Efficiency factors and radiation characteristics of spherical scatterers in an absorbing medium

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The radiative properties of bubbles or particles embedded in an absorbing medium are investigated. We aim first to determine the conditions under which absorption by the surrounding medium must be accounted for in the calculation of the efficiency factors by comparing results from Mie theory and the far-field and near-field approximations. Then, we relate 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 at which absorption by water must be accounted for. © 2006 Optical Society of America

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1. INTRODUCTION

Light and radiation transfer in nonabsorbing 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] that 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 However, when these scatterers are embedded in semitransparent media the conventional Mie theory is no longer valid.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 in 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 are 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 that strongly depend on thermal radiation transfer through the glass foam layer covering part of the molten glass.[4] Light scattering by bubbles has also been used to noninvasively monitor the bubble dynamics in sonoluminescence.[5–8] 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 media containing bubbles or particles can be divided into four different regimes whether one considers a single scatterer or an ensemble of scatterers and whether the matrix is nonabsorbing 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

A. Single Scatterer and Electromagnetic Wave Theory

1. Mie Theory

Mie theory[1] describes the absorption and scattering of radiation by a single spherical particle surrounded by a nonabsorbing 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 $x = 2\pi a/\lambda$, (ii) the complex index of refraction of the particle $m' = n' - ik'$, and (iii) the complex index of refraction of the nonabsorbing surrounding medium $m = n$. The efficiency factors of scattering $Q_{\mathrm{sca}}^{\mathrm{M}}(a)$, absorption $Q_{\mathrm{abs}}^{\mathrm{M}}(a)$, and extinction $Q_{\mathrm{ext}}^{\mathrm{M}}(a)$ are expressed as[1,10]

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

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

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

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 nonabsorbing medium, attempts were made to expand the theory to an absorbing matrix based on either the far-field approximation[12–15] or the near-field approximation.[16–19]

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 formulas of the Mie theory [Eqs. (1)–(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 Eqs. (1)–(5):

(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 Eqs. (1) and (2) must be replaced by the coefficient

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

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 $Q_{\text{abs}}^{\text{FF}}(a)$ can be defined as $Q_{\text{abs}}^{\text{FF}}(a) = Q_{\text{ext}}^{\text{FF}}(a) - Q_{\text{sca}}^{\text{FF}}(a)$, 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 Eq. (6), making the coefficient C^{FF} independent of r and equal to

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

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

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

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.[16–19]

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 et al.[16] derived analytical expressions for the efficiency factors of absorbing spherical particle in an absorbing medium,[17]

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

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

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

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]

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

$$b_j' = \frac{m' \varphi_j(mx)\varphi_j'(m'x) - m\varphi_j'(mx)\varphi_j(m'x)}{m' \zeta_j(mx)\varphi_j'(m'x) - m\zeta_j'(mx)\varphi_j(m'x)}, \tag{15}$$

$$c_j' = \frac{m'\varphi_j(mx)\varphi_j'(mx) - m'\zeta_j'(mx)\varphi_j(mx)}{m'\zeta_j(mx)\varphi_j'(m'x) - m\zeta_j'(mx)\varphi_j(m'x)}, \quad (16)$$

$$d_j' = \frac{m'\zeta_j'(mx)\varphi_j(mx) - m'\zeta_j(mx)\varphi_j'(mx)}{m'\zeta_j'(mx)\varphi_j(m'x) - m\zeta_j(mx)\varphi_j'(m'x)}. \quad (17)$$

Note that when the matrix is nonabsorbing, i.e., $m = n$, the above defined coefficients a_j' and b_j' are identical to a_j and b_j defined in Eqs. (4) and (5) for the conventional Mie theory.

Therefore, this approach eliminates the ambiguity in the definition of the extinction efficiency factor since the formulas 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[17–19] have shown 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 $Q_{\text{ext}}^{\text{NF}}(a)$ approaches unity as diffraction can be neglected.[19] These results contrast with the case of large spheres in a nonabsorbing matrix where the Mie theory predicts that $Q_{\text{ext}}^{\text{M}}(a)$ approaches 2.[18,11] In addition, the scattering efficiency factor $Q_{\text{sca}}^{\text{NF}}(a)$ 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]

B. Multiple Scatterers and Radiation Transfer

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

First, radiation transfer has often been treated as a diffusion process accounting for multiple scattering events.[22–26] Durian and co-workers[22–25,27] performed experimental, theoretical, and numerical studies on the angular distribution of the diffusely transmitted and backscattered 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 nonabsorbing 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 MC simulations can be performed. For example, Wong and Mengüç[29] simulated depolarization of a collimated and polarized light through nonabsorbing foams consisting of large spherical bubbles by using a combined MC–ray tracing approach as a 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) by using MC 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 by 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 detail by Pilon and Viskanta[33] for various porosities and bubble sizes. In brief, the following models for the effective absorption coefficient was proposed,[31]

$$\kappa_{\text{eff}} = \kappa - \pi \int_0^\infty [Q_{\text{abs},m}^{\text{M}}(a) - Q_{\text{abs},m'}^{\text{M}}(a)]a^2 f_1(a)\mathrm{d}a, \quad (18)$$

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 is defined as the number of bubbles per unit volume having radius between a and $a + \mathrm{d}a$. The efficiency factors $Q_{\text{abs},m}^{\text{M}}(a)$ and $Q_{\text{abs},m'}^{\text{M}}(a)$ are computed for a sphere of continuous phase (m) or dispersed phase (m'), respectively. They are estimated using the asymptotic formulas (see 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 nonparticipating matrix,[11,35]

$$\sigma_{\text{eff}} = \pi \int_0^\infty Q_{\text{sca}}^{\text{M}}(a)a^2 f_1(a)\mathrm{d}a, \quad (19)$$

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

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, $Q_{\text{sca}}^{\text{M}}(a)$ is calculated based on the anomalous diffraction approximation.

