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Technical Notes

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Effective Optical Constant of Alumina Particle Containing Carbon

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Nomenclature

a	=	radius of inner core, μm
b	=	radius of outer concentric shell, μm
f_c	=	volume fraction of carbon
$f_{Al_2O_3}$	=	volume fraction of alumina
k	=	imaginary part of the complex refractive index (absorption index)
m	=	mixture's effective complex refractive index
$m_{Al_2O_3}$	=	complex refractive index of alumina
m_c	=	complex refractive index of carbon
n	=	real part of complex refractive index
T_m	=	melting temperature of alumina particle, K
δ	=	free parameter
ε	=	mixture's effective dielectric constant
ε_c	=	dielectric constant of carbon
$\varepsilon_{Al_2O_3}$	=	dielectric constant of alumina
λ	=	wavelength, μm

Subscripts

Al_2O_3	=	alumina
c	=	carbon soot

I. Introduction

ALUMINA (Al_2O_3) particles are present in many types of flames including solid propellant rocket exhaust. Accurate modeling of exhausted alumina particle properties is essential for assessment of the rocket base heat transfer, plume diagnosis, and investigation. The complex refractive index of alumina particles is one of the essential optical properties needed for the radiation calculation. These properties of pure alumina have been extensively studied and fairly well understood [1–3]. The alumina's optical properties in a plume, however, are rather difficult for an accurate consideration, because they are strongly dependent on the crystalline structures, the phase status, and the purity of the material [4–6]. It has been concluded that the high radiance observed in solid rocket plumes is due to impurities, specifically, unburned metallic aluminum or carbon [7,8]. We mainly focus on the effect of the impurity of carbon in alumina particles. The process of phase transition is also considered when particle temperature is crossing the melting point.

Different from the pure alumina particle, the alumina particle in a plume has some different components, so that the optical properties of an alumina particle in an exhaust plume are effective properties. The concept of an effective medium has been used to deal with the optical properties of atmospheric aerosols for a long time. Here, we try to use this method to predict the optical constants of alumina particles in an exhaust plume. The volume-averaged, Maxwell–Garnett, and Bruggeman mixing rules for effective optical constants have been applied to many types of materials [9,10], such as porous materials, solutions, composite films, and atmospheric aerosols. In the case of an alumina particle surrounded by a shell of carbon, the concentric shell model [11] has been used, in which the spherical particle is composed of two concentric regions, the shell and the core, each with its own uniform complex refractive index. All of these mixing rules can be used for the heterogeneous particle to be replaced by a homogeneous particle with an effective optical constant. In Sec. II, several expressions commonly used for an effective medium and their applications are introduced. In Sec. III, the optical constant of an alumina particle containing carbon impurities is simulated by using the four mixing rules. Section IV compares the results from the four different mixing rules to those of measurements. Conclusions are given in Sec. V.

II. Theory

Generally speaking, the morphology of a mixture should be taken into consideration when calculating its effective optical constant. If a composite particle can be modeled as a core of alumina sphere surrounded by a concentric shell of carbon, then the effective optical constant of the particle can be modeled by using the concentric shell model [11]. Suppose a particle of radius b is replaced by a composite particle whose inner core is spherical alumina of radius a and dielectric constant $\varepsilon_{Al_2O_3}$, while the outer concentric shell is carbon of radius b and dielectric constant ε_c. The effective dielectric constant ε of the concentric sphere can be approximated by [11]

$$\varepsilon = \varepsilon_c + \frac{\delta}{1/(\varepsilon_{Al_2O_3} - \varepsilon_c) + (1-\delta)/3\varepsilon_c} \tag{1}$$

where $\delta = (a/b)^3$, which is equal to the volume fraction of alumina $f_{Al_2O_3}$ in the composite particle. In this model, the volume fraction of alumina is equal to $(1 - f_c)$. In the case of $a = 0$, the material of the outer concentric shell composes the whole particle, and the effective dielectric constant of the particle is equal to that of carbon soot ($\varepsilon = \varepsilon_c$). In the case of $a = b$, there is no carbon in the particle, and the effective dielectric constant is that of alumina $\varepsilon = \varepsilon_{Al_2O_3}$.

If the components are mixed in such a way that a layered model is not appropriate, there is not an exact analytical solution for the geometry of the mixture. Then, an appropriate mixing rule is required. In this section, we present a brief introduction to the three common mixing rules: the volume-averaged, Maxwell–Garnett, and Bruggeman rules, which can be employed to predict macroscopic dielectric properties for a material consisting of various components.

