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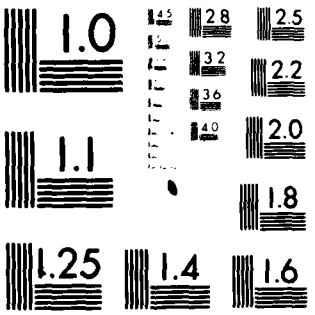

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EXTINCTION BY AEROSOL CLOUDS OF NONSPHERICAL PARTICLES AT ARBITRARY WAVELENGTHS

by

Janon F. Embury, Ph.D.

Research Division

March 1982

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US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
Chemical Systems Laboratory
Aberdeen Proving Ground, Maryland 21010

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Extinction by a cloud consisting of many aerosol particles has been found to fall into two distinct wavelength regions which are adequately described by the geometric optics theory and the Rayleigh theory. Because the cloud consists of many different particles, the narrow extinction resonance structure of individual particles is lost, and extinction is governed by shape and size as predicted by geometric optics. At longer wavelengths, particles are in the Rayleigh region, and extinction for a large variety of particle shapes is predicted by the

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20. ABSTRACT (Contd)

Rayleigh ellipsoidal theory. The transition region lying between the applicability of these two simple theories occupies only about one wavelength decade.

PREFACE

The work described in this report was authorized under project 1L162622A554-C, Research in Aerosol/Obscuration Science. This work was started in January and completed in June 1981.

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EXTINCTION BY AEROSOL CLOUDS OF NONSPHERICAL PARTICLES AT ARBITRARY WAVELENGTHS

1. INTRODUCTION

Most aerosol clouds consist of a polydispersion of particles. Distributions in size greatly simplify extinction spectra of clouds by averaging out the size-dependent extinction resonance peaks of individual particles. If the aerosol particles are not liquid, then they can be nonspherical and have a collection of orientations, surface irregularities, or a shape distribution, all of which serve to further average out narrow extinction resonance peaks. What remains of the extinction spectra may be treated by two limit theories; the geometric optics limit and the Rayleigh limit. In the geometric optics limit, extinction is governed by shape and size, independent of complex refractive index. In the Rayleigh limit extinction by an absorbing particle is governed by shape and complex refractive index, independent of size.

If the particles are convex, then the shape and size dependence in the geometric optics limit takes the form of a direct proportionality between extinction and surface area per unit volume. The Rayleigh ellipsoidal theory describes extinction by a wide variety of particle shapes represented by elliposoids. For example, high aspect ratio prolate spheroids, ellipsoids of revolution, can be used to represent fibers, and high aspect ratio oblate spheroids can be used to represent disks or flakes reasonably well.

2. DISCUSSION OF THEORY

The extinction coefficient, α, defines the beam transmittance, T, through a cloud as a function of its concentration, c, and pathlength l. A convenient set of consistent units puts the pathlength in meters, the concentration in grams per cubic meter, and the extinction coefficient in square meters per gram of material.

$$T = e^{-\alpha c l} \tag{1}$$

The extinction coefficient depends upon the geometric cross section, G, of a mass of material ρV, having extinction efficiency factor, Q. These must be averaged over particle size, w, and solid angle, Ω, to get an average extinction coefficient to represent the complete cloud.

$$\alpha = \left\langle \left\langle \frac{GQ}{\rho V} \right\rangle_{\Omega} \right\rangle_{w} \tag{2}$$

Assuming that the angular average of the product GQ is equal to the product of the averages and that the average efficiency factor is equal to two,

$$\alpha = 2 \left\langle \frac{\langle G \rangle_{\Omega}}{\rho V} \right\rangle_{w} \tag{3}$$

For convex particles it is well known that the geometric cross section averaged over all angles is equal to one-fourth the surface area, S.

$$\alpha = \frac{1}{2\rho} \left\langle \frac{S}{V} \right\rangle w \tag{4}$$

Values of surface area per unit volume appear in table 1 for four typical shapes. These values for monodispersions appear in the first column, while the corresponding values for log normal polydispersions, having geometric standard deviations, σ, and number averaged dimensions subscripted n, appear in the second column. All dimensions are in microns when the density is in grams per cubic centimeter.

