(Received June 15, 1978)

APPLICATION OF LIGHT SCATTERING FROM DIELECTRIC CYLINDER BASED UPON MIE AND RAYLEIGH-GANS-BORN THEORIES TO POLYMER SYSTEMS. II.

SCATTERING FROM A COAXIAL AND CIRCULAR CYLINDER.

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Mie theory was applied to the light scattering from a coaxial and circular cylinder. The methods were proposed to evaluate the parameters of the cylinders from the light scattering experiment. The light scattering experiments were performed on the E-glass fiber of 2.5 μm in radius coated and uncoated with polystyrene, and the results were compared to the calculated ones. It was shown that the parameters obtained from the light scattering experiments agree with those obtained by other methods like an optical microscope investigation.

The theoretical expression for the light scattering from the coaxial cylinder in Rayleigh-Gans-Born (R.G.B.) criterion was also derived and compared with the Mie results.

1. INTRODUCTION

The fiber reinforced plastics (FRP) have been developed as a fundamental structure material. As for the fiber material, glass fibers of micron order in radius are used most popularly. The orientation of the fiber in the matrix polymer and the compatibility to the polymer are the important factors for the practical applications. Light scattering studies on these fibers will provide a powerful method for studying the FRP properties.

A light scattering apparatus with a laser as a light source makes it possible to study the fibers with fairly large radius compared with the wavelength of the incident light.

The method has been applied to polymeric systems in the studies of the supermolecular structures of crystalline polymers⁽¹⁾, in the determination of the anisotropy existing in the polymer system^(2,3) and of molecular weight in dilute solution^(4,5). In these studies, the light scattering can be described by Rayleigh-Gans-Born (R.G.B.) approximation^(6,7), where the conditions

$$|m_r - 1| \ll 1 \quad (1)$$

$$2k_0a|m_r - 1| \ll 1 \quad (2)$$

must be satisfied; m_r is the ratio of refractive indices of the scattering particles and the surrounding medium, a is the particle size, and k_0 is the wavenumber of the incident light in vacuo. The polymer systems described above satisfy the criteria (1) and (2). However, for a wider application of the light scattering technique, the restrictions (1) and (2) must be removed (Mie scattering)^(8,9).

In a previous article⁽¹⁰⁾, we discussed the Mie and R.G.B. scattering from an infinitely long, simple (not coaxial) and circular cylinder with a radius much larger than the wavelength of light. In this paper, we will discuss the Mie scattering from a coaxial circular cylinder. The scattering from the coaxial cylinder with fairly large radius compared with the wavelength has many practically important applications, e.g., the scattering from a glass fiber coated with a silane coupling agents to improve the compatibility of the fiber to the matrix polymer, the scattering from crazes produced in the periphery of the fiber in the FRP subjected to strain, the scattering from coated

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optical fibers, and the scattering from voids appeared in the interfaces between two polymers in strained polymer blend.

In this article we will discuss the Mie scattering from a coaxial circular cylinder having size of order of microns, which is the size frequently encountered in practical plastic industries.

The corresponding R.G.B. scattering for a coaxial cylinder was also derived and compared with the Mie results. Moreover, a method was proposed to determine the structural parameters of the cylinders based upon the Mie and R.G.B. theory.

2. THEORIES

2.1 R.G.B. Scattering by an Infinitely Long Coaxial Cylinder.

Let us first consider the R.G.B. scattering from an infinitely long coaxial and circular cylinder of inner and outer radii of R_1 and R_2 with refractive indices m_1 and m_2 , respectively. The cylinder is perfectly oriented parallel to z -axis and embedded in isotropic medium of refractive index m_3 as shown in Figure-1. We consider the equatorial scattering i.e., the scattering in the plane of x - o - y , (i.e., azimuthal angle $\mu = 90^\circ$) as a function of the scattering angle (Figure-2). The V_v scattering is then given by

Fig-1. The coordinate system for a coaxial cylinder and definitions of parameters (R_1 , R_2 , m_1 , m_2 , m_3).

Fig-2. The coordinate system for the scattering system and definitions of θ and μ .

$$E_{V_v}(\theta) = \frac{k_0^2}{R} \int (\mathbf{M} \cdot \mathbf{O})_{V_v} \exp(i\mathbf{h} \cdot \mathbf{r}) d\mathbf{r} \quad (3)$$

where $\mathbf{M} = \alpha(\mathbf{r})E_0\mathbf{e}_3$, $\mathbf{O} = \mathbf{e}_3$, $\alpha(\mathbf{r})$ is the polarizability at point $\mathbf{r}$, $\mathbf{e}_3$ is the unit vector along the z -axis, and $k_0 = 2\pi/\lambda_0$ is the propagation constant of light in vacuo. The cylinder is assumed to be optically isotropic. Therefore

$$(\mathbf{M} \cdot \mathbf{O})_{V_v} = \alpha(\mathbf{r})E_0 \quad (4)$$

where E_0 is the electric field of incident beam. Using the cylindrical coordinate of Figure-1, it follows that

$$\mathbf{r} = \rho(\mathbf{e}_1 \cos \phi + \mathbf{e}_2 \sin \phi) + z\mathbf{e}_3 \quad (5)$$

and

$$\mathbf{h} = k(\mathbf{s}' - \mathbf{s}_0) = k[(1 - \cos \theta)\mathbf{e}_1 - \mathbf{e}_2 \sin \theta], \quad k = 2\pi/\lambda \quad (6)$$

where $\mathbf{e}_1$ and $\mathbf{e}_2$ are the unit vectors along x and y -axes, respectively, $\mathbf{s}'$ and $\mathbf{s}_0$ are the unit vectors along the propagation directions of the scattered and incident beams, respectively, and λ is the wavelength of light in the medium.

