40] based on the Maxwell's equations and the concept of statistical

electromagnetics could be extended to particles in an absorbing medium and compared with solutions of the RTE combined with one the above effective property models.

ACKNOWLEDGEMENTS

References

1. G. Mie, "Beiträge zur Optik trüber Medien, speziell kolloidaler Metallsungen", *Annalen der Physik*, **25**, 377–445 (1908).

2. P. L. Marston, "Light scattering from bubbles in water", in *Oceans' 89, IEEE Publication No. 89CH2780-5*, New York, NY (1989), IEEE, pp. 1186–1193.

3. X. Zhang, M. Lewis, and B. Johnson, "Influence of bubbles on scattering of light in the ocean", *Applied Optics*, **37** 6525–6536 (1998).

4. A. G. Fedorov and L. Pilon, "Glass foam: formation, transport properties, and heat, mass, and radiation transfer", *Journal of Non-Crystalline Solids*, **311** 154–173 (2002).

5. K.R. Weninger, S.J. Putterman, and B.P. Barber, "Angular correlations in sonoluminescence: Diagnostic for the sphericity of a collapsing bubble", *Physical Review E*, **54** R2205–R2208 (1996).

6. S.Hayashi, S. Uchiyama, and N. Harba, "Double scattering of light by a sonoluminescing bubble and many small particles", *Journal of the Physical Society of Japan*, **70** 3544–3549 (2000).

7. K.R. Weninger, P.G. Evans, and S.J. Putterman, "Comment on "Mie scattering from a sonoluminescing bubble with high spatial and temporal resolution -[Physical Review E 61, 5253 (2000)]"", *Physical Review E*, **64** (2001).

8. C. Villarreal, R. Esquivel-Sirvent, and R. Jauregui, "Mie scattering and the physical mechanisms of sonoluminescence", *Physical Review E*, **61** 403–406 (2000).

9. H. Berberoglu, J. Yin, and L. Pilon, "Simulating light transfer in a bubble sparged photobioreactor for simultaneous hydrogen fuel production and CO_2 mitigation", in *Eurotherm Seminar 72, Computational Thermal Radiation in Participating Media II, Poitiers, France* (April 5-7, 2006), pp. 297–306.

10. H. C. van de Hulst, *Light Scattering by Small Particles*, John Wiley & Sons, New York, NY (1957).

11. M. F. Modest, *Radiative Heat Transfer*, Academic Press, San Diego, CA (2003).

12. W.C.Mundy, J.A. Roux, and A.M. Smith, "Mie scattering in an absorbing medium", *Journal of the Optical Society of America- A*, **64** 1593–1597 (1974).

13. P. Chylek, "Light scattering by small particles in an absorbing medium", *Journal of the Optical Society of America*, **67** 561–563 (1977).

14. C. F. Bohren and D. P. Gilra, "Extinction by a spherical particle in an absorbing

medium", *Journal of Colloid and Interface Science*, **72**, 215–221 (1979).

15. M. Quintem and J. Rostalki, "Lorenz-mie theory for sphere immersed in an absorbinh host medium", *Particle & Particle Systems Characterization*, **13** 89–96 (1996).

16. A.N. Lebedev, M. Gartz, U. Kreibig, and O. Stenzel, "Optical extinction by spherical particles in an absorbing medium: Application to composite absorbing films", *The European Physical Journal D*, **6** 365–373 (1999).

17. Q. Fu and W. Sun, "Mie theory for light scattering by a spherical particle in an absorbing medium", *Applied Optics*, **40**, 1354–1361 (2001).

18. I.W. Sudiatra and P. Chylek, "Mie scattering efficiency of a large spherical particle embedded in an absorbing medium", *Journal of Quantitative Spectroscopy and Radiative Transfer*, **70** 709–714 (2001).

19. I.W. Sudiatra and P. Chylek, "Mie scattering formalism for spherical particles embedded in an absorbing medium", *Journal of the Optical Society of America-A*, **18** 1275–1278 (2001).

20. P.Yang, B.-C. Gao, W.J. Wiscombe, M.I. Mishchenko, S.E. Platnik, H.-L. Huang, B.A. Baum, Y.X. Hu, D.M. Winker, S.-C. Tsay, and S.K. Park, "Inherent and apparent scattering properties of coated or uncoated spheres embedded in an absorbing host medium", *Applied Optics*, **41** 2740–2759 (2002).