More recently, Dombrovsky[21] questioned the validity of the above model on the basis that Eq. (18) had not been validated and that $Q_{\text{sca}}^{\text{M}}(a)$ was estimated by 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,

$$\kappa_{\text{eff}} = \kappa + \pi \int_0^\infty Q_{\text{abs}}^{\text{FF}}(a)a^2 f_1(a)\mathrm{d}a, \quad (21)$$

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

Moreover, the second term on the right-hand side of Eq. (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., $\kappa_{\text{eff}} \leqslant \kappa$.

Finally, two practical questions remain unanswered and are addressed in this paper: (1) Among all the abovementioned theories, which one should be used to estimate the efficiency factors of a spherical scatterer in an absorbing media? (2) What would be the expressions of the associated radiation characteristics needed to solve the RTE? The present study aims first to determine 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 Mie theory and the far-field and the near-field approximations for specific absorbing media and particle or 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 scatters 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 four times their radius.[37,38]

A. Difference between Far-Field and Near-Field Approximations

This subsection compares the results for the different efficiency factors obtained by (1) Mie theory, (2) the far-field approximation, and (3) the near-field approximation. The results for the Mie theory were computed based on the code provided by Bohren and Huffman.[39] The same code was adapted for the far-field approximation following the suggestions by Mundy *et al.*[12] reviewed in Section 2 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 Sun and was vali-
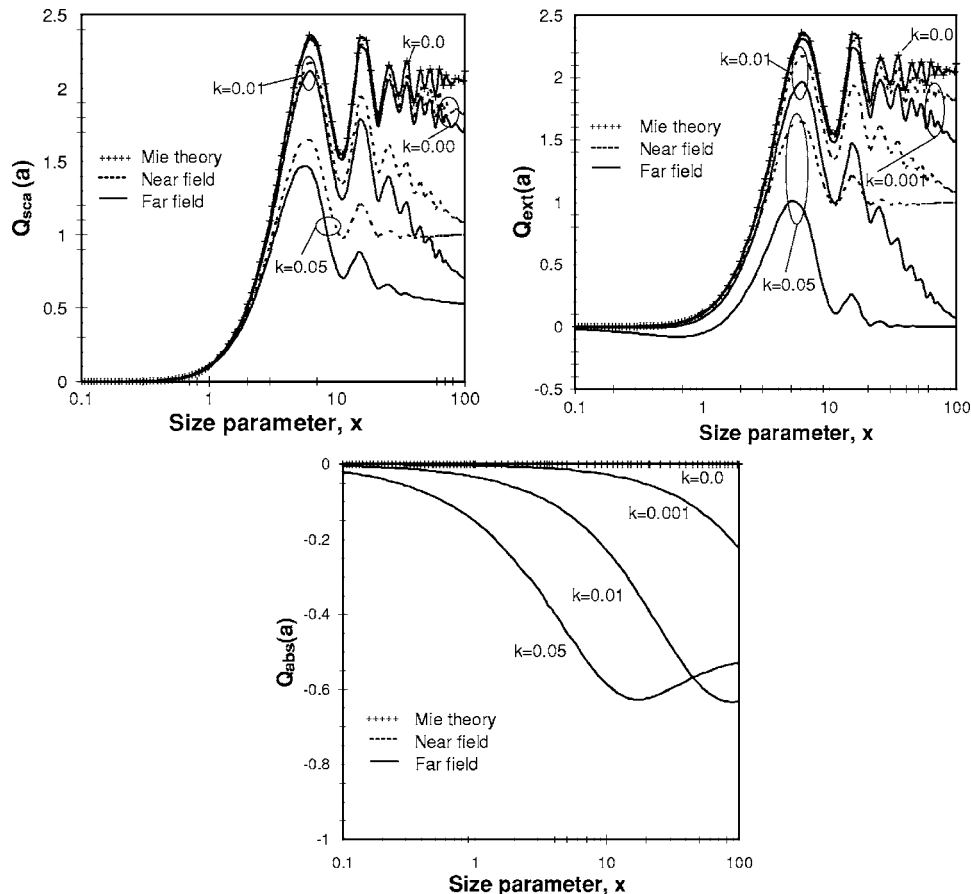

