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THERMAL RADIATION OF SPHERICAL AND CYLINDRICAL SOOT PARTICLES

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Thermal Radiation of Spherical and Cylindrical Soot Particles

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Abstract

The effect of soot shape on soot radiation from flame and smoke is considered in the present study. It is realized that soot particles generally conglomerate into different shapes, while the limiting bounds are being either spherical or long chains. By modelling the long chains as infinite cylinders, it is shown that the radiation extinction characteristics of the spherical and cylindrical soot are distinctly different, with that of the spherical particles exhibiting a cut-off wavelength phenomenon. Emissivity from a cloud of cylindrical particles is always higher than that of the spheres due to their higher extinction coefficients. Moreover, the effect of soot shape on radiation is more pronounced at typical smoke temperatures (300°K) than at flame temperatures. A simple experimental method is also proposed for determining the amount of cylindrical and spherical particles in a cloud.

Introduction

The combustion products from burning hydrocarbon fuels are generally consisted of several gaseous constituents and numerous small solid particulates called soot. It has long been recognized that soot plays an important role in the radiative heat transfer from flames and smoke. For example, soot in flames is responsible for the flame luminosity and contributes greatly to the infrared radiation of flames, while that in smoke accounts for light obscuration and infrared absorption. In order to estimate the radiative contribution by soot to flame and smoke, detailed soot information must be known.

Soot particles are characterized by the optical properties, composition (Hydrogen/Carbon ratio), size distributions, and shapes. By assuming that soot is spherical, other properties have been explored in several previous investigations.[1-6] Computations were all based on the Mie theory[7] for spheres. However, it is well known that soot particles usually conglomerate into larger chunks and long chains during and subsequent to the combustion process. The effect of conglomeration is seldom taken into consideration due to the complexities involved.

Although a number of studies have been made on the interaction of electromagnetic waves with assemblies of particles,[7-9] the radiative heat transfer characteristics of these assemblies have not been explored. From the engineering standpoint, a careful assessment is needed concerning the effect of soot conglomeration on radiation both in flames and smoke. It is the objective of the present work to investigate specifically the effect of soot shape on the radiation of soot clouds.

Analytical Basis

The radiative heat transfer characteristics of a sooty medium may vary greatly due to soot conglomeration. Information on the amount of various conglomerated shapes is required for the calculation of flame and smoke radiation. This is particularly important in predicting soot ratiation from the combustion of hydrocarbon fuels, in which case coagulation of soot particles in flames and smoke is generally observed.[10,11] The complexities and uncertainities involved in accounting for all particle shapes indeed present a formidable problem and may not justify any detail elaborate computation for practical applications. It is, however, important to realize the limiting bounds of the radiation calculations when the particles are either all spherical or long chains. The extinction and emission characteristics of a cloud with different shapes of conglomerated particles should lie somewhere between the limits obtained for the two types of particles. Exploitation of these two cases would then reveal the effect of soot shape on soot radiation.

Spherical Particles

The light scattering and extinction characteristics of spherical particles can be obtained using the Mie theory.[7,9] Assuming homogeneous and spherical soot particles, the extinction efficiency is given by

$$Q_s\ (\underset{\sim}{m},\alpha) = \frac{2}{\alpha^2} \sum_{m=1}^{\infty} (2m + 1)\ \mathrm{Re}\ (a_m + b_m) \qquad (1)$$

where $\underset{\sim}{m}$ (=n - i κ) is the refractive index, α (= 2π r/λ) the size parameter, r the radius, λ the wavelength, a_m and b_m the coefficients in terms of Ricatti-Bessel functions, and Re the real parts of the coefficients. Theoretical prediction of the spectral extinction coefficient of a polydisperse cloud requires information on the optical properties and size distribution

$$K_\lambda = \int^{\infty} \pi r^2 N(r) Q_s dr \quad (2)$$

where N(r) is the particle size distribution. A size distribution generally used is[12,13]

$$N(r) = a r^3 e^{-br} \quad , \quad b > 0 \quad (3)$$

The constants a and b may be represented in terms of physical parameters as

$$a = N_t b^4/\Gamma(4) \quad (4a)$$

$$b = 3/r_m \quad (4b)$$

where N_t is the total number of particles, r_m the most probable radius, and Γ the Gamma function. The soot volume fraction is defined by

$$f_v = \int_0^{\infty} \frac{4}{3} \pi r^3 N(r) dr \quad (5)$$

and from Eq. (3), it follows that

$$f_v = \frac{4}{3} \pi a \frac{\Gamma(7)}{b^7} \quad (6)$$

Substituting Eq. (6) in (2), the spectral extinction coefficient becomes

$$K_\lambda = 2.278 \frac{f_v}{r_m^7} \int_o^\infty r^5 \exp \left(- 3 \frac{r}{r_m}\right) Q_s \, dr \quad (7)$$