21. L.A. Dombrovsky, "The propagation of infrared radiation in a semitransparent liquid containing gas bubbles", *High Temperature*, **42** 143–150 (2004).

22. M. U. Vera, A. Saint-Jalmes, and D. J. Durian, "Scattering optics of foams", *Applied Optics*, **40**, 4210–4214 (2001).

23. D. J. Durian, "The diffusion coefficient depends on absorption", *Optics Letters*, **23** 1502–1504 (1998).

24. M. U. Vera and D. J. Durian, "Angular distribution of diffusely transmitted light", *Physical Review E*, **53**, 3215–3224 (1996).

25. M. U. Vera, P.-A. Lemieux, and D. J. Durian, "Angular distribution of diffusely back-scattered light", *Journal of the Optical Society of America - A*, **14**, 2800–2808 (1997).

26. M. R. Jones, V. P. Solovjov, B. W. Webb, and S. M. Salisbury, "Identification of appropriate source model for accurate diffusion modeling of radiative transfer in a non-absorbing foam layer", in *Proceedings of the 4th International Symposium on Radiation Transfer*, M. P. Mengüç and N. Selçuk, Eds. Begell House, Inc., New York (2004), pp. 143–152.

27. D.J. Durian, "Influence of boundary reflection and refraction on diffusive photon transport", *Physical Review E*, **50** 857–866 (1994).

28. S. M. Salisbury, M. R. Jones, B. W. Webb, and V. P. Solovjov, "Sensitivity analysis of an inverse method for characterizing industrial foams", in *Proceedings of the 2005 ASME Summer Heat Transfer Conference*. ASME, San Francisco, California, USA (2005), pp. HT2005 − 72720.

29. B.T. Wong and M. P. Mengüç, "Depolarization of radiation by non-absorbing foams", *Journal of Quantitative Spectroscopy and Radiative Transfer*, **73** 273–284 (2002).

30. M. Tancrez and J. Taine, "Direct identification of absorption and scattering coefficients and phase function of a porous medium by a Monte Carlo technique", *International Journal of Heat and Mass Transfer*, **47** 373–383 (2004).

31. A. G. Fedorov and R. Viskanta, "Radiative transfer in a semitransparent glass foam blanket", *Physics and Chemistry of Glasses*, **41**, 127–135 (2000).

32. A. G. Fedorov and R. Viskanta, "Radiative characteristics of glass foams", *Journal of the American Ceramic Society*, **83**, 2769–2776 (2000).

33. L. Pilon and R. Viskanta, "Radiation characteristics of glass containing bubbles", *Journal of the American Ceramic Society*, **86**, 1313–1320 (2003).

34. D. Deirmendjian, *Electromagnetic scattering on spherical polydispersions*, Elsevier, New York, NY (1969).

35. R. Siegel and J.R. Howell, *Thermal radiation heat transfer - Third Edition*, Hemisphere Publishing Co., New York, NY (2001).

36. C. L. Tien and B. L. Drolen, "Thermal radiation in particulate media with dependent and independent scattering", in *Annual Review of Numerical Fluid Mechanics and Heat Transfer*, T.C. Chawla, Ed., vol. 1, pp. 1–32. Hemisphere, New York, NY (1987).

37. M. I. Mishchenko, D. W. Mackowski, and L. D. Travis, "Scattering of light by bispheres with touching and separated components", *Applied Optics*, **34** 4589–4599 (1995).

38. M. I. Mishchenko, J. W. Hovenier, and D. W. Mackowski, "Single scattering by a small volume element", *Journal of the Optical Society of America - A*, **34** 71–87 (2004).

39. C. F. Bohren and D. R. Huffman, *Absorption and scattering of light by small particles*, John Wiley and Sons, New York (1983).

40. M. Mishchenko, "Vector radiative transfer equation for arbitrarily shaped and arbitrarily oriented particles: A microphysical derivation from statistical electromagnetics", *Applied Optics*, **41** 7114–7134 (2002).

41. Q. Fu and W.B. Sun, "Apparent optical properties of spherical particles in absorbing medium", Personal communications (2005).

42. G. M. Hale and M. R. Querry, "Optical constants of water in the 200-nm to 200-μm wavelength region", *Applied Optics*, **12** 555–563 (1973).

43. H. Randrianalisoa, D. Baillis, and L. Pilon, "Improved inverse method for determining radiation characteristics of fused quartz containing bubbles", *Journal of Thermophysics and Heat Transfer*, **(in press)** (2006).