Fig. 1.   Scattering, extinction, and absorption efficiency factors as functions of size parameters for a spherical bubble ($m' = 1.0$) embedded in a medium having $m = 1.34 - ik$ with $k = 0.0, 0.001, 0.01,$ and 0.05.
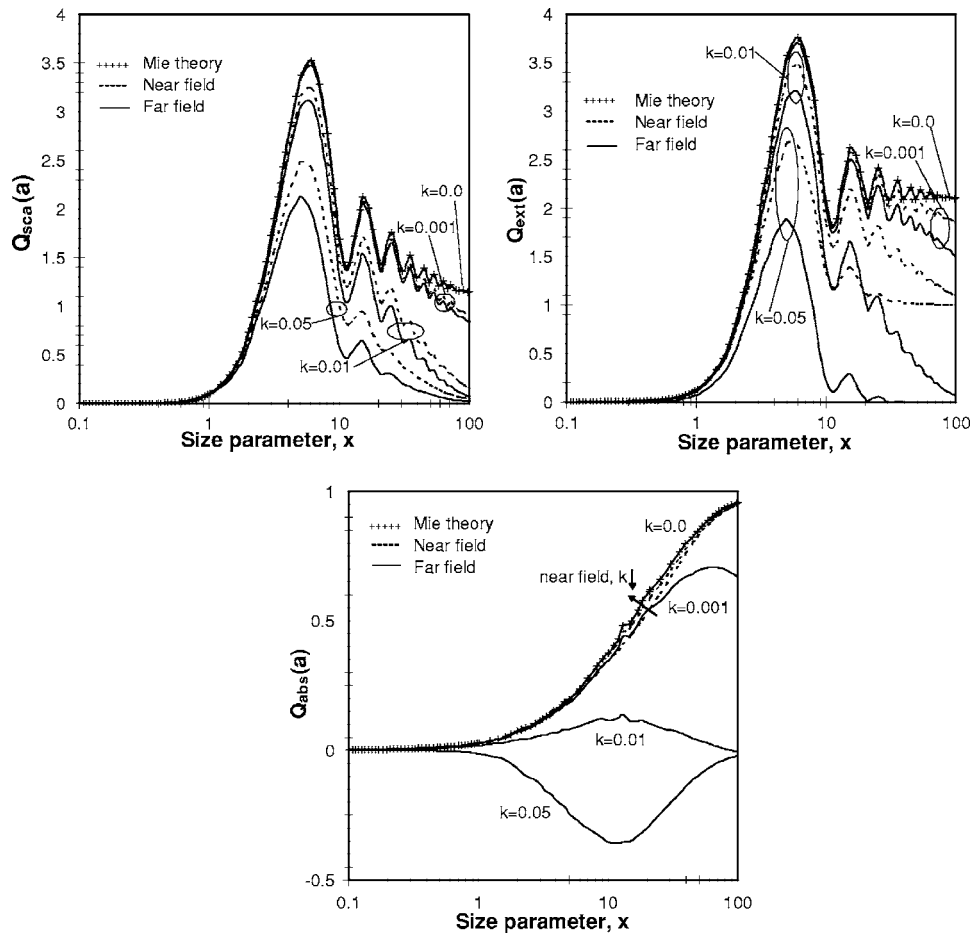
Fig. 2.   Scattering, extinction, and absorption efficiency factors as functions of size parameters for an absorbing spherical particle ($m' = 1.34 - 0.01i$) embedded in a medium with refractive index $m = 1.0 - ik$ with $k = 0.0$, 0.001, 0.01, and 0.05.

dated against Fu and Sun's results.[17] Here, the same situations as those explored by Fu and Sun[17] were investigated. In all cases, the series in Eqs. (1), (2), (9), and (10) were truncated and terminated when the summation index j was 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 nonabsorbing bubble ($m' = 1$) embedded in an absorbing medium of refractive index $m = 1.34 - ik$ with k equal to 0, 0.001, 0.01, and 0.05. First, for nonabsorbing matrix ($k = 0$), Mie theory, the far-field, and the near-field approximations gave identical results with $Q_{abs}(a) = 0$ and $Q_{sca}(a) = Q_{ext}(a)$. In addition, the extinction efficiency factor converged to 2 as the size parameter tended to infinity, corresponding to the well-known diffraction paradox.[11]

Moreover, Fig. 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. In contrast,

$Q_{sca}^{FF}(a)$ 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, Fig. 3 shows the results for a 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 nonabsorbing 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. In contrast, the absorption and extinction efficiency factors obtained by the far-field approximation can be negative if k is larger than k' as shown in Figs. 1 and 2 when $k = 0.05$ and $k' = 0.01$. Note also that $Q_{abs}^{NF}(a)$ was 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 Fig. 4. They indicate 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. A 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 Mie theory and the near-field approximation for the efficiency factors are shown in Fig. 5. They indicate that the relative difference in the absorption efficiency factors between 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 matrices 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 Mie theory and the far-field approximation are much larger than those shown in Fig. 5. Thus, one can see that Mie theory deviates significantly from the near-field and far-field approximations for matrices with large absorption indices and/or for large size parameters. 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 and is unimportant for all practical purposes.