In the limit of small particles ($\alpha << 1$), the extinction efficiency Q_s is independent of particle size, K_λ then reduces to

$$K_\lambda = \frac{36 \pi n \kappa}{(n^2 - \kappa^2 + 2)^2 + 4 n^2 \kappa^2} \cdot \frac{f_v}{\lambda} \quad (8)$$

which is the well-known Rayleigh limit expression. The emissivity from a soot cloud is

$$\varepsilon = \frac{\int_o^\infty (1 - e^{-K_\lambda L}) \, e_{b\lambda} \, d\lambda}{\sigma T^4} \quad (9)$$

where L is the pathlength, $e_{b\lambda}$ the spectral black body emissive power, σ the Stefan-Boltzmann constant, and T the temperature. If n and κ in Eq. (8) may be replaced by some suitable average values in the infrared, a closed-form expression for the soot emissivity may be obtained

$$\varepsilon = 1 - \frac{15}{\pi^4} \psi^{(3)} \left(1 + \frac{C_s f_v L T}{C_2}\right) \quad (10)$$

where $\psi^{(3)}$ is the Pentagamma function, C_2 the Planck's second constant and

$$C_s = \frac{36 \pi n \kappa}{(n^2 - \kappa^2 + 2)^2 + 4 n^2 \kappa^2} \tag{11}$$

Computations of the luminous flame emissivity using these approximate expressions can then be greatly simplified.[14,15]

Cylindrical Particles

The other limiting case of soot conglomeration is the long chains which may be approximated as cylinders. Since the length to diameter ratio of such chains is usually very large, a good approximation would be to treat these chains as infinitely long cylinders. The interaction of electromagnetic wave with infinite cylinders have been studied in detail.[7,9,16] Rigorous solution of the Maxwell's equations yields the extinction efficiency per unit length for unpolarized radiation as

$$Q_c\ (\underset{\sim}{m},\alpha,\phi) = \frac{2}{\alpha} \mathrm{Re}\ \{\frac{A_o + B_o}{2} + \sum_{m=1}^{\infty} (A_m + B_m)\} \tag{12}$$

where ϕ is the angle between the incident electromagnetic wave and the normal to the cylinder axis, and Re the real parts of the coefficients A_m and B_m which are functions of integral order Bessel functions. Since particles in a cloud are generally randomly oriented, the extinction efficiency averaged over all orientations is

$$\overline{Q}_c = \frac{2}{\pi} \int_0^{\pi/2} Q_c(\phi)\ d\phi \tag{13}$$

The spectral extinction coefficient for a monodisperse cloud is

$$K_\lambda = \lim_{\ell\to\infty} (2\ r\ \ell\ N_t\ \overline{Q}_c) \tag{14}$$

where ℓ is the length of the particles. The volume fraction is

$$f_v = \lim_{\ell\to\infty} (\pi\ r^2\ \ell\ N_t) \tag{15}$$

Equation (14) then becomes

$$K_\lambda = \frac{2}{\pi r}\ \overline{Q}_c\ f_v \tag{16}$$

which is independent of length. Rigorous consideration of the extinction characteristics of finite cylinders yield similar results as long as $\ell \gg r$ and λ.[7]

An exact closed-form limiting expression for the extinction efficiency of small cylinders ($\alpha \ll 1$) analogous to that of spherical particles is not available due to their complicated angular dependence. However, an approximate expression may be obtained based on the values at perpendicular incidence such that

$$\overline{Q}_{RAY} = \beta\ \frac{\alpha}{4}\ [\pi\ n\ \kappa + \frac{4\ \pi\ n\ \kappa}{(n^2 - \kappa^2 + 1)^2 + 4\ n^2\ \kappa^2}] \tag{17}$$

which approximates $\overline{Q}_c$ quite well in the infrared if $\beta = 1.1$. The spectral extinction coefficient then becomes

$$K_\lambda = C_c\ \frac{f_v}{\lambda} \tag{18}$$

where

$$C_c = \beta \pi n \kappa \quad [1 + \frac{4}{(n^2 - \kappa^2 + 1)^2 + 4 n^2 \kappa^2}] \tag{19}$$

If n and κ can be replaced by some suitable average values in the infrared, emission from a cylindrical soot cloud may be obtained using Eq. (10).

Soot radiation calculations may also be greatly simplified if a mean coefficient can be defined. In the optically thick region, the Rosseland mean coefficient is appropriate:

$$k_R = \int_0^{\infty} \frac{\partial e_{b\lambda}}{\partial e_b} d\lambda \Big/ \int_0^{\infty} \frac{1}{K_\lambda} \frac{\partial e_{b\lambda}}{\partial e_b} d\lambda \tag{20}$$

Using Eqs. (8) or (18), assuming that C_s and C_c are some average values in the infrared, Eq. (20) becomes[17]

$$k_R = 3.6 \frac{C_o}{C_2} f_v T \tag{21}$$

where C_o equals either C_s or C_c or a combination. The total emissivity may be obtained as

$$\varepsilon = 1 - e^{-k_R L} \tag{22}$$

A further extension is to consider the concept of radiation conductivity. This is appropriate in engineering radiative heat flux computations since the need to consider the equation of transfer can be eliminated. For an optically thick medium bound by black surfaces at temperatures T_1 and T_2, the heat flux is

$$q \approx \frac{\sigma (T_1^4 - T_2^4)}{\frac{3}{4} k_R L} \tag{23a}$$

$$\approx \frac{16 \sigma T_m^3}{3 k_R} \cdot \frac{dT}{dx} \tag{23b}$$

where T_m is the mean temperature. The radiation conductivity is defined as

$$k_{RAD} = \frac{16 \sigma T_m^3}{3 k_R} \tag{24}$$

By using k_{RAD}, radiative heat transfer can be easily incorporated into other modes of heat transfer mechanisms.