44. L.A. Dombrovsky, H. J. Randrianalisoa, D. Baillis, and L. Pilon, "The use of Mie theory in analysis of experimental data on infrared properties of fused quartz containing bubbles", *Applied Optics*, **44**, 7021–7031 (2006).
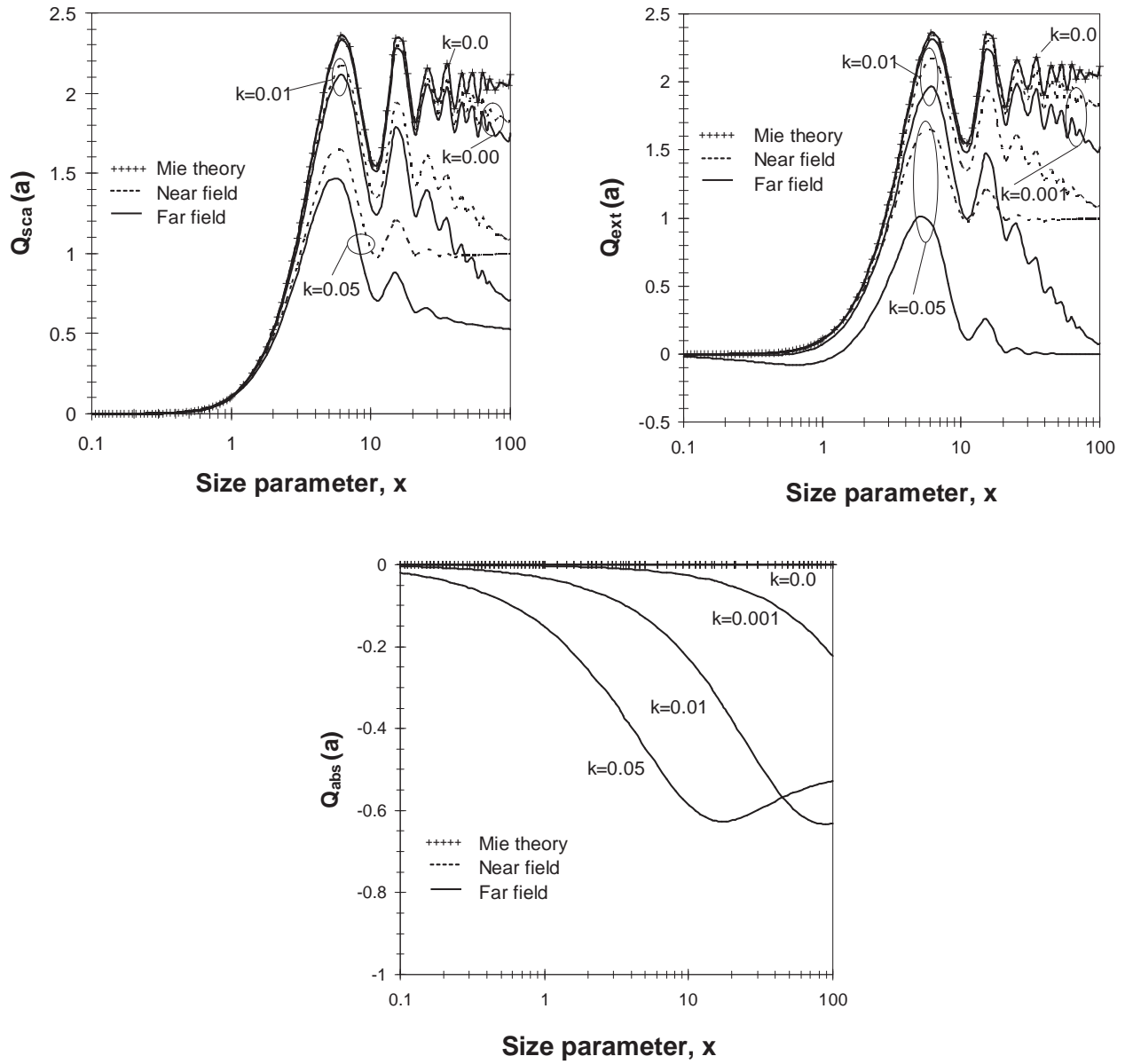
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Fig. 2. Scattering, extinction and absorption efficiency