From eqs (3) to (6), it follows that

$$E_{V_v}(\theta) = (\text{const}) \int_0^\infty m(\rho) J_0(2k\rho \sin(\theta/2)) \rho d\rho \quad (7)$$

where $\alpha(\mathbf{r})$ or $m(\mathbf{r})$ (the local refractive index) is assumed to be dependent upon ρ only, $J_0(z)$ is the first kind Bessel function of order zero, and the constant is related to the absolute intensity. After integration, we obtain an expression for the V_v scattered intensity for the coaxial cylinder,

$$I_{V_v} = (\text{const}) \left\{ (m_1 - m_2) \frac{J_1(U_1)}{U_1} \right\}$$

$$+(m_2 - m_3) \left(\frac{R_2}{R_1} \right)^2 \frac{J_1(U_2)}{U_2} \Big\}^2 \quad (8)$$

where

$$\begin{aligned} U_1 &= (4\pi/\lambda) \cdot R_1 \sin(\theta/2), \\ U_2 &= (4\pi/\lambda) \cdot R_2 \sin(\theta/2) \end{aligned} \quad (9)$$

The cylinder is assumed to be homogeneous with constant m_1 and m_2 .

2.2 Mie Scattering by an Infinitely Long Coaxial Cylinder.

Let us consider Mie scattering for the same cylinder under the same condition as above. The solution of the Maxwell equation for the scattered electromagnetic field must be obtained under proper boundary conditions⁽⁹⁾. If U is the solution of the scalar wave equation satisfying the Maxwell equations, U in each region of Figure-1 is given by

$$U = \sum_{n=-\infty}^{\infty} F_n (B'_n J_n(m_1 k_0 \rho)), \quad \text{for region (i),} \quad (10)$$

$$U = \sum_{n=-\infty}^{\infty} F_n (B''_n J_n(m_2 k_0 \rho) - b''_n H_n(m_2 k_0 \rho)), \quad \text{for region (ii),} \quad (11)$$

$$U = \sum_{n=-\infty}^{\infty} F_n (J_n(m_3 k_0 \rho) - b_n H_n(m_3 k_0 \rho)), \quad \text{for region (iii),} \quad (12)$$

where

$$F_n = (-1)^n \exp i(n\theta + \omega t) \quad (13)$$

and ω is the angular frequency of the incident field. The functions J_n and H_n are the first kind Bessel and the second kind Hankel functions of the order n , respectively. The constant B'_n , B''_n , b''_n , and b_n are determined by the boundary conditions for the electromagnetic field that mU and $m(\partial U/\partial r)$ should be continuous at each boundary.

The constant b_n in the region (iii) is associated with the scattered intensity at far field. Defining α_1 and α_2 as

$$\alpha_1 = k_0 R_1, \quad \alpha_2 = k_0 R_2,$$

then b_n is given by

$$b_n = \begin{vmatrix} J_n(m_3 \alpha_2) & H_n(m_3 \alpha_2) & J_n(m_2 \alpha_2) & 0 \\ m_3 J'_n(m_3 \alpha_2) & m_2 H'_n(m_2 \alpha_2) & m_2 J'_n(m_2 \alpha_2) & 0 \\ 0 & H_n(m_2 \alpha_1) & J_n(m_2 \alpha_1) & J_n(m_1 \alpha_1) \\ 0 & m_2 H_n(m_2 \alpha_1) & m_2 J'_n(m_2 \alpha_1) & m_1 J'_n(m_1 \alpha_1) \end{vmatrix} \quad (14)$$

At far field the Hankel function is approximated by

$$H_n(m_3 k_0 \rho) = (2/(\pi m_3 k_0 \rho))^{1/2} \times \exp(-i(m_3 k_0 \rho - \frac{(2n+1)\pi}{4})) \quad (15)$$

and the incident wave propagating along x-axis is given by

$$U = \exp(-i(m_3 k_0 x - \omega t)) \quad (16)$$

Therefore the scattered wave at far field is given by

$$U = \sqrt{\frac{2}{\pi m_3 k_0 \rho}} \exp(-i(m_3 k_0 \rho - \omega t - \frac{3\pi}{4})) \times \sum_{n=-\infty}^{\infty} b_n \exp(in\theta) \quad (17)$$

The z component of the electric vector, E_z , is proportional to U , and the intensity I is proportional to $|U|^2$

The scattered intensity $I_{V_v}(\theta)$ under the V_v polarization is then given from equations (12) to (17) by

$$\begin{aligned} I_{V_v}(\theta)/I_0 &= (\lambda_0/\pi^2 \rho m_3) \left| \sum_{n=-\infty}^{\infty} b_n \exp(in\theta) \right|^2 \\ &= (\lambda_0/\pi^2 \rho m_3) \left| b_0 + 2 \sum_{n=1}^{\infty} b_n \cos n\theta \right|^2 \end{aligned} \quad (18)$$

where I_0 is the incident intensity.