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The effective optical constants of the mixture can be calculated as a function of the constituent optical constants, their volume fractions, and possibly some other parameters characterizing the microstructure of the mixture. A full review of the theory and application of Maxwell–Garnett and Bruggeman mixing rules can be found in texts dealing with effective medium approximations [11]. Only a summary of some key points relevant to the current study are included here. We now present the definitions of the mixing rules.

The Maxwell–Garnett mixing rule should be used for the case of many separated spherical inclusions randomly distributed throughout the particle. For the alumina particles with carbon impurities, alumina is the host medium with dielectric constant $\varepsilon_{\mathrm{Al_2O_3}}$, and isolated spheres of carbon having dielectric constant ε_c are embedded in the host medium. Then, the Maxwell–Garnett mixing rule is defined as [11]

$$\frac{\varepsilon - \varepsilon_{\mathrm{Al_2O_3}}}{\varepsilon + 2\varepsilon_{\mathrm{Al_2O_3}}} = f_c \frac{\varepsilon_c - \varepsilon_{\mathrm{Al_2O_3}}}{\varepsilon_c + \varepsilon_{\mathrm{Al_2O_3}}} \tag{2}$$

where ε is the mixture's dielectric constant. It is well known that the Maxwell–Garnett mixing rule is valid only when the values of f_c are very small.

Unlike the Maxwell–Garnett mixing rule, the Bruggeman mixing rule is also valid for large values of f_c. The Bruggeman mixing rule allows a calculation of effective dielectric constant ε for a mixture where the carbon impurity is interspersed. It is defined as [11]

$$f_c \frac{\varepsilon_c - \varepsilon}{\varepsilon_c + 2\varepsilon} + (1 - f_c) \frac{\varepsilon_{\mathrm{Al_2O_3}} - \varepsilon}{\varepsilon_{\mathrm{Al_2O_3}} + 2\varepsilon} = 0 \tag{3}$$

For a homogeneous mixture, a simple volume average of the individual complex refractive indices is used. The volume-averaged complex refractive index m is defined by [10]

$$m = f_c m_c + (1 - f_c) m_{\mathrm{Al_2O_3}} \tag{4}$$

Note that the effective dielectric constant mixing rule is generally valid in the limit that the sizes of the constituents (besides the host material) are small as compared to the wavelength of the incident radiation [9].

III. Calculations

The effective optical constant of a composite particle depends on the optical constants of all constituents and the morphology of the mixture. Several formulations of effective medium theory and their applications have been introduced in Sec. II. In this section, the effective optical constants of an alumina particle with carbon impurity are calculated by using these mixing rules. Their algebraic expressions are shown in Eqs. (1–4). Because the real part of the complex refractive index does not significantly contribute to the emissive properties of particles in a plume [3], this paper is primarily concerned with the imaginary part, which is the absorption index.

The optical constants of the constituents are important parameters needed for calculations. It is well known that size effects may occur for particles with very small diameters, and the optical constants may differ significantly from those of the corresponding bulk values. The optical constant data chosen for calculation should adequately represent those of the alumina particles and the carbon soot in a rocket plume. Pure alumina's absorption index (imaginary part) and the carbon soot's complex refractive index can be found in [3]. The real part of an alumina particle's complex refractive index has little dependence on temperature and wavelength [12,13], and so the real part of the complex refractive index in this Note only changes in accordance with phase status. According to Oliver and Moylan [14], an alumina particle is assumed to be in the liquid phase when its temperature is above the melting point ($T_m = 2327$ K), and when below the melting point, solid alumina is in the stable alpha phase. Krishnan et al. [1] and Oliver and Moylan [14] give the real parts of an alumina particle's liquid and alpha-phase complex refractive indices. Note that the complex refractive index is the square root of the dielectric constant.

IV. Results and Discussion

The calculated effective absorption indices of an alumina particle containing various levels of carbon impurity, at a wavelength of 0.5 μm, are shown in Figs. 1–4. It is indicated that the concentric shell model gives results similar to those of the other three mixing rules at the least volume fraction of 0.001 (Fig. 1). The differences between the results of these mixing rules become bigger as the volume fraction of carbon increases. The Bruggeman mixing rule and the Maxwell–Garnett mixing rule also give very similar results at the moderate volume fractions of 0.01 and 0.05 (Figs. 2 and 3). The

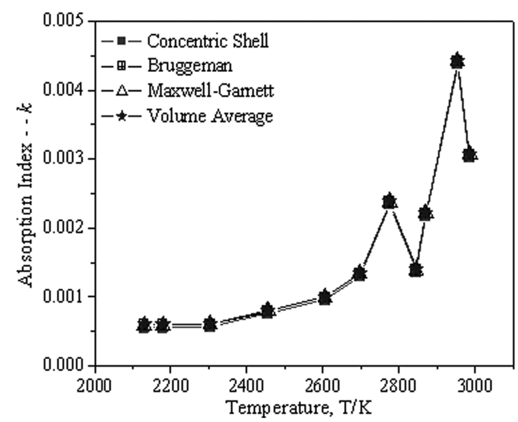

Fig. 1 Effective absorption index of an alumina particle with $f_c = 0.001$, $\lambda = 0.5$ μm.