factors as functions of size parameters for an absorbing spherical particle ($m' = 1.34 - 0.01i$) embedded in a medium with refractive index $m = 1.0 - ik$ with $k = 0.0$, 0.001, 0.01, and 0.05.
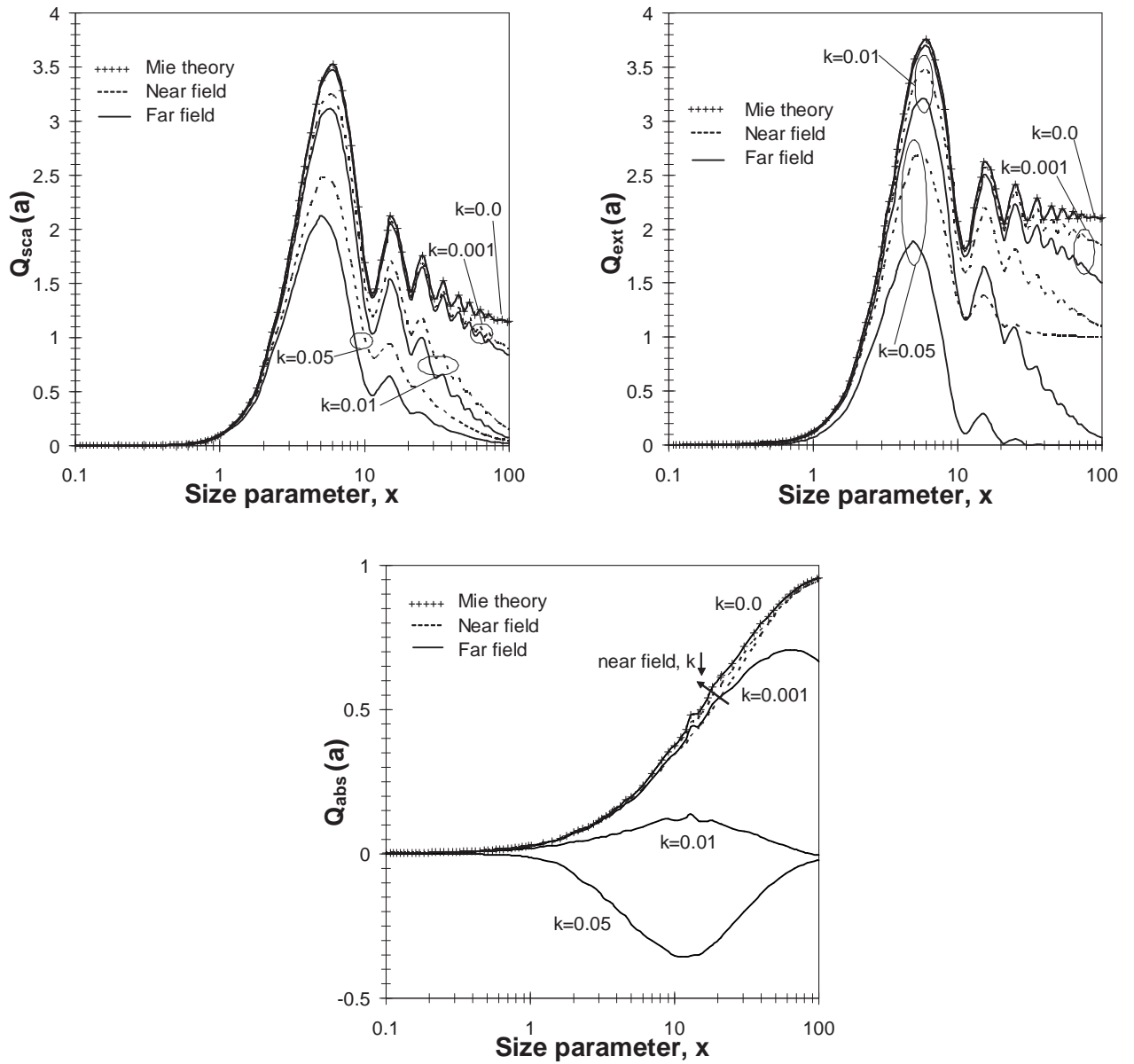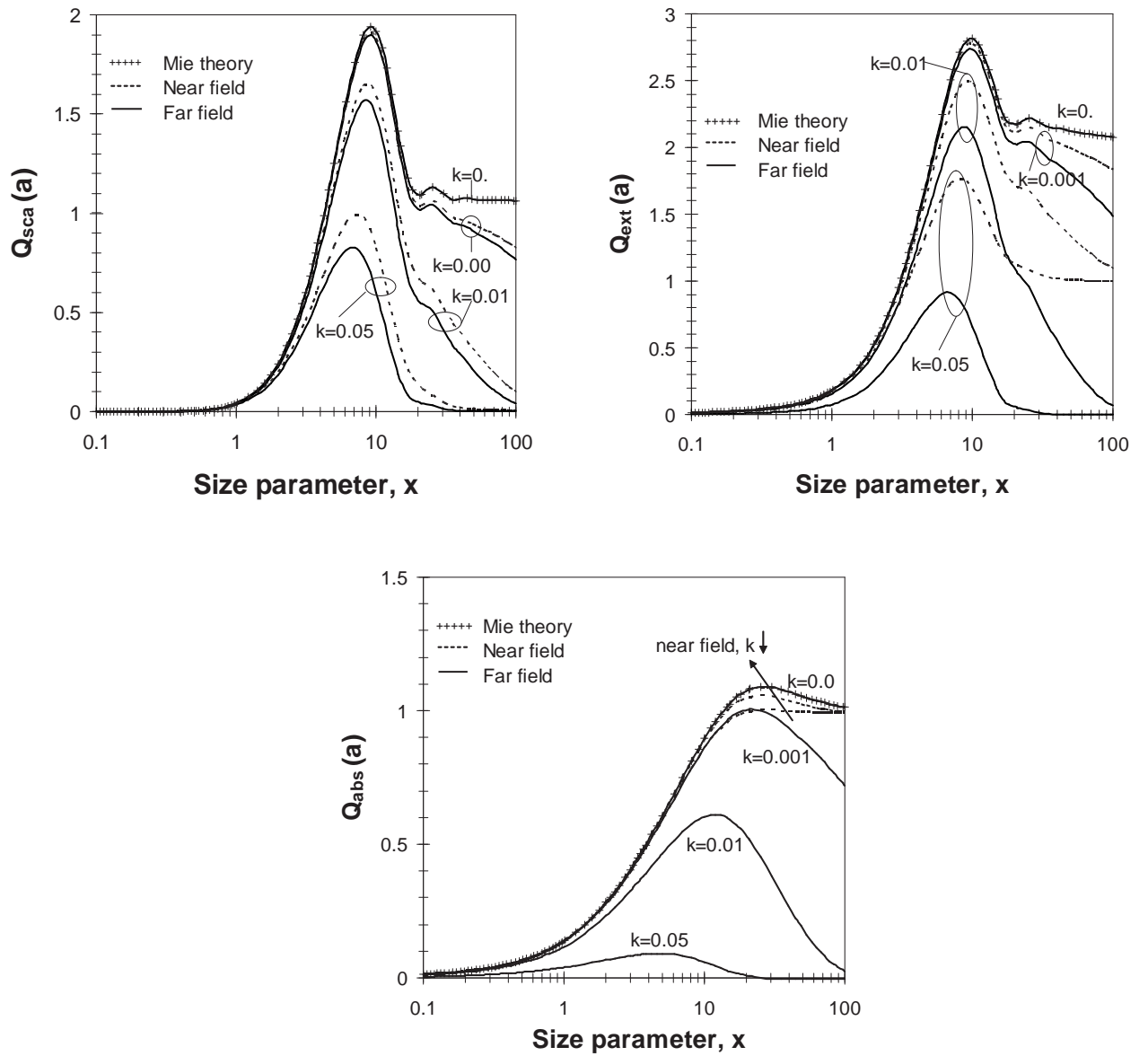
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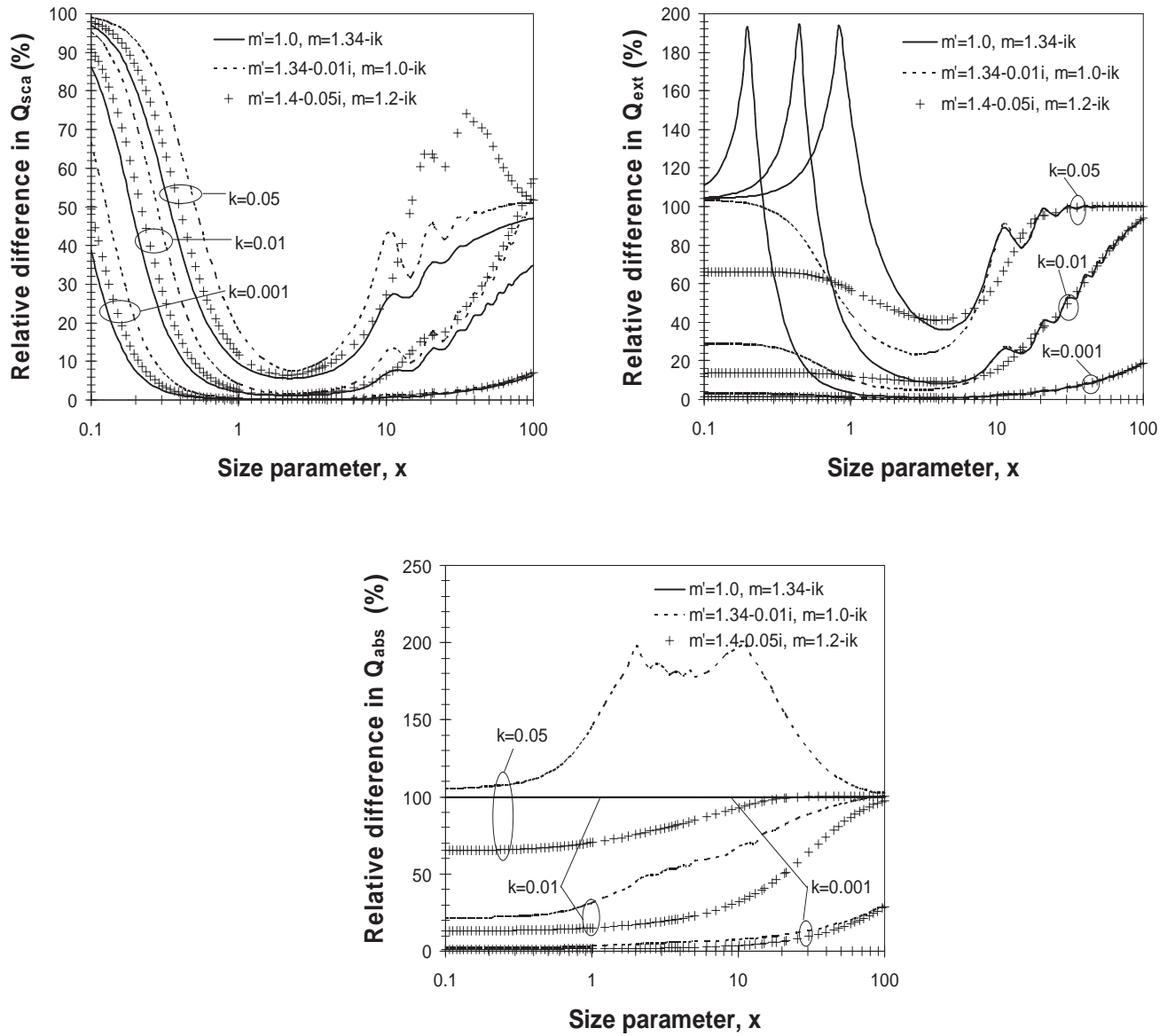