Table 1. Surface Area Per Unit Volume for Randomly Oriented Particles

Shape	Monodisperse	Polydisperse
	μ	μ
Sphere (r = radius)	$\frac{3}{r}$	$\frac{3}{r_n} e^{\frac{-5}{2}(\ln\sigma)^2}$
Cube (a = edge)	$\frac{6}{a}$	$\frac{6}{a_n} e^{\frac{-5}{2}(\ln\sigma)^2}$
Flake (t = thickness)	$\frac{2}{t}$	$\frac{2}{t_n} e^{\frac{-1}{2}(\ln\sigma)^2}$
Filament (r = radius)	$\frac{2}{r}$	$\frac{2}{r_n} e^{\frac{-3}{2}(\ln\sigma)^2}$

As these particles become smaller, they eventually become Rayleigh particles, the exact particle volume of transition depending upon shape and complex refractive index. Once this transition occurs, the extinction efficiency factor can no longer be considered as a constant equal to two, but instead becomes size dependent in such a way that it exactly cancels out the size dependence of the surface area per unit volume, resulting in a size-independent extinction coefficient so long as the particle is absorbing. Even if the particle's complex refractive index has a relatively small imaginary component, absorption will dominate over

scatter at longer wavelengths because absorption efficiency factors have an explicit inverse wavelength dependence, while scatter eficiency factors have an explicit inverse fourth-power wavelength dependence. When the complex refractive index does not change rapidly with wavelength, the inverse wavelength dependence appears directly in the spectra for an absorbing particle, just as the inverse fourth-power wavelength dependence appears in the spectra for a dielectric particle. On the other hand, there will be peaks in the extinction spectra in wavelength regions of anomolous dispersion.

A theory treating nonspherical Rayleigh particles has been in existence since before the turn of the century and was developed by Rayleigh to predict the optical properties of small elliposoidal particles. Gans extended the theory to randomly oriented spheroids, elliposoids of revolution. The spheroid is a particularly valuable yet simple shape with the flexibility of predicting everything from rods, in the limit of high aspect ratio prolate spheroids, to discs, in the limit of high aspect ratio oblate spheroids. The extinction coefficient for an absorbing randomly oriented spheroid is

$$\alpha = \frac{8\pi^3}{3\lambda} \operatorname{Im} \left[\frac{1 - n^2}{4\pi + (n^2-1)P} + \frac{2\,(1 - n^2)}{4\pi + (n^2 - 1)\,(4\pi - P)/2} \right] \tag{5}$$

where P is the depolarization factor for the one unequal axis, n is the complex refractive index, and λ is the wavelength.

3. EXPERIMENTAL VERIFICATION

Aerosols are produced from powders by means of a nozzle which aspirates particles into a shear field produced by a sonic flow of air. This shear field has been sufficient to deagglomerate the particles without grinding individual particles against one another, as would occur in an air mill, to produce particles smaller than those originally manufactured. This stream of deagglomerated particles is injected into a 6-cubic-meter chamber, and the transmittance is measured using a Barnes Radiometer to continuously scan wavelengths throughout the mid infrared. In addition, the transmittance is measured at two laser lines, one in the visible and another in the near infrared. Filter samples of aerosol material are collected to determine mass concentrations and then, with the pathlength known, extinction coefficient spectra are calculated. This procedure has been used to produce the spectra in figures 1 and 2, where subsequent scans taken at 5-minute intervals lie at progressively higher extinction values. In figure 1 a graphite flake powder having a greater thickness than the graphite flake in figure 2 produced a flat spectra predicted for the averaged resonance region. The spectral scans in figure 1 made at later times represent progressively thinner flakes, as the thicker flakes fall out of the air first. In figure 2 the sequential scans are clumped together because the average flake thickness is smaller and should require more time to fall then those in figure 1. If the time required to fall is tripled, then the spectra in the second figure would be separated by only one-third the distance in the first figure, for example. This of course ignores the effect of initial delay due to falling through the vertical distance above the beam.