B. Application to Radiation Transfer

One of the main motivations in determining single particle efficiency factors is for radiative transfer calculations that 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.[20,40] 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
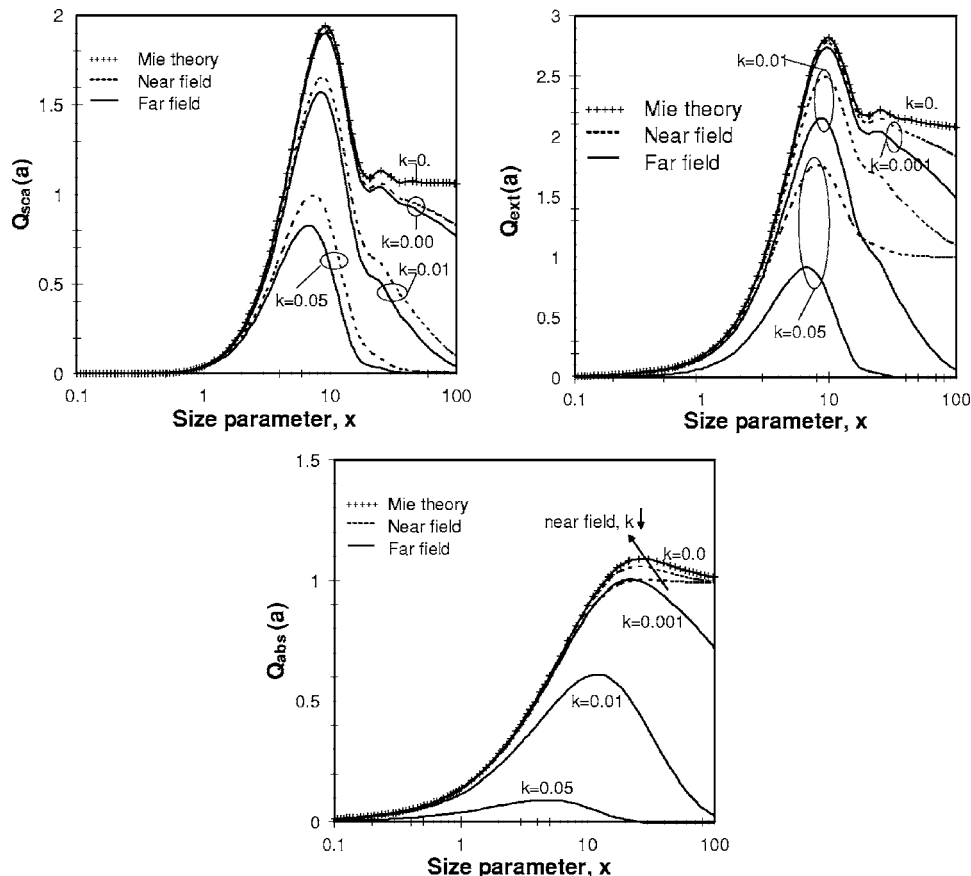

Fig. 3. Scattering, extinction, and absorption efficiency factors as functions of size parameters for an absorbing spherical particle ($m' = 1.4 - 0.05i$) embedded in a medium with refractive index $m = 1.2 - ik$ with $k = 0.0$, 0.001, 0.01, and 0.05.
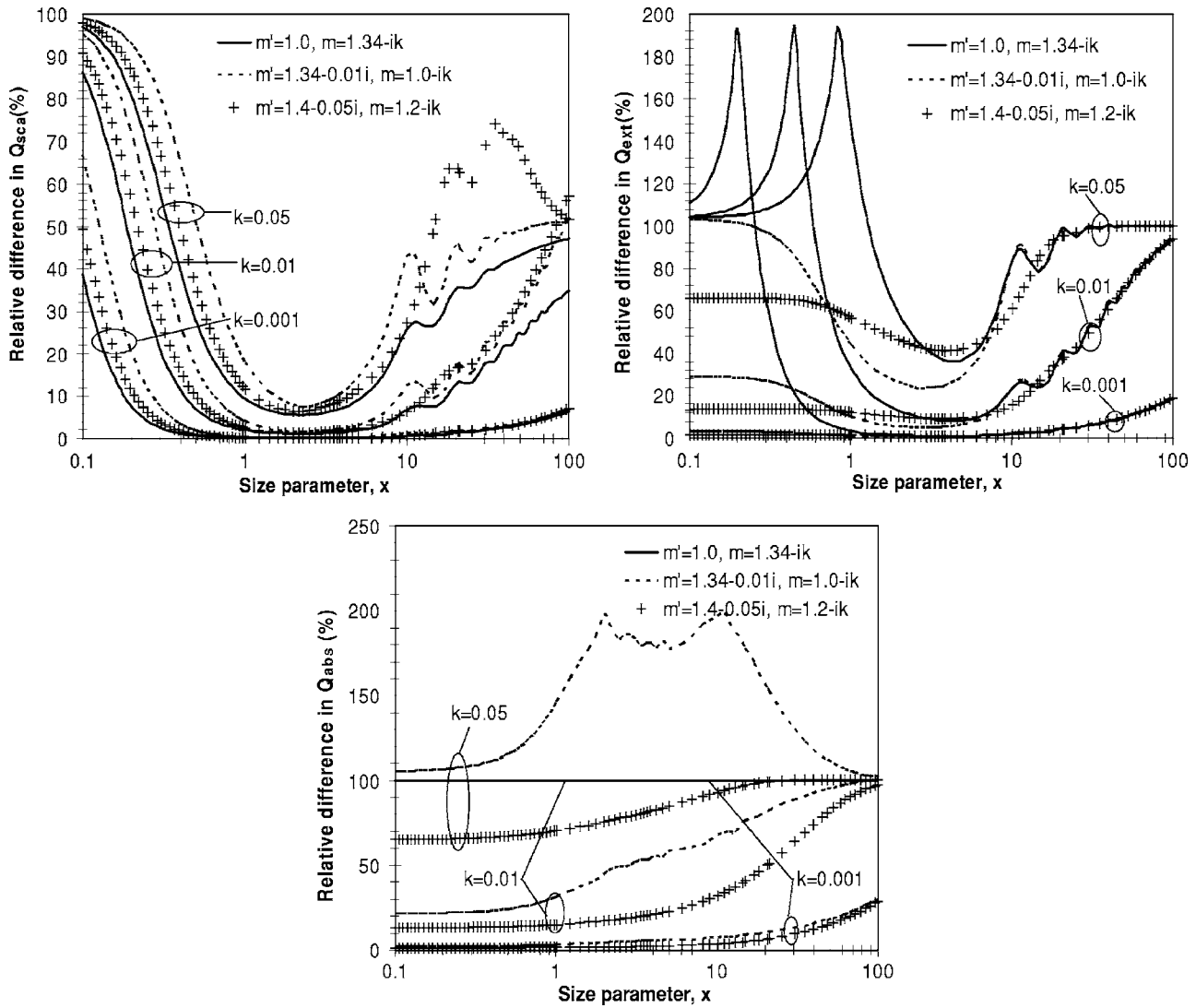
Fig. 4. Relative difference between near-field and far-field approximations for the scattering, extinction, and absorption efficiency factors as functions of size parameters.