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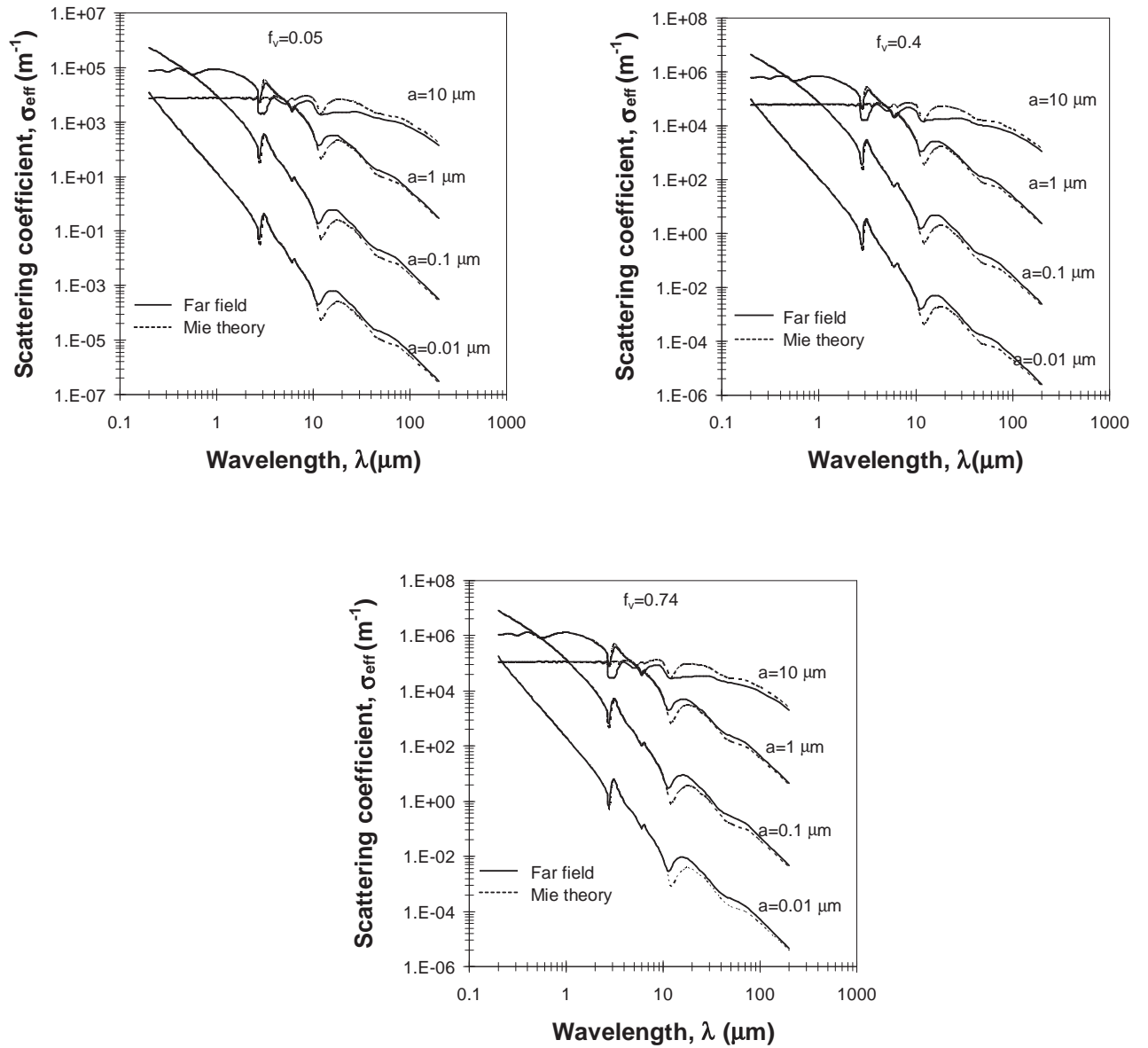
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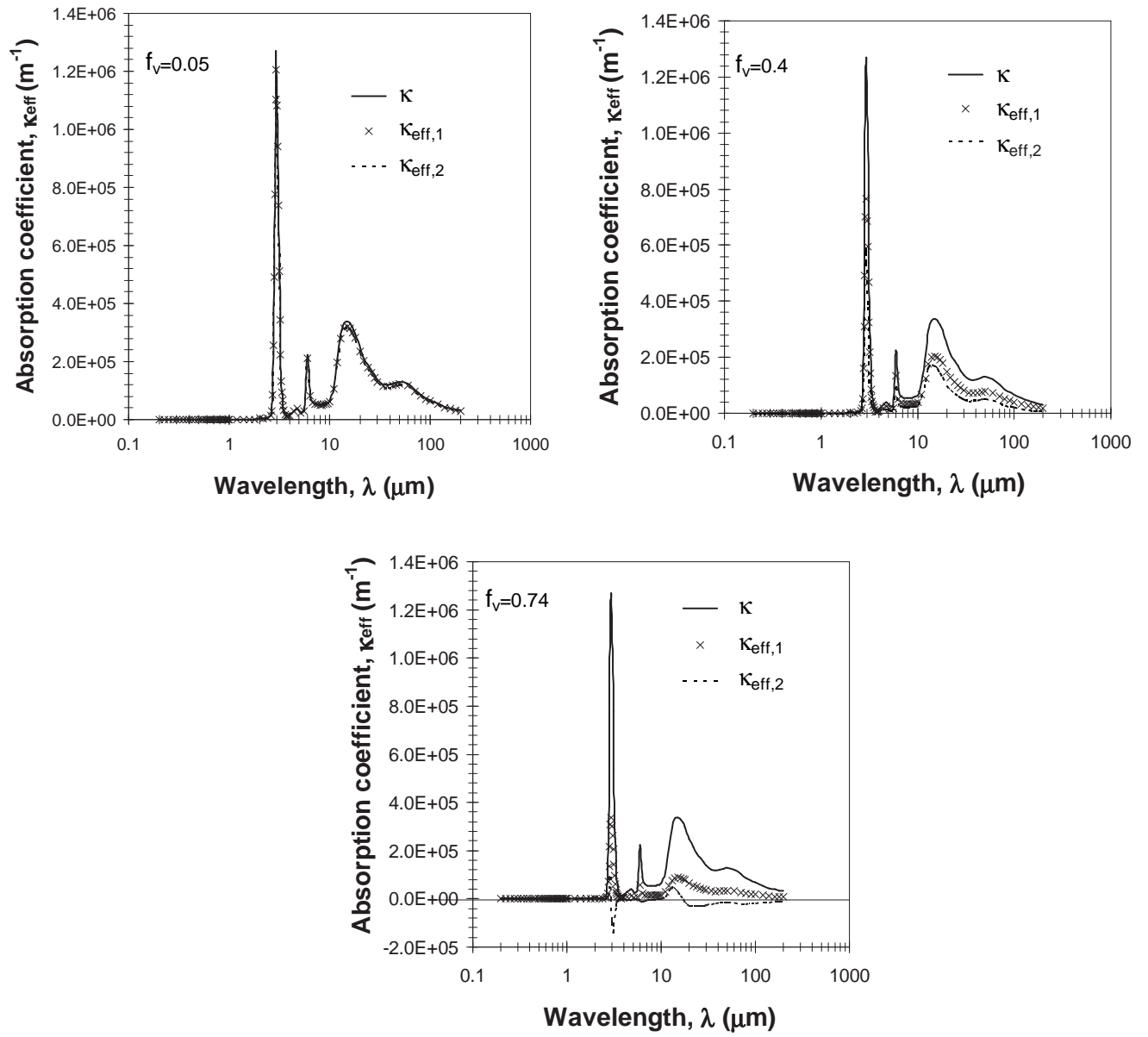