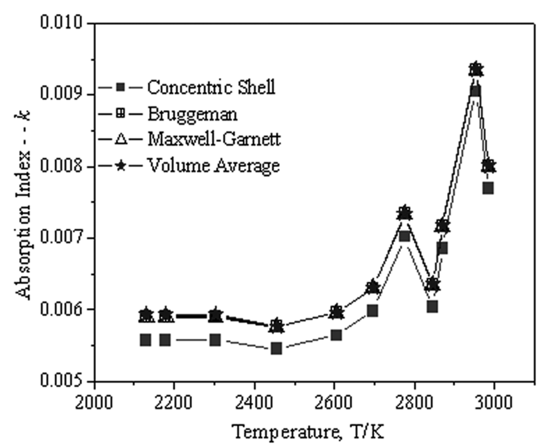

Fig. 2 Effective absorption index of an alumina particle with $f_c = 0.01$, $\lambda = 0.5$ μm.

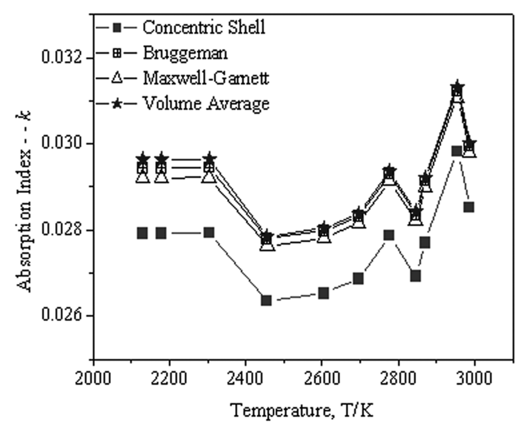

Fig. 3 Effective absorption index of an alumina particle with $f_c = 0.05$, $\lambda = 0.5$ μm.

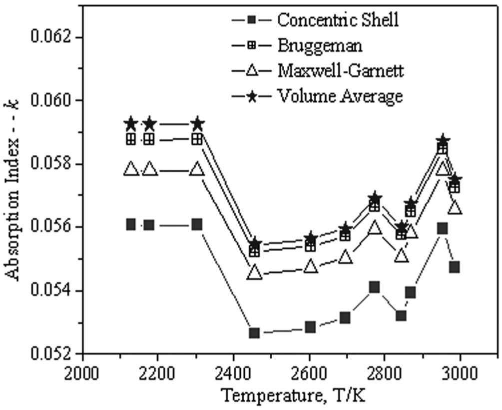

Fig. 4 Effective absorption index of an alumina particle with $f_c = 0.1$, $\lambda = 0.5\ \mu$m.

percentage differences between the Bruggeman and the Maxwell–Garnett results are ±0.1% for the volume fractions of 0.01 and ±0.7% for the volume fractions of 0.05. When the volume fraction increases to 0.1 (Fig. 4), the results of the Maxwell–Garnett mixing rule begin to deviate from the results of the Bruggeman mixing rule by up to 1.6%.

The effect of wavelength is also considered in this Note. In Table 1, the results for an alumina particle with a carbon volume fraction of 0.05 are shown at three wavelengths (0.5, 4.03, and 7.30 μm). The temperature of the particle is 2695 K in this calculation. It is found that the effective absorption index increases with an increase in the wavelength. The results of the concentric shell model are lower than those of other mixing rules at the wavelength of 0.5 μm, and when the wavelength increases to 7.30 μm, the result of the concentric shell model is the highest.

