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Improved thermal radiation extinction in metal coated polypropylen microfibers

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Abstract—If polypropylen microfibers with 2–3 μm diameters are coated with aluminum the infrared extinction increases by a factor of 20 as compared to the non-coated fibers. This has been shown by Mie scattering calculations and infrared optical transmission and reflection measurements using an integrating sphere. Large infrared extinction is important for the reduction of thermal radiative heat transfer in low density insulations. Insulation systems with Al-coated polypropylen microfiber fleeces can be expected to provide the same excellent insulation performance as natural downs.

1. INTRODUCTION

IN SLEEPING bags, beddings and cold weather clothing, fiber materials or natural feathers and downs are used in order to provide sufficient thermal insulation. The total thermal conductivity of such low density fillings is usually about 0.050 W m^{-1} K^{-1}, i.e. twice the one of still air (λ_{air} = 0.026 W m^{-1} K^{-1}). It has been shown that despite the large porosity, which can be higher than 99%, natural air convection does not occur within such insulations [1]. Also the conductivity via the solid structure of the fiber material can usually be neglected [1]. Thus radiative heat transfer and gaseous conduction are the two main effects responsible for heat losses.

Usually the finer the filling material the larger is the effective surface which blocks thermal radiation by absorption and scattering. It is well known that in the case of powder pigments an optimum particle diameter exists, which can be calculated according to Mie scattering theory [2]. This is also true for dielectric fibers, where the optimum fiber diameter lies in the range of the typical wavelengths of the thermal radiative spectrum (see for example [1, 3]). At room temperature an optimum fiber diameter for maximum extinction is about 5 μm for polyester fibers [1] and about 4 μm for natural keratin fibers; the latter are the structural units of downs [4]. The diameters of the hairs in fine goose downs are near the optimum fiber diameter for keratin. The maximum specific extinction e or effective blocking surface per unit of mass for polyester and downs is in the range e = 80–100 m^2 kg^{-1}.

For smaller dielectric fibers below the optimum diameter the radiation blocking efficiency rapidly decreases as the comparably large radiation wavelengths only weakly interact with such fibers. This is different, however, for metallic fibers or for metallized dielectric fibers [5]. Due to the electrical conductivity the fibers act like little antennae, which scatter and absorb radiation even if the fiber diameter is far smaller than the incoming wavelengths of the electromagnetic radiation.

For very fine metallized glass fiber samples with fiber diameters in the range of 0.1 μm specific extinction values $e > 1400$ m^2 kg^{-1} have been measured [6]. Despite this success it is rather difficult to produce sufficiently thin and homogeneous layers with these ultra-thin glass fibers or even to disperse single fibers in an appropriate matrix, which provides the mechanical stability and high porosity of the insulation. Thus this approach, to our knowledge, had no practical use until now.

In this paper we present measurements on the extinction properties of coated and non-coated polypropylen micro-fiber fleeces with fiber diameters in the range of 2 μm. Such fleeces are commercially produced with a very low mass per area down to 2 g m^{-2} and excellent homogeneity [7].

2. METHODS FOR DETERMINATION OF THE SPECIFIC EXTINCTION

According to the diffusion model for radiative transfer the blocking efficiency or specific extinction e and the filling density ρ determine the radiative conductivity

$$\lambda_r = \frac{16}{3} n^2 \frac{\sigma T_r^3}{e\rho}. \quad (1)$$

This equation only holds for optically thick insulations in which the photon mean free path is much smaller than the thickness of the insulating specimen. In low density insulations the effective index of refraction n is very close to 1 and can be neglected. T_r is the mean radiative temperature of the insulation and σ the Stefan–Boltzmann constant. There are three independent methods to determine the specific extinction e: total thermal conductivity measurements, Mie

NOMENCLATURE

a_i, b_i	complex scattering coefficients
d	