Assuming that the spherical particles are polydisperse and cylindrical particles are monodisperse, the extinction and emission of radiation by the soot clouds are investigated using the exact expressions Eqs. (2), (9) and (16). The soot optical constants recently reported[6] are employed in all calculations, and they are adjusted accordingly for their temperature dependence.

Results and Discussion

A good assessment of the radiative heat transfer characteristic of conglomerated particles may be obtained by examining the extinction and

and emissions of radiation by such particles. In particular, a study of the limiting bounds of conglomeration, i.e., spherical particles and long chains, can offer much insight into the problem. By modelling the long chains as infinite cylinders, the extinction coefficiencies of the spherical and cylindrical particles (Q_s and $\overline{Q}_c$) are presented in Fig. 1 for typical soot sizes. It is observed that $\overline{Q}_c$ is generally higher than Q_s which decreases rapidly in the infrared range, exhibiting a cut-off wavelength phenomenon; whereas $\overline{Q}_c$ falls off relatively much slower, thus attenuating radiation far into the infrared. Assuming that spherical particles are polydisperse and cylindrical particles are monodisperse, a comparison of their spectral extinction coefficients is shwon in Fig. 2. The extinction coefficients K_λ are generally higher for larger particles except in the visible range. While in the far infrared, they all converge to their limiting values respectively, denoting that Rayleigh limit is approached. The rapid decrease of K_λ with wavelength of the spherical particles reveals the basic difference of radiation extinction between the two types of particles. Consequently, emission from a cylindrical soot cloud is always higher than that from the spherical soot as shown in Fig. 3. As temperature is decreased, the peak blackbody emissive power shifts toward longer wavelengths where extinction by cylindrical particles becomes more dominant. This accounts for the increasingly large difference of emission between the two types of particles toward lower temperatures. It is also interesting to see that even for a ten-fold difference in particle size, the emissivities of the cylindrical particals converge rapidly to the limiting value of 300°K. This is, of course, due to the rapid convergence of K_λ to their limiting values in the Rayleigh limit.

Having explored the spherical and cylindrical particles separately, emission from a cloud containing both types of particles is next considered. As shown in Figs. 4 and 5, the emissivity of such a soot cloud decreases with increasing spherical soot content. This is especially significant at low temperatures due to the cut-off wavelength characteristics exhibited by the spherical particles. Whereas, the emissivity at high temperature is almost independent of the type of particles present. Since spherical and cylindrical particles represent limiting bounds of conglomeration, these emissivities may be interpreted as the limiting values indicating the maximum effect of soot conglomeration on soot radiation. Consequently, it may be concluded that the effect of soot shape on soot emission is most pronounced at low temperatures; while at typical flame temperatures, the soot emissivity is only slightly affected.

All the above results have been obtained based on the assumption of monodisperse cylindrical particles. This assumption is now relaxed to include a bimodal size distribution containing particles of radii 0.01 μm and 0.1 μm. At typical smoke temperatures, the emissivity is almost independent of size as shown in Fig. 6, since the small particles limit is approached. The effect of polydispersion becomes more pronounced at elevated temperatures. Although a more realistic particle size distribution should really be used in considering a truely polydisperse system, a simple bimodal distribtution of typical soot sizes should be sufficient for an illustration of its effect on soot radiation.

An experimental method for the determination of the amount of different types of particles in a cloud is next investigated. One possibility is the sampling technique, where particles are collected using probes. The amount of each particle shapes may be inferred from studying the micrographs. However, this technqiue is clearly inappropriate, since introducing the probe in the path of particle flow would inevitably cause the breaking up of long chains as well as enhancing agglomeration. An optical diagnostic technique is favored since it would not disturb the system. Consider a cloud containing only spherical particles and long chains, the extinction coefficient for typical soot sizes in the infrared is

$$K_\lambda = \frac{C_c}{\lambda} x f_v + \frac{C_s}{\lambda}(1 - x) f_v \tag{25}$$

where x is the fraction of cylindrical particles. Observing the cut-off wavelength characteristic of the spherical particles (fig. 2), attenuation of radiation in the far infrared is mostly due to the cylindrical particles. Thus, a transmission measurement in the far infrared would yield $x f_v$, the volume fraction of cylindrical particles. Using this information and a transmission measurement in the near infrared, both x and f_v may also be determined.