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 by 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 Eq. (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 nonnegative. 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.[12–15]

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 nonexponential decay of the near-field scattered radiation in the absorbing matrix. This approach is referenced by the superscript NE. The nonexponential 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]

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

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: (i) the unattenuated absorption efficiency factor $Q_{abs}^{FF}(a)$ defined

by Mundy et al.[12] with the constant C^{FF} given by Eq. (7), (ii) the near-field absorption efficiency factor $Q_{abs}^{NF}(a)$ given by Eq. (10), and (iii) the absorption efficiency factor $Q_{abs}^{NE}(a)$ given by Eq. (23).

The absorption efficiency factor to be used to estimate the effective absorption coefficient will depend on the model selected [e.g., Eq. (18) or Eq. (21)]. In Fedorov and Viskanta's model,[31] $Q_{abs}^{M}(a)$ and $Q_{abs,m}^{M}(a)$ were calculated by 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 by using $Q_{abs}^{FF}(a)$ (Ref. 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 $Q_{sca}^{NF} - Q_{sca}^{FF}(a) \geqslant 0$ and $Q_{abs}^{NF}(a) \geqslant 0$, even for bubbles. Thus, it cannot be used in combination with Eq. (21). In addition, the term $Q_{abs,m}^{NE}(a) - Q_{abs,m'}^{NE}(a)$ can be negative or positive depend-

ing 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 also appear to be incompatible with Eq. (18). Then, a new model for κ_{eff} conceptually compatible with the definition of $Q_{abs}^{NE}(a)$ should be developed.

C. Application: Radiation Characteristics of Water Containing Bubbles
This subsection discusses the effective radiation characteristics of water containing bubbles that are 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]

1. Effective Scattering Coefficient
First, note that the expression for the effective scattering coefficient proposed by Fedorov and Viskanta[31] [Eq. (19)] and by Dombrovsky[21] [Eq. (22)] differ only by the choice of the model for the scattering efficiency factor. As discussed above, $Q_{sca}^{FF}(a)$ 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
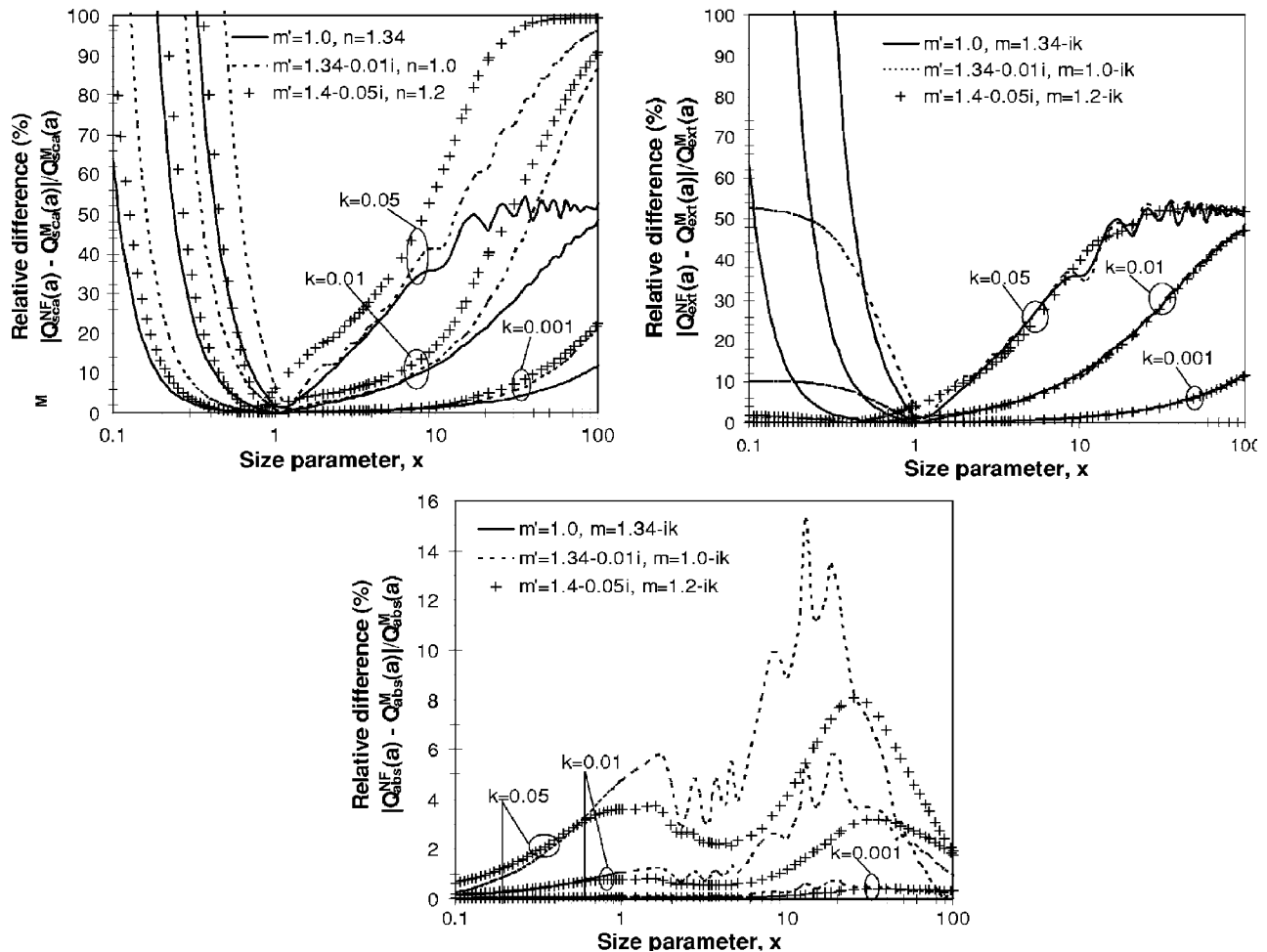