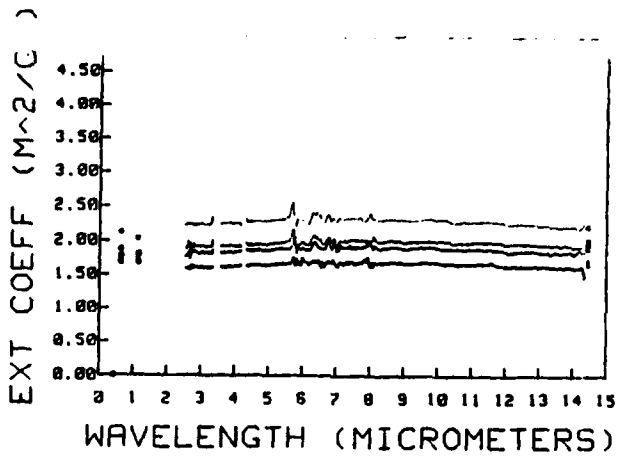

Figure 1. Dixon Graphite HPN-5

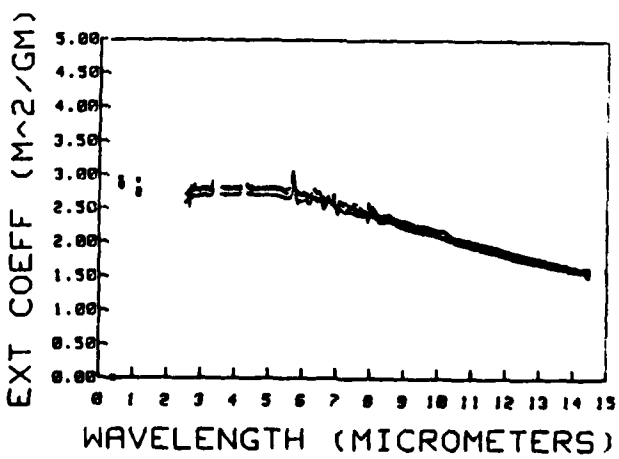

Figure 2. Asbury Graphite Micro #950

In figure 2, the transition between flat and Rayleigh inverse wavelength spectra occurs at wavelengths around 5 microns. This occurs in figure 2, but not in figure 1, for two reasons. First, the flake particles in figure 2 are nearly half the thickness of those in figure 1. Second, the major flake dimensions in figure 2 are only about a tenth those in figure 1, so that the transition at a fixed thickness would occur at shorter wavelengths due to the smaller aspect ratio.

4. CONCLUSION

Extinction spectra for a collection of particles in a cloud have been demonstrated to be either flat, in the case of larger particles, or to have structure superimposed onto an inverse wavelength dependence, in the case of smaller particles. Flat spectra occur as a result of the superposition of numerous, individual particle-resonant structure spectra where the particles have a variety of sizes and orientations, both of which influence the wavelength position of resonant peaks. Smaller particle spectra is structured where refractive index changes take place rapidly with wavelength. An underlying inverse wavelength dependence appears in small, absorbing particle spectra because the extinction efficiency factor has such explicit dependence. The transition wavelengths lying between the short wavelength, flat and longer wavelength, Rayleigh-structured spectral regions are determined by particle size and shape, which move the plateau up and down, as well as by the refractive index and aspect ratio, which move the Rayleigh spectra up and down. Analytic expressions were given for a variety of shapes, both monodisperse and polydisperse, outside the Rayleigh region. An analytic expression was also given for the spheroid in the Rayleigh region.

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