Conclusion

In the study of soot radiation from flame and smoke, it is generally assumed that the soot particles are spherical and polydisperse, while neglecting soot conglomeration. The calculation of soot emission is consequently greatly simplified, particularly when calculating the luminous flame emissivity. However, the effect of soot conglomeration on radiation must be realized to evaluate the sphericity assumption of soot. In the present simplified treatment, the limiting bounds of conglomeration, i.e., spherical particles and long chains modelled as infinite cylinders, are considered. It is shown that the extinction characteristics of the two particle shapes are distinctly different, with that of the spherical particles exhibiting a cut-off wavelength phenomenon. A simple in-situ experimental method for the determination of the amount of different particle shapes in a soot cloud is then discussed based on this observation. Consequently, emission from cylindrical soot is always higher due to their higher extinction coefficients. The difference in soot emission between the two types of particles is largest at low temperatures, while it diminishes with increasing temperature. Therefore, the effect of soot shape on soot radiation is more pronounced in smoke than in flames. On the other hand, relaxing the monodisperse assumption of cylindrical particles shows that the effect of a size distribution is only important at elevated temperatures.

Acknowledgment

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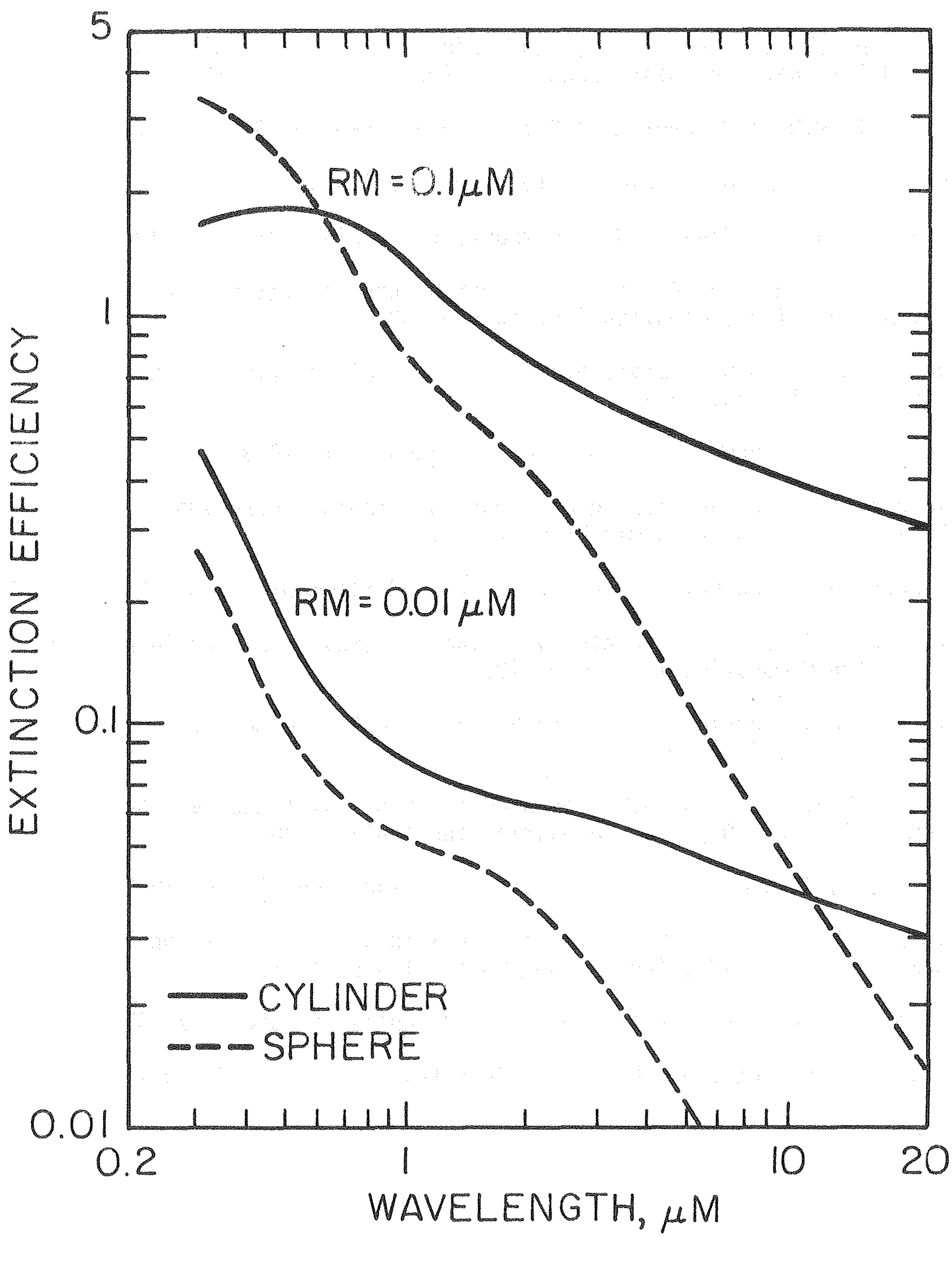