Fig. 5.   Relative difference between Mie theory and near-field approximation for the scattering, extinction, and absorption efficiency factors as functions of size parameters.
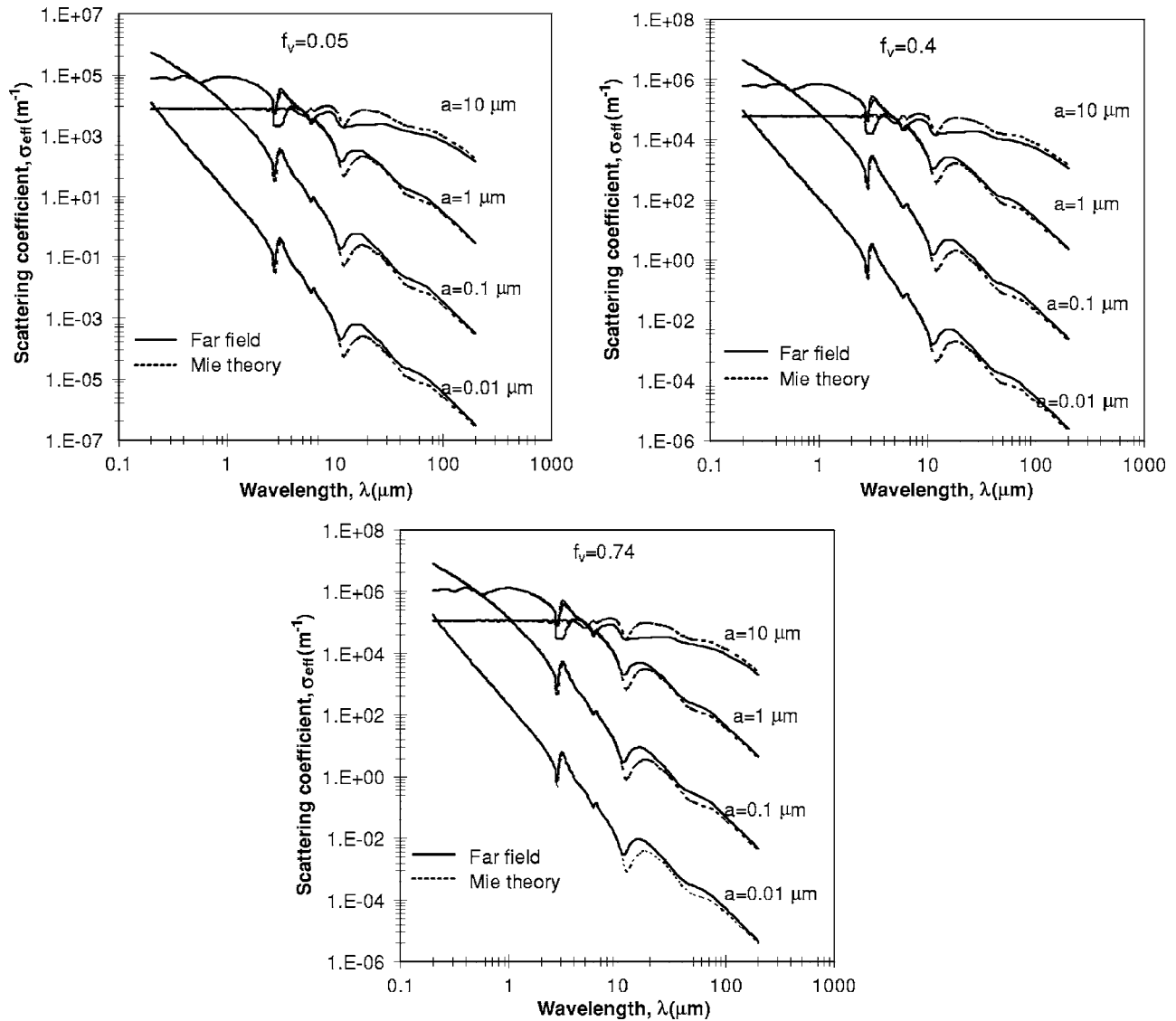
Fig. 6. Effective scattering coefficients accounting (far-field approximation) for or neglecting (Mie theory) the matrix absorption of air bubbles in water as functions of wavelength and bubble radius for void fractions f_v = 0.05, 0.4, and 0.74.