Simmons [3] presents the values of the absorption indices of a pure alumina particle and an alumina particle from an SB-15 motor exhaust plume. Assume carbon is the only impurity in the particle, and its effective absorption index is calculated. The concentric shell model requires that the absorbing core is situated exactly in the center of the sphere. The volume-averaged mixing rule is used for a homogeneous mixture, such as a mixture of solutions [10]. The Maxwell–Garnett and Bruggeman mixing rules are the most commonly used formulas, and can be used for an arbitrary composite system, but Bruggeman made a significant improvement to the Maxwell–Garnett mixing rule [11]; it treats the two composites in a symmetrical fashion and is valid for large values of f_c, and so the Bruggeman mixing rule is chosen for this calculation. The calculations are compared with reported measurements [3]. In Figs. 5 and 6, the calculated results for a particle containing different volume fractions of carbon are shown. The effective absorption index increases with increasing volume fraction of carbon for the two wavelengths of 2.65 and 4.03 μm. The calculated effective absorption index is adjacent to the measurements when the volume fraction of carbon is in the region of $0.025 \sim 0.125\%$. For the wavelength of 2.65 μm, the average percentage difference between the Bruggeman results and the measurements is the least at a carbon volume fraction of 0.05%; it is about 33.3%. When the wavelength is 4.03 μm, this minimum average percentage difference is at a carbon volume fraction of 0.075%; it is about 25.8% . As the volume fraction of carbon in an alumina particle increases, the discontinuity in the absorption index at the melting point ($T_m = 2327$ K) is significantly

Table 1 Wavelength dependence of effective absorption index

	$\lambda = 0.5\ \mu m$	$\lambda = 4.03\ \mu m$	$\lambda = 7.30\ \mu m$
Concentric shell	0.02686	0.06786	0.11809
Bruggeman	0.02831	0.06048	0.08186
Maxwell–Garnett	0.02815	0.0568	0.07416
Volume–average	0.02837	0.0692	0.10798

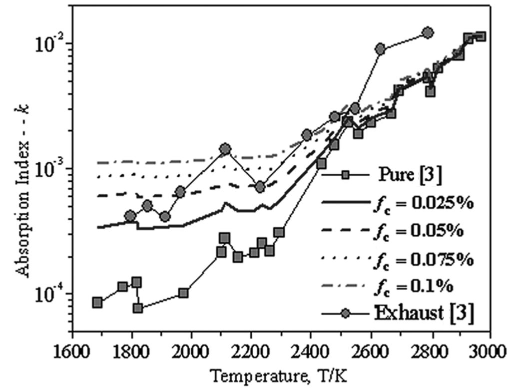

Fig. 5 Comparison between calculations and reported measurements, $\lambda = 2.65\ \mu$m.

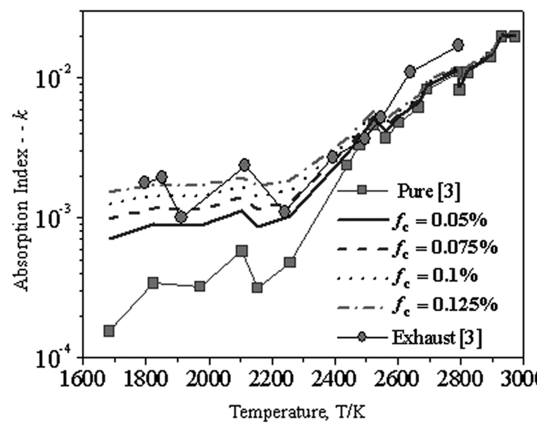

Fig. 6 Comparison between calculations and reported measurements, $\lambda = 4.03\ \mu$m.

reduced. This is consistent with measurements in the literature [3]. Therefore, carbon impurity is an important factor that can significantly reduce the discontinuity in the absorption index of pure alumina through the melting point.

V. Conclusions

The volume-averaged complex refractive index, Bruggeman mixing rule, Maxwell–Garnett mixing rule, and the concentric shell model for determining the effective optical constant of a binary mixture were tested for an alumina particle having carbon impurities. The effect of mixing morphology was considered by using different kinds of mixing rules. When the volume fraction of carbon is below 0.001, the results of these mixing rules are very similar. The differences among these results increase with increasing volume fraction of carbon impurity. The Maxwell–Garnett and Bruggeman mixing rules give similar results at carbon volume fractions below 0.1. The simulated results show that the presence of carbon impurities in alumina particles can significantly reduce the discontinuity in the absorption index at the melting point. The calculated effective absorption index is adjacent to the measured absorption index of an alumina particle in the exhaust plume when the volume fraction of carbon is in the region of $0.025 \sim 0.125\%$.

Carbon is not the only impurity in exhaust plume alumina particles. There may be a small fraction of unburned metallic aluminum or some other material contained in the particle. Then, the mixing rules for ternary or multicomponent mixtures need to be applied. Exhaust plume alumina particles tend to form spheroidal hollow particles during the solidification process. These factors have not been considered in our calculations, primarily due to the lack of experimental data.

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