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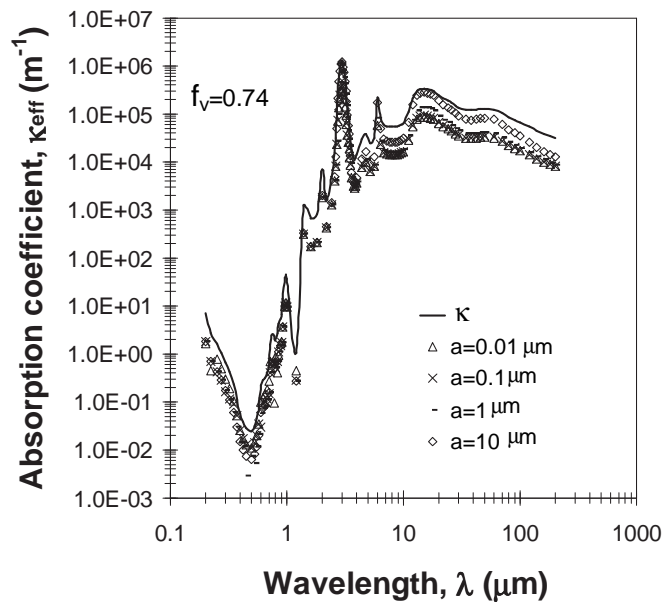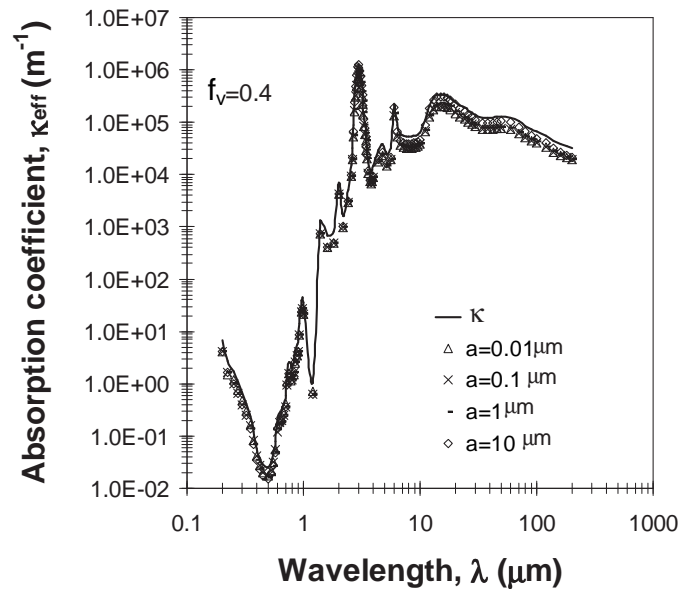
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