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

Figure 6 compares the effective scattering coefficient predicted by Eq. (24) (solid curves) with that predicted by the Mie theory (assuming $k = 0$) (dotted curves) as a function of wavelength for different void fractions and bubble radii. For a 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 the void fraction. For wavelengths smaller than 2 μm, the relative error is less than 10%, while it can be larger than 50% for wavelengths at approximately 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 in the effective scattering coefficient.

2. Effective Absorption Coefficient

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

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

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

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 were investigated over the spectral range from 0.2 to 200 μm.

Figure 7 compares the effective absorption coefficient predicted by Eqs. (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 dominated the overall absorption of the composite medium. In other works, $\kappa_{eff} \approx \kappa$ 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 with average radii of 0.64 mm and void fraction of 4% in the spectral region from 1.67 to 3.5 μm where the quartz was weakly absorbing ($k < 10^{-4}$) and $\kappa x \ll 1$.

However, when the void fraction increases, the differences become large (Fig. 7). Then the second term on the right-hand sides of Eqs. (25) and (26) dominates. Also, $\kappa_{eff,2}$ is much smaller than $\kappa_{eff,1}$. In addition, $\kappa_{eff,2}$ is nega-

tive for wavelengths at approximately 3 and 11–200 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 nonphysical 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 Fig. 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

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

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 wave-
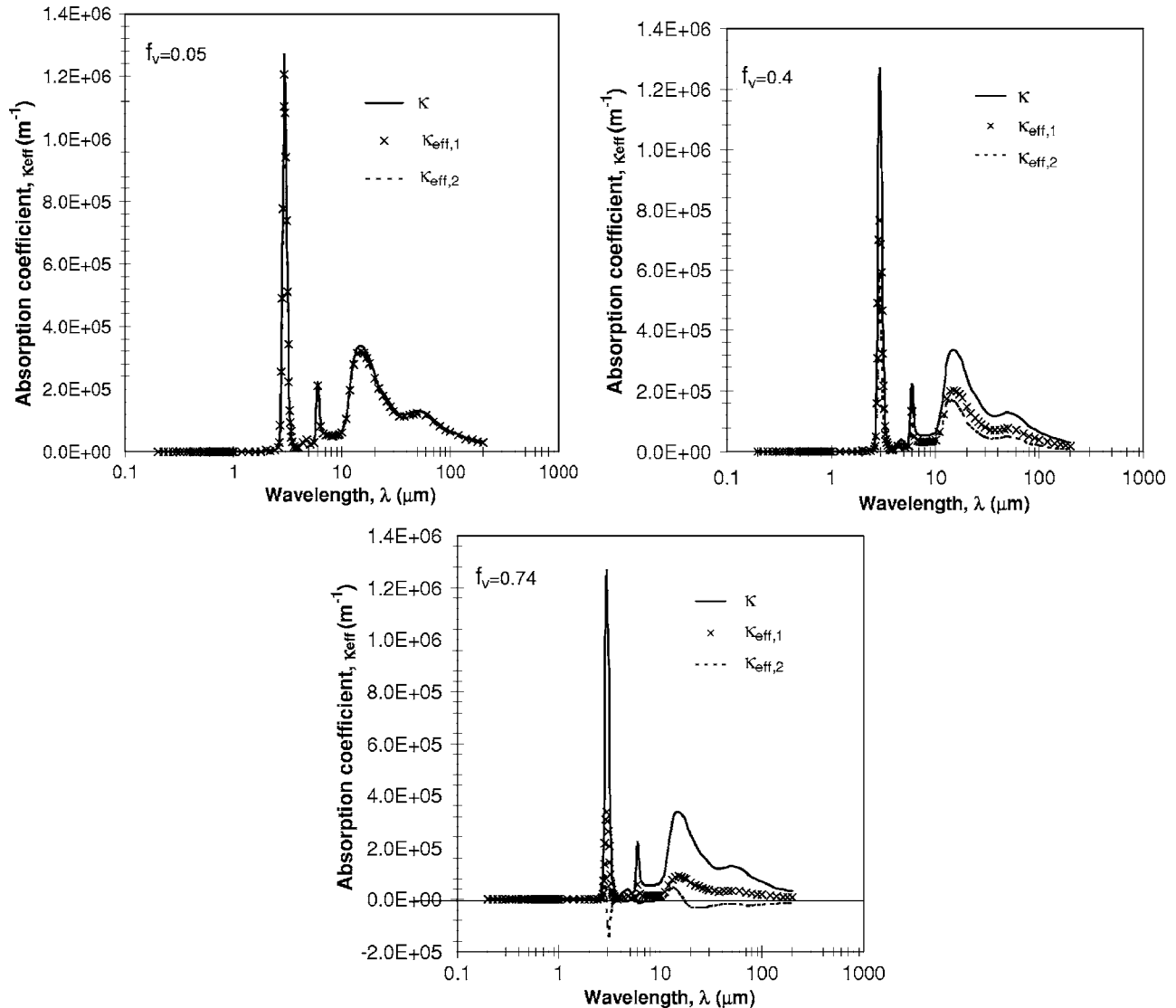