Figure 1 Spectral Extinction Efficiencies of Cylindrical and Spherical Particles

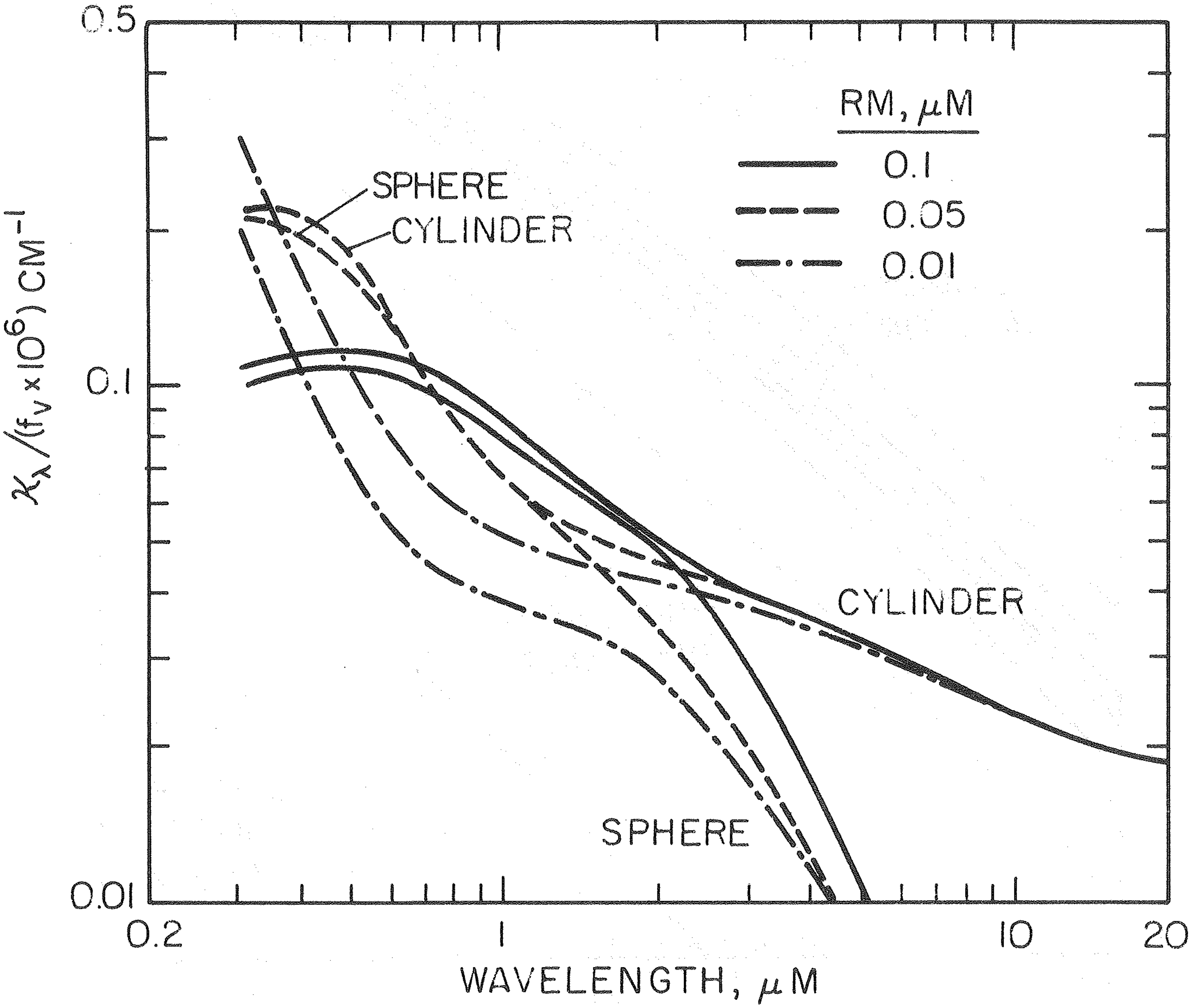

Figure 2 Spectral Extinction Coefficients of Cylindrical and Spherical Particles

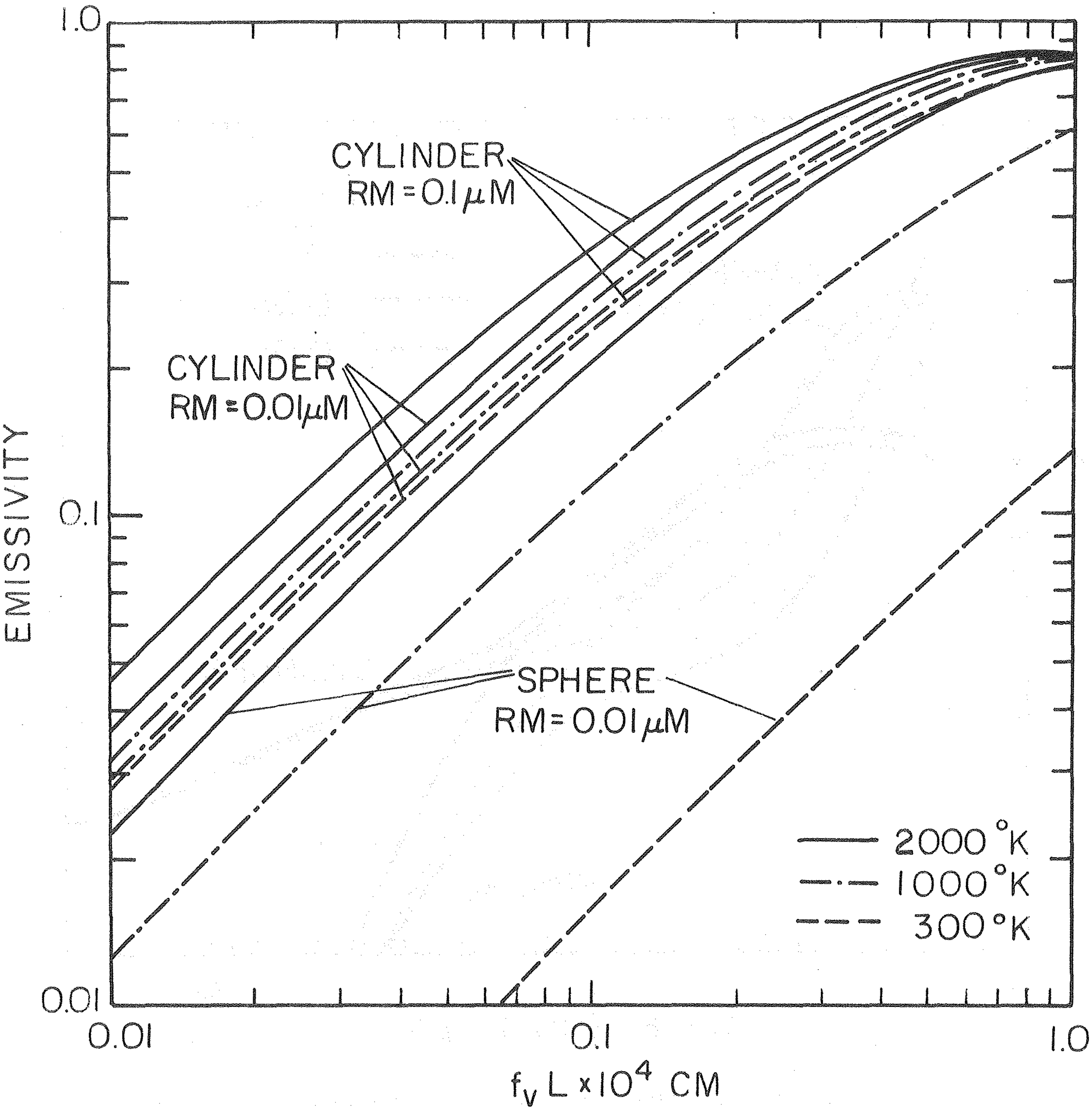