The distribution of the scattered intensity can be numerically calculated from equations (14) and (18), if m_1 , m_2 , m_3 , R_1 and R_2 are all known.

The scattering efficiency Q which is the scattering cross section divided by the geometrical cross section of the scattering object is obtained by integrating equation (18) in whole directions.

$$Q = \frac{\lambda_0}{\pi R_2} (|b_0|^2 + 2 \sum_{n=1}^{\infty} |b_n|^2) \quad (19)$$

3. RESULTS AND DISCUSSIONS

3.1 Numerical Calculations.

The coefficients b_n (equation (14)) for the V_v light scattering were obtained from a similar method as described in our previous work⁽¹⁰⁾, though the computation is much more complicated than for the simple cylinder. The Bessel functions $J_n(x)$ and $H_n(x)$ were evaluated with the Miller's method⁽¹¹⁾. Then the scattering intensity was calculated at each 0.5° step in scattering angles. In order to obtain the scattering intensity distribution from the simple cylinder, m_2 should be

set equal to m_1 ($m_1=m_2=m$) and R_1 may be arbitrarily assigned ($R_1 \leq R_2$). The calculated results were compared to the published data for some special values of parameters⁽¹²⁾, and a complete agreement was obtained.

The R.G.B. scattering from the coaxial cylinder can be numerically calculated based upon equations (8) and (9). For values of the parameters for which the R.G.B. criteria are satisfied, the Mie scattering coincides with the R.G.B. scattering, which verifies the validity of equation (8).

3.2 Effect of Structural Parameters on Mie Scattering.

The Mie equation (18) contains five parameters, the inner (R_1) and outer cylinder radius (R_2), the refractive indices of the inner and outer cylinder (m_1) and (m_2) respectively, and the refractive index of the medium (m_3). Equation (18) cannot be expressed by some reduced parameters as in the R.G.B. scattering (U_1 or U_2 in equation (8)). Each set of parameters gives a different pattern of scattering. In this section we attempted to obtain the typical light scattering

intensity distributions with these parameters and to find the method of determining the parameters. The calculations were performed for the cylinders having size of order of microns, because the parameters of this size have many practical applications. The wavelength is assumed to be $\lambda_0 = 0.6328 \mu\text{m}$, the wavelength of a He-Ne laser. Table-1 summarize the set of parameters for each figures.

(1) Effect of varying the size of inner cylinder R_1 :

In Figure-3, the effect of varying the inner cylinder radius R_1 is shown. The set of parameters are ($R_1, R_2=5.0 \mu\text{m}, m_1=1.548, m_2=1.6, m_3=1.0$), where R_1 was varied from 2.5 (μm) (a), 3.0 (b), 4.0 (c) to 5.0 (d). Case (d) corresponds to a simple cylinder. The general behavior of each curve does not change so much, except at small angle region, where the angles between two adjacent maxima $\Delta\theta_i$ varies in a complicated manner depending upon R_1 value. At large scattering angles, however, the separation angles $\Delta\theta_i$ of all curves for various R_1 values coincide almost completely for the

Table 1. The set of parameters (R_1, R_2, m_1, m_2, m_3) for each Figure.

Variables	curve number	$R_1 (\mu\text{m})$	$R_2 (\mu\text{m})$	m_1	m_2	m_3	Comments
R_1 (Fig-3)	a	2.5	5.0	1.548	1.6	1.0	simple cylinder
	b	3.0	5.0	1.548	1.6	1.0	
	c	4.0	5.0	1.548	1.6	1.0	
	d	5.0	5.0	1.548	1.6	1.0	
R_2 (Fig-4)	e	2.5	2.5	1.548	1.6	1.0	simple cylinder
	f	2.5	3.0	1.548	1.6	1.0	same to (a)
	g	2.5	4.0	1.548	1.6	1.0	
	h	2.5	5.0	1.548	1.6	1.0	
m_1 (Fig-6)	i	2.5	4.0	1.0	1.6	1.0	hollow cylinder
	j	2.5	4.0	1.2	1.6	1.0	same to (g) simple cylinder
	k	2.5	4.0	1.548	1.6	1.0	
	l	2.5	4.0	1.6	1.6	1.0	
m_2 (Fig-7)	m	2.5	4.0	1.8	1.6	1.0	simple cylinder (e)
	n	2.5	4.0	1.548	1.0	1.0	
	o	2.5	4.0	1.548	1.2	1.0	
	p	2.5	4.0	1.548	1.6	1.0	
m_3 (Fig-9)	q	2.5	4.0	1.548	2.0	1.0	same to (g)
	r	2.5	4.0	1.548	1.6	1.0	same to (g)
	s	2.5	4.0	1.548	1.6	1.4	
	t	2.5	4.0	1.548	1.6	1.8	
	u	2.5	4.0	1.548	1.6	2.0	

($\lambda_0 = 0.6328 \mu\text{m}$)

Fig-3. Mie scattering for a various size of the inner cylinder R_1 .

particular set of parameters (R_2 , m_1 , m_2 , m_3) close to the experimental condition. The separation angles at scattering angles larger than 30° are independent of the inner radius when the difference of the refractive indices between the inner and outer cylinders is relatively small. The angular dependence of the scattering pattern is primarily determined by the outer radius in this case. The separation angle gradually decreases with the scattering angle.

The curve (d) corresponds to a simple cylinder of radius $5.0 \mu\text{m}$. No discontinuous changes in the intensity distribution can be seen by changing the scatterer from the coaxial cylinder to the simple cylinder.

(2) Effect of varying the size of outer cylinder R_2 ;

Figure-4 shows the variation of the Mie scattering with the outer cylinder radius R_2 . The parameters are ($R_1=2.5 \mu\text{m}$, R_2 , $m_1=1.548$, $m_2=1.6$, $m_3=1.0$), where R_2 is $2.5 (\mu\text{m})$ (curve (e) corresponding to a simple cylinder), 3.0 (f), 4.0 (g) and 5.0 (h). Figure-5 shows the corresponding angular separations $\Delta\theta_i$ as a function of order of the scattering maximum i . There is no discontinuous change in the scattering curves by changing the scatterer from a simple cylinder (curve (e)) to a coaxial cylinder (curve (f)). The angular separation decreases as the radius of the cylinder becomes large. The numbers of the peaks within a certain range of scattering angle, depend primarily upon

Fig-4. Mie scattering for various size of the outer cylinder R_2 .

Fig-5. The separation angles between two adjacent maximum peaks as a function of order of scattering maxima for various values of R_2 .

the outer radius rather than the inner radius, when m_1 and m_2 are close as in this case.

If the refractive index, the radius of the inner cylinder and the refractive index of the sheath are known, the coat thickness of the sheath material can be estimated by fitting calculated and measured intensity distribution or by fitting the measured separation angle $\Delta\theta_i$ with the calculated $\Delta\theta_i$ as shown in Figure-5.

A remarkable change of the scattering with R_2 can be also found at the first intensity minimum, which is a plateau in curve (e) ($R_2=2.5 \mu\text{m}$) and a shallow minimum in curve (f) ($R_2=3.0 \mu\text{m}$) and a sharp and deep minimum in curve (h) ($R_2=5.0 \mu\text{m}$). From the depth of the first minimum, it may be possible to determine the thickness of the sheath. In curve (h), for which $R_2=5.0 \mu\text{m}$, the scattering curve of the coaxial cylinder becomes close to that of simple cylinder of $R_1=5 \mu\text{m}$

as previously reported⁽¹⁰⁾ or as shown in curve (d) in Figure-3. The small differences between the two come from the difference of the optical thickness corresponding to the sheath part ($R_2 - R_1 = 2.5 \mu\text{m}$, where the refractive index of curve (h) is 1.60, and that of curve (d) is 1.548).

(3) Effect of varying the refractive index m_1 ;

Figure-6 shows the Mie scattering as a function of the refractive index of the inner cylinder m_1 . The parameters are ($R_1 = 2.5 \mu\text{m}$, $R_2 = 4.0 \mu\text{m}$, m_1 , $m_2 = 1.6$, $m_3 = 1.0$), where $m_1 = 1.0$ (curve (i), corresponding to a hollow cylinder), 1.2 (j), 1.548 (k, E-glass), 1.6 (l, corresponding to a simple cylinder) and 1.8 (n).

When the difference between m_1 and m_2 becomes large (curve (i)) $m = m_2 - m_1 = 0.6$, or curve (j) $m = 0.4$, the scattering curves become complicated. However when the difference becomes small (curve (k) (l) or (n)), the intensity distribution curves become simple. The coaxial cylinders behave, as if they were a simple cylinder with the radius R_2 , and the refractive index m_1 (or m_2).

The slow modulations or beatings of the scattering maxima in (i) and (j) curves (occurring at almost the same scattering angles) may be attributed to the parameters ($R_2 - R_1$) and m_2 corresponding to the sheath thickness.

(4) Effect of varying the refractive index m_2 ;

Figure-7 shows the Mie scattering as a function of the refractive index of the outer cylinder, m_2 . The parameters are ($R_1 = 2.5 \mu\text{m}$, $R_2 = 4.0 \mu\text{m}$, $m_1 = 1.548$, m_2 , $m_3 = 1.0$), where m_2 is 1.0 (curve (n)),

Fig-6. Mie scattering for various values of refractive indices of the inner cylinder m_1 .

a simple cylinder), 1.2 (o), 1.6 (p), and 2.0 (q). When the refractive index m_2 is close to m_3 (curve (n)) or m_1 (curve (p)), the coaxial cylinder approaches a simple cylinder, and the scattering pattern shows a simple curve, but when the difference of the refractive indices is large, the scattering curves become much complicated (curve (o) or (q)). The curve (n) where m_2 equals to m_3 , shows a light scattering pattern of a simple cylinder with a radius $2.5 \mu\text{m}$. The coaxial cylinders (o), (p) and (q) which are no longer simple cylinders show more peaks than the simple cylinder.