Fig. 7.   Effective absorption coefficients of air bubbles of radius $a=0.01$ μm in water as functions of wavelength for void fraction f_v =0.05, 0.4, and 0.74.
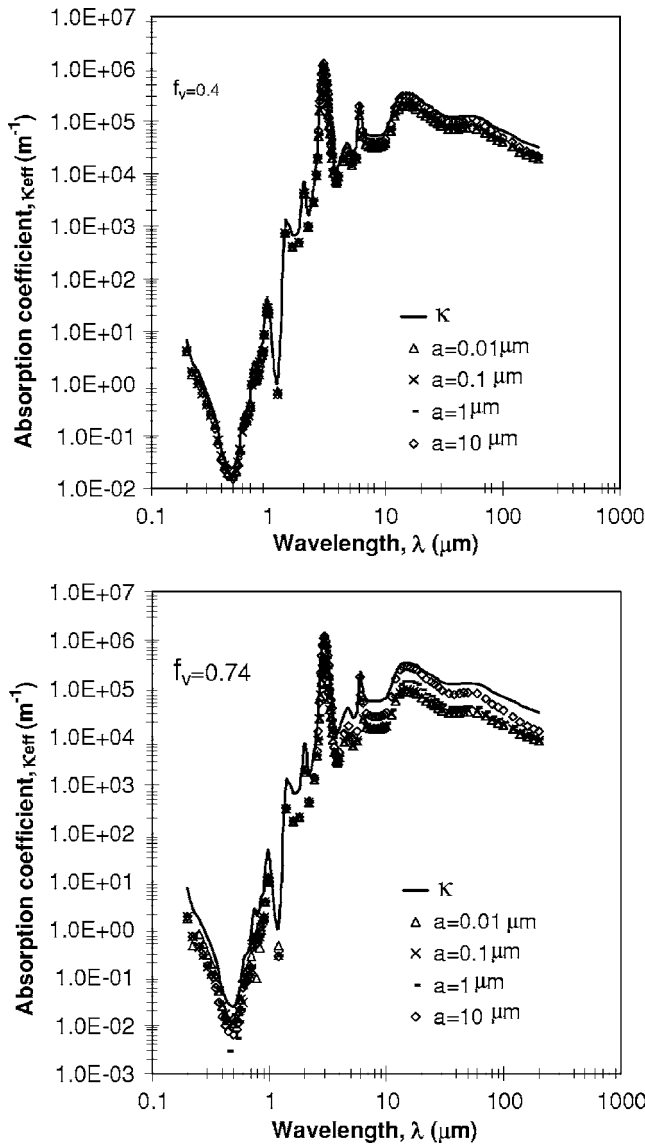
Fig. 8.   Effective absorption coefficients of monodispersed air bubbles in water as functions of wavelength predicted by Eq. (25) for $f_v = 0.4$ and 0.74 and various bubble radii.

lengths. Increasing the void fraction results in larger relative errors. For example, for $f_v = 0.4$, the relative error can be as high as 20% at some wavelengths, and for $f_v = 0.74$ it may exceed 80% for some combinations of wavelengths and bubble radii. 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 a two-phase mixture consisting of bubbles in an absorbing medium. The efficiency factors predicted by the conventional Mie theory and the far-field and 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 were drawn:

1. Ignoring the absorption index of the matrix can result in significant error on the scattering and extinction efficiency factors predicted by the conventional Mie theory except when the host medium is nonabsorbing or weakly absorbing. This is particularly true if the size parameter is large. Then the near-field and 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 $Q_{\text{sca}}^{\text{FF}}(a)$.
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 a 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 nonabsorbing media can be used. However, at some wavelengths, neglecting the absorption of media results in large errors in the efficiency factors and in the associated radiation characteristics.

Finally, note that the experimental data for media with large volume fractions of scatters and/or for matrices with relatively large absorption indices are still needed to validate the above effective radiation characteristic models. Alternatively, the rigorous approach developed by Mishchenko[40] based on 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.

APPENDIX A: NOMENCLATURE

1. Variables and Functions

$$
\begin{aligned}
a &= \text{Radius of particles or bubbles.} \\
a_j, b_j, c_j, d_j &= \text{Mie coefficients.} \\
a_j', b_j', c_j', d_j' &= \text{Coefficients in Eqs. (12) and (13).} \\
C &= \text{Coefficient.} \\
f_1 &= \text{Size distribution function of particles.} \\
f_v &= \text{Volume void fraction or porosity.} \\
\text{Im} &= \text{Imaginary part of a complex number.} \\
j &= \text{Index number.}
\end{aligned}
$$

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, $2\pi a/\lambda$.

2. 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.

3. Subscripts

eff = Refers to the effective properties.
sca = Scattering.
abs = Absorption.
ext = Extinction.

4. Superscripts

M = Refers to the Mie theory.
FF = Refers to the far-field approximation.
NF = Refers to the near-field approximation.
NE = Refers to the nonexponential decay model (Ref. 41).

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