Figure 3 Total Emissivity of Cylindrical and Spherical Soot Particles

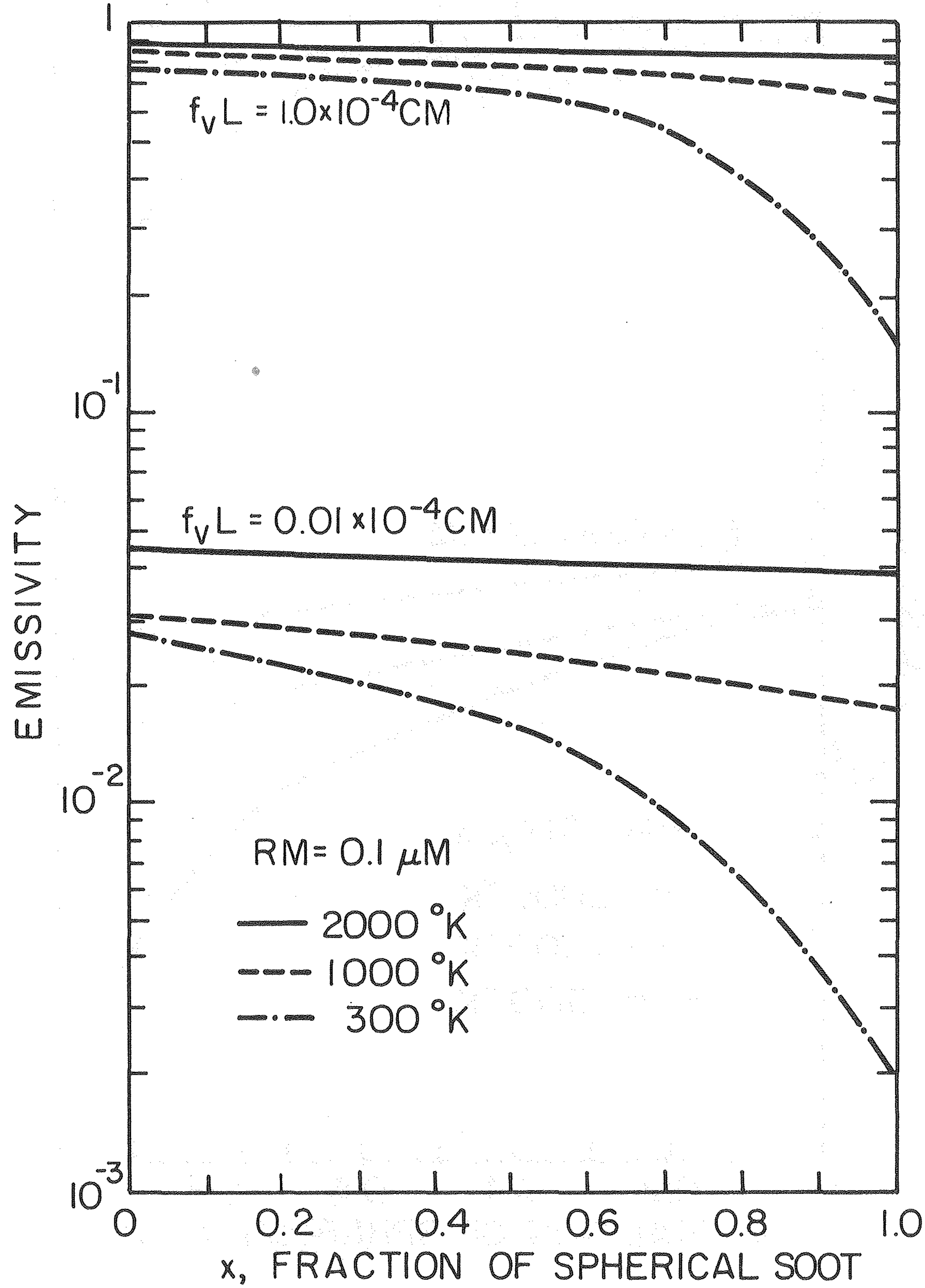

Figure 4 Total Emissivity of a Soot Cloud Containing Both Cylindrical and Spherical Particles