The angular separations of these curves as a function of order of the scattering maxima are shown in Figure-8. The separations for the curves (n), (p) and (q) show a smooth curve and reach to the limiting values, but the cylinder (o) shows

Fig-7. Mie scattering for various values of refractive indices of the outer cylinder m_2 .

Fig-8. The separation angles between two adjacent maximum peaks as a function of order of maxima for various values of m_2 .

irregular angular separations.

(5) Effect of varying the refractive index m_3 ;

Figure-9 shows the Mie scattering as a function of the refractive index of the medium, m_3 . The parameters are ($R_1=2.5 \mu\text{m}$, $R_2=4.0 \mu\text{m}$, $m_1=1.548$, $m_2=1.6$, m_3), where m_3 is 1.0 (curve (r)), 1.4 (s), 1.8 (t) and 2.0 (u).

For a case where m_3 value is less than m_2 (curve (r) and (s)) the intensity distribution of the scattered light shows relatively regular oscillations and the angular separation decreases with increasing m_3 (Figure-10). The separation angle becomes almost constant for the higher order peaks. For a case where m_3 value is larger than m_2 (curve (t) or (u)), abrupt irregularities around the angle less than 40° appear, though the angular separation decreases with increasing m_3 value. At the scatter-

Fig-9. Mie scattering for various values of refractive indices of the medium m_3 .

Fig-10. The separation angles between two adjacent maximum peaks as a function of order of maxima for various values of m_3 .

ing angles larger than 40° , however, the angular separation becomes almost constant, the value of which depends upon m_3 . By observing the abrupt change of the scattering pattern from the regular to the irregular oscillation pattern, we may decide the refractive index of medium, if the values of the other parameters of the cylinder are known, or on the contrary, the refractive index of the cylinder may be determined by immersing the cylinder with several liquids of known refractive indices. When the cylinder is simple, its refractive index can be determined by measuring the refractive index of the immersion liquids which give rise to the minimum scattered intensity⁽¹⁰⁾.

3.3 Comparison of Mie Scattering with R.G.B. Scattering.

In Figure-11 are shown the calculated scattering curves for a coaxial cylinder with the parameters ($R_1=2.5 \mu\text{m}$, $R_2=4.0 \mu\text{m}$, $m_1=1.548$, $m_2=1.54$, $m_3=1.548$). The calculations were performed based on R.G.B. theory as well as Mie theory. The difference of the refractive indices between the outer cylinder and the inner cylinder or the medium ($\Delta m=(m_2-m_1)/m_1$) is 0.52% and the quantity of equation (2), $2k_0 \cdot a |m_r - 1|$, is 0.41, for which the R.G.B. criterion is supposed to be satisfied. The agreement between the Mie and R.G.B. curves are fairly good, though there are small discrepancies between them. The discrepancies are expected to vanish, if the difference of

Fig-11. Calculated Mie and R.G.B. scattering curves for a coaxial cylinder with the parameters (2.5, 4.0, 1.548, 1.54, 1.548).

Fig-12. Calculated Mie and R.G.B. scattering curves for a coaxial cylinder with the parameters (2.5, 4.0, 1.548, 1.5, 1.548).

the refractive indices becomes even smaller, which was found to be true.

In Figure-12 are shown Mie and R.G.B. curves for the parameters ($R_1=2.5 \mu\text{m}$, $R_2=4.0 \mu\text{m}$, $m_1=1.548$, $m_2=1.5$, $m_3=1.548$). The quantity of equation (2) is 2.54. In this case, the coincidence of the two curves are no more good. The scattering angle of each maximum is identical for both Mie and R.G.B. scattering. The slow modulation of the scattering curve as clearly seen in the R.G.B. scattering becomes less remarkable in the Mie scattering. It is concluded that the R.G.B. equation cannot be adopted any more for this case in which the quantity $2k_0 \cdot a |m_1 - 1|$ is more than 2.54. Origin of the slow modulation (or beating) will be discussed in next section.

In Figure-12, the Mie scattering intensity varies with scattering angle with a smaller amplitude between the maximum and minimum intensity than the R.G.B. The large amplitude of the R.G.B. (in logarithmic scale) is attributed to the simple equation (8), consisting of just two oscillating terms $J_1(U_1)$ and $J_1(U_2)$. The small amplitude of the Mie curve may be due to the superposition of the many oscillating terms as seen in equation (18).

3.4 The Physical Meanings of the Slow Modulation or Beating of The R.G.B. Scattering Intensity Distribution.

Mie and R.G.B. scattering with the set of the

parameters ($R_1=2.5 \mu\text{m}$, $R_2=4 \mu\text{m}$, $m_1=1.548$, $m_2=1.54$, $m_3=1.548$) show almost the same scattering pattern (Figure-11), since R.G.B. criterion is satisfied in this case. The refractive index difference $(m_1 - m_2) = -(m_2 - m_3) = 0.008$, which is small enough, and the ratio of the inner and outer radii R_2/R_1 is 1.6. Parameter U_1 and U_2 of equation (8) can be expressed

$$U_1 = m R_1 a \quad (20)$$

$$U_2 = m R_2 a \quad (21)$$

where a is defined

$$a = \frac{4\pi}{\lambda_0} \sin \frac{\theta}{2} \quad (22)$$

For the special case where $m_1 \approx m_3$ the equation (8) can be rewritten by

$$I_{V_v}(\theta) = (m_1 - m_2)^2 \left[\frac{J_1(U_1)}{U_1} - \frac{R_2}{R_1} \frac{J_1(U_2)}{U_1} \right]^2 \quad (23)$$

The Bessel function $J_1(x)$ is expressed asymptotically⁽¹³⁾ for $3 \leq x < \infty$,

$$J_1(x) = x^{-1/2} f_1 \cos \theta_1 \quad (24)$$

$$f_1 = 0.79788456 + 0.00000156 \left(\frac{3}{x} \right) + 0.01659 \left(\frac{3}{x} \right)^2 + \dots$$

$$\theta_1 = x - 2.35619449 + 0.12499612 \left(\frac{3}{x} \right) + \dots$$

Taking the first term of f_1 and the first and the second terms of θ_1 for sufficiently large x , $J_1(x)$ is approximated

$$J_1(x) = \frac{\alpha \cos(x - \beta)}{x^{1/2}} \quad (25)$$

$$\alpha = 0.79788, \quad \beta = 2.35619$$

U_1 and $U_2 > 3.0$ is satisfied for the scattering angle larger than 6 degrees in this case.

Then I_{V_v} can be written

$$I_{V_v}(\theta) = \frac{(m_1 - m_2)^2 \alpha^2}{U_1^3} \{ \cos(U_1 - \beta) - (R_2/R_1)^{1/2} \cos(U_2 - \beta) \}^2 \quad (26)$$

Putting the term $(R_2/R_1)^{1/2} \approx (1.265)$ as unity, we obtain the approximate equation (27)

$$I_{V_v}(\theta) = \frac{4\alpha^2 (m_1 - m_2)^2}{U_1^3} \sin^2 \left(\frac{U_1 + U_2}{2} - \beta \right) \sin^2 \left(\frac{U_1 - U_2}{2} \right) \quad (27)$$

The term U_1^3 shows the decreasing intensity with the scattering angle, which is seen in every scattering intensity curve. The term $\sin[(U_1 + U_2)/2 - \beta]$ represents the maxima and minima in the oscilla-

tion pattern of the scattering intensity distribution. The maxima and minima appear at the angles satisfying equation (28).

$$\frac{U_1 + U_2}{2} - \beta = \frac{\pi}{2} n \quad (28)$$

or

$$\sin \frac{\theta}{2} = 0.01577n + 0.02366 \quad (29)$$

where I_{V_v} maximum emerges at an odd number n and I_{V_v} minimum emerges at an even number n .

The modulation or beat pattern on the scattering intensity distribution comes from the term $\sin(U_1 - U_2)/2$.

$$\frac{U_1 - U_2}{2} = \frac{\pi}{2} n \quad (30)$$

or

$$\sin(\theta/2) = 0.06908n \quad (31)$$

where an odd number n corresponds to the maximum of the modulation, and an even number n corresponds to the minimum.

The first term of equation (8) comes mainly from the light scattering by the inner cylinder, and the second term comes mainly from the outer cylinder. It is deduced from the discussions above that oscillating peaks appeared in Mie or R.G.B. scattering from a coaxial cylinder is due to the beating between the waves scattered from the two cylinders. The rapidly oscillating pattern is the term associated with an addition of the phases U_1 and U_2 of the two waves, whereas the slow modulation pattern is due to that associated with a subtraction of the phases of the two waves.

The relationship (26) or in a special case (28) and (30) can be utilized for determining the parameters of a coaxial cylinder from the light scattering pattern, if the R.G.B. criterion is satisfied. Equation (28) and (30) are written

$$m(R_1 + R_2) \sin \frac{\theta_m}{2} = \frac{\lambda_0}{2\pi} \left(\frac{\pi}{2} n + \beta \right), \quad n=1, 3, 5, 7, \dots \quad (32)$$

$$m(R_2 - R_1) \sin \frac{\theta_{bm}}{2} = \frac{\lambda_0}{4} n, \quad n=1, 3, 5, 7, \dots \quad (33)$$

where θ_m is the scattering angle at the maximum intensity, and θ_{bm} is the scattering angle at the maximum intensity of the beating pattern. The use of equation (32) and (33) determines the parameters m , R_1 or R_2 , if one of these values

are previously known.

3.5 Determination of the Thin Sheath Thickness of a Coaxial Cylinder.

Figure-13 and 14 show the calculated light scattering intensity as a function of the sheath thickness for a coaxial cylinder having a thin sheath. In all cases the inner cylinder is assumed to have radius 5.0 μm and the refractive index 1.548. The cylinder is coated with a very thin sheath material of refractive index 1.45, and is put in the air ($m_3=1.0$, Figure-13) and in the material

Fig-13. Calculated light scattering intensity as a function of the sheath thickness of a coaxial cylinder in the air ($m_3=1.0$) $R_1=5 \mu\text{m}$, $m_1=1.548$ and $m_2=1.45$.

Fig-14. Calculated light scattering intensity as a function of the sheath thickness of a coaxial cylinder in the medium of refractive index $m_3=1.550$. $R_1=5 \mu\text{m}$, $m_1=1.548$, and $m_2=1.45$.

of refractive index 1.550 (Figure-14). The sheath thickness are 0 (simple cylinder), 10 (Figure-14 only), 100 and 1000 (Å) respectively.

The immersion liquids brings the system near the R.G.B. criterion. In the reality, the former model (coaxial cylinder in the air) corresponds to an E-glass fiber coated with a silane coupling reagent such as n - β (amino ethyl)- γ (aminopropyl) methyl diethoxy silane or some cyclic epoxy silane ($m_2=1.45$) to improve the compatibility between the glass fiber and the polymer. In the latter model (coaxial cylinder in the medium of $m_3=1.550$) the same treated glass fibers are put in some polymer matrix, for example, an epoxy resin ($m_3=1.550$) (FRP).

The silane coated layer is said to be most effective, when the thickness is monomolecular layer. It is difficult to measure the thickness of such a thin layer with ordinary methods.

As in Figure-13, the difference of the scattering pattern from the coaxial cylinders with the sheath thickness of 0 Å and 100 Å is small. An increase of the sheath thickness from 100 Å to 1000 Å produces a slight shift of the scattering angles of the maximum intensity, but the scattering curve as a whole are not much affected.

The situation is different in case when the coaxial cylinder is immersed in the medium having the refractive index close to that of the inner cylinder as depicted in Figure-14. Even in 10 Å sheath thickness, the scattering at large angle region are definitely different from that of the simple cylinder and a precise angular distribution measurement will be utilized to determine the sheath thickness. The thicker the sheath, the stronger is the scattering intensity at large angle region, though the number of peaks does not

Table 2. The ratio of the maximum intensity as a function of the sheath thickness for the set of parameters (5, R_2 , 1.548, 1.45, 1.55).

sheath thickness (Å)	I_0/I_{17}	I_1/I_{17}	I_3/I_{10}
1	7.05×10^4	1.25×10^3	2.45×10
10	5.48×10^4	9.26×10^2	2.18×10
100	3.23×10^2	8.27×10	5.45
1000	2.86×10^2	2.13×10	3.94

$R_1=5.0 \mu\text{m}$, $m_1=1.548$, $m_2=1.45$, $m_3=1.55$

change so much.

The intensity ratio of the maximum intensity at zeroth, 1st, and 3rd maximum peaks to n th maximum peak I_0/I_n etc. are shown in table-2 as a function of the sheath thickness. When the refractive indices are known, the thickness of the coupling reagent layer may be strictly determined from the light scattering intensity ratio I_1/I_{17} or I_3/I_{10} . Accuracy of the evaluation of the sheath thickness increases by putting the coaxial cylinder in a matrix whose refractive index is close to that of the inner cylinder.

The Q value defined by equation (19) is the measure of the total scattering power or the transparency for a practical purpose. The value depends on sizes and refractive indices of the fiber, the sheath, and the surrounding matrix. The Q values are also listed in table-3. The transparency is expressed as a function of the sheath thickness. When the coated fiber is in a matrix (FRP), the thinner is the sheath, the material is more transparent. But when the coated fiber is in the air, the transparency is not much affected.

Table 3. Scattering efficiency as a function of the sheath thickness and the refractive index of the medium.

sheath thickness (Å)	$Q (m_3=1.55)$	$Q (m_3=1.0)$
0	0.0406	2.51
1	0.0407	2.51
10	0.0418	2.51
100	0.0545	2.46
1000	0.3513	2.26

$R_1=5.0 \mu\text{m}$, $m_1=1.548$, $m_2=1.45$, $m_3=1.0$ or 1.55

3.6 Parameter Determination for the Coaxial Cylinder from V_v Light Scattering.

Unfortunately the Mie equation is fairly complicated to derive a simple relationship to evaluate the parameters of scattering systems. The calculated curve for a set of parameters (R_1 , R_2 , m_1 , m_2 , m_3) is compared with and fitted to the experimental curve in terms of the peak positions and peak intensity. In the real experiment, some parameters in R_1 , R_2 , m_1 , m_2 or m_3 are known so that the estimation of the values for other parameters becomes easier. This method of fittings has enough accuracy.

Every parameter of a coaxial cylinder can be

determined, in principle, from light scattering experiments as follows;

- (1) m_1 can be determined at first by the minimum intensity method⁽¹⁰⁾ (i.e., by finding the refractive index of the liquid giving rise to minimum scattering intensity). For this experiment the sheath part must be first removed to obtain the inner cylinder only.
- (2) The radius R_1 can also be determined from the scattering angles of maximum intensity as discussed in the previous article⁽¹⁰⁾.
- (3) The measurements are performed for the coated cylinder immersed in several kinds of liquids of various refractive indices m_3 . The refractive index m_3 for which the calculated scattering pattern for a simple cylinder with parameters (R_1, m_1) in the medium of refractive index m_3 coincides to the experimental scattering pattern should be equal to the refractive index of the sheath material m_2 .
- (4) Then the calculated scattering curves of a coaxial cylinder with parameters $(R_1, R_2, m_1, m_2, m_3)$ (where R_1, m_1 and m_2 are all known and R_2 is assumed for several values, and m_3 may be 1.0, when the experiment is performed in the air) are compared with the experimental light scattering curves to determine the outer radius R_2 .

Though somewhat lengthy, this is a general principle to determine all values of parameters of a coaxial cylinder. The light scattering method is especially valuable in determining the refractive indices for such a thin sheath. When some of the parameters are predetermined, the determination becomes much simpler. The knowledge on the general trend of the scattering intensity distributions with the parameters (as discussed in section 3-2, 3 and 4) will facilitate the determination of the parameters.

4. APPLICATION OF THE LIGHT SCATTERING THEORY TO THE SIMPLE AND COATED GLASS FIBERS

Figure-15(a) shows an optical micrograph of a glass fiber (E-glass⁽¹⁴⁾, refractive index, $m_1=1.548$) of radius $2.5\ \mu\text{m}$, while Figure-15(b) shows the photograph of the same glass fiber coated with polystyrene. The coated layer is uniform and cylindrically symmetric. The corresponding photo-

graphic V_v light scattering patterns from the simple and coaxial cylinder are shown, respectively, in Figure-16(a) and 16(b). Both patterns show a strong streak in a direction perpendicular to the cylinder axis with many scattering maxima and minima. The coated sheath of polystyrene decreases the angular intervals of the adjacent scattering maxima and minima. Figure-17(a) and 17(b) show, respectively, the V_v scattering intensity distribution of the simple and coaxial fibers (open circle) measured by a photometrical apparatus together with the calculated curves (solid lines). As shown in Figure-16 and 17, the scattering from the simple and coaxial fibers show the strong maxima and minima depending upon the size R_1 and R_2 of the cylinder, refractive indices m_1 and m_2 , and the wavelength of the incident light.

From Figure-17 the separation angles $\Delta\theta_1$ in the measured scattered intensity distribution from

Fig-15. Optical micrographs of a glass fiber of radius $R_1=2.5\ \mu\text{m}$ (a), and the same glass fiber coated by polystyrene ($R_2=4.5\ \mu\text{m}$).

Fig-16. Photographic V_v light scattering pattern for the simple (a) and coaxial cylinders (b).

the coated cylinder can be estimated and plotted as a function of i , the order of the scattering maxima as shown in Figure-18 with the solid circles. A comparison of this result with those shown in Figure-5 give the outer radius R_2 of the coated cylinder approximately $4.5 \mu\text{m}$ for a given set of parameters $R_1=2.5 \mu\text{m}$, $m_1=1.548$, $m_2=1.6$ and $m_3=1.0$ (these parameters except for m_2 being estimated as described already). In order to confirm the estimated value ($R_2=4.5 \mu\text{m}$), the measured separation angles were more closely compared with the calculated results for various values of m_2 as in Figure-18. The fine agreement assures accuracy of the values and gives the estimated value of m_2 in the range from 1.58 to 1.62. The estimated R_2 and m_2 values agree, respectively,

Fig-17. Photometric V_v light scattering patterns of (a) the simple and (b) coaxial fibers (open circles) and the calculated curves (solid lines).

Fig-18. Separation angles for the glass fibers coated by polystyrene as a function of the refractive indices of polystyrene.

with the values obtained from the optical micrograph and the literature value ($m_2=1.59-1.60$)⁽¹⁵⁾.

In Figure-17(b), the calculated curve with the estimated parameters ($R_1=2.5 \mu\text{m}$, $R_2=4.5 \mu\text{m}$, $m_1=1.548$, $m_2=1.58$, $m_3=1.0$) is shown, which reproduces the experimental peak position very well. The agreement of the calculated Mie curve for a set of parameters ($R_1=2.5 \mu\text{m}$, $R_2=2.5 \mu\text{m}$, $m_1=1.548$, $m_2=1.548$, $m_3=1.0$) and the experimental curve for the simple cylinder is also good, as shown in Figure-17(a).

These results confirm the wide applicability of the Mie theory to the light scattering experiments.

5. CONCLUSION

The Mie equation for a coaxial cylinder is expressed by an infinite series, whereas the R.G.B. equation consists of only two terms concerning to the inner and outer cylinders. Any parameter of a coaxial cylinder can be principally determined by the combination of the Mie theory and the light scattering experiment. When the R.G.B. criterion is satisfied, i.e., $2k_0 \cdot a |m_r - 1| \leq 0.4$, the analysis of the light scattering phenomena becomes simpler. The application of the Mie or R.G.B. theory to the polymer coated or uncoated glass fibers shows the wide validity of the theory to the practical purpose.

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