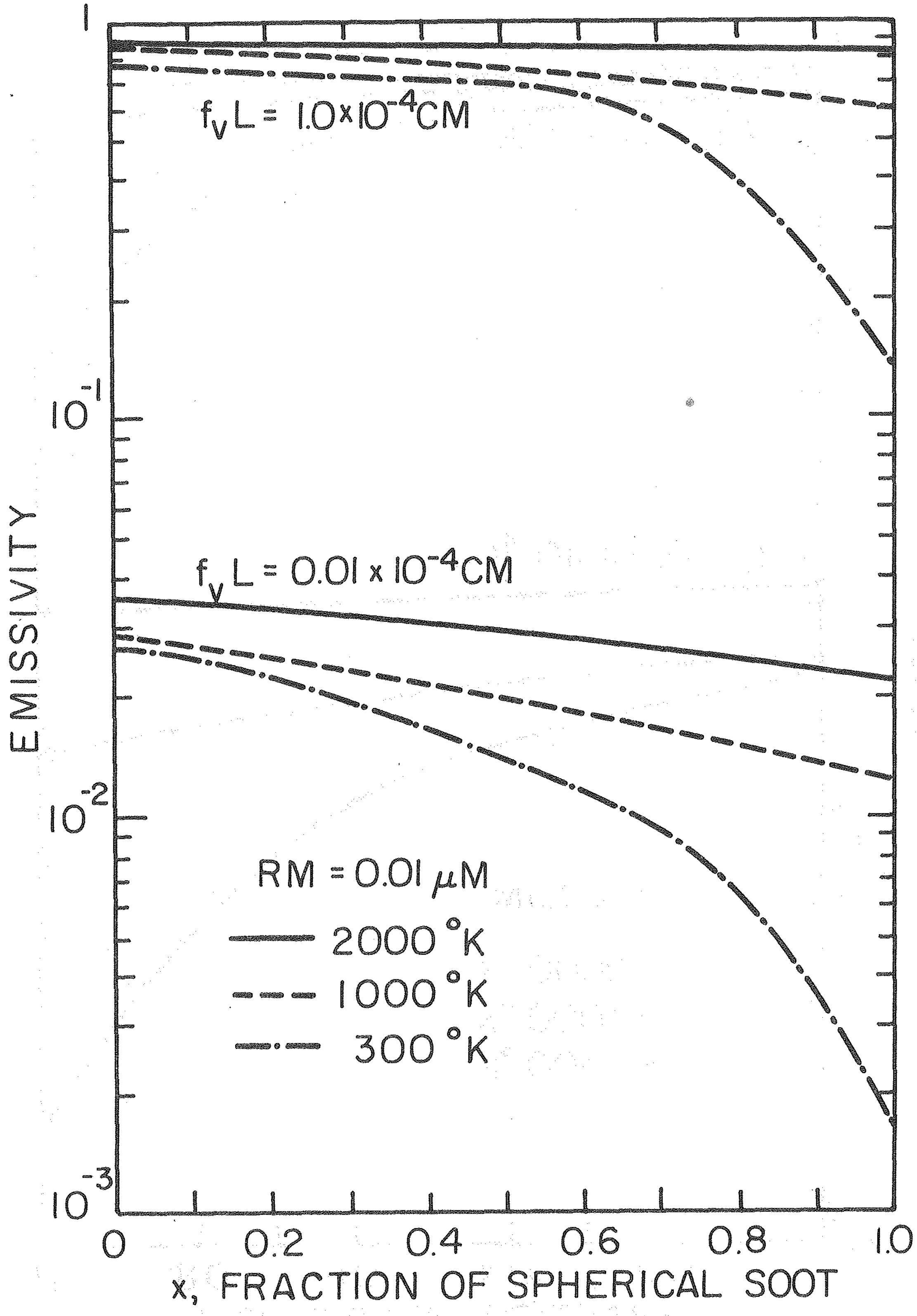

Figure 5 Total Emissivity of a Soot Cloud Containing Both Cylindrical and Spherical Particles

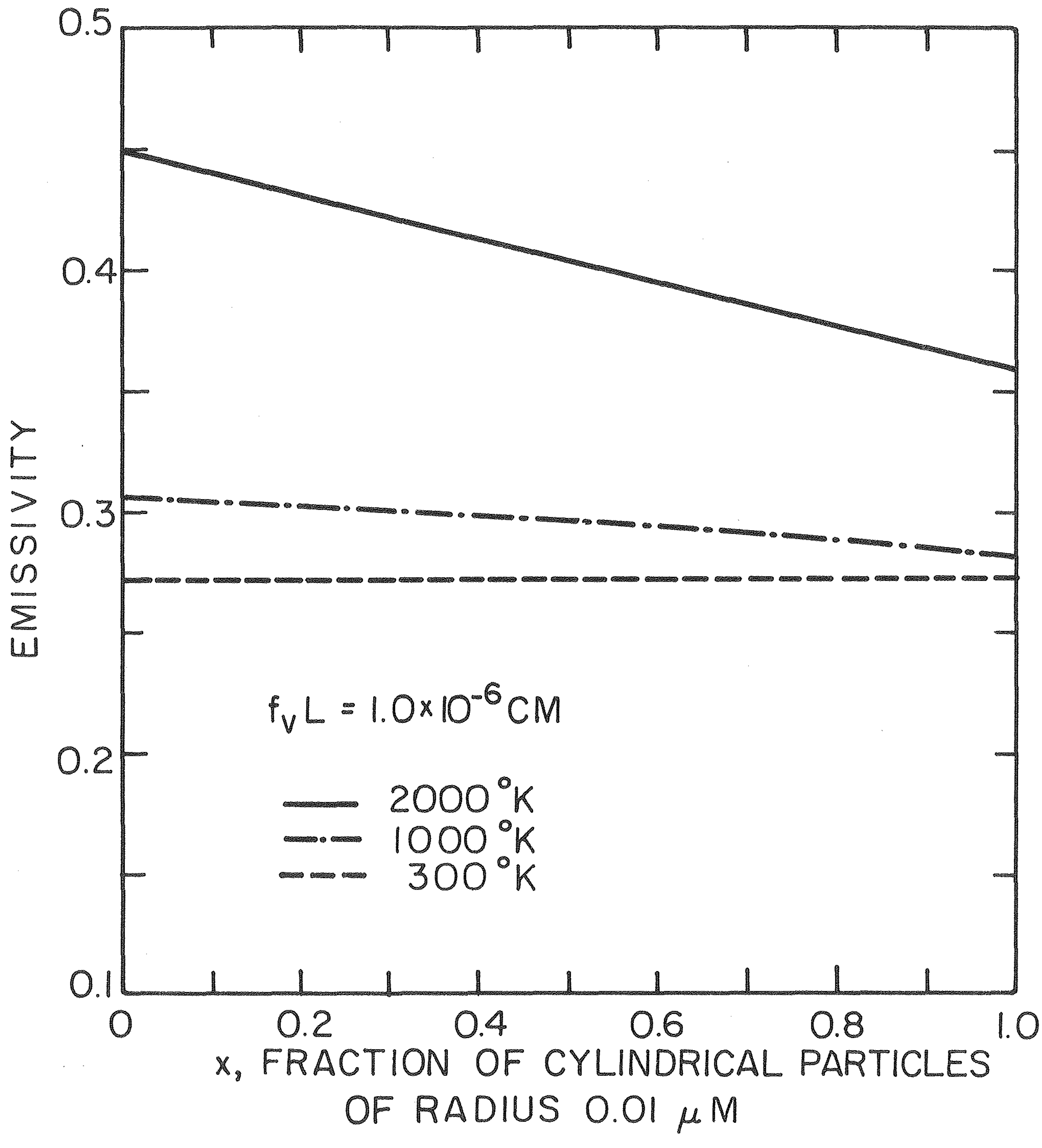

Figure 6 Total Emissivity of a Cloud of Cylindrical Particles with Two Sizes (RM=0.01 & 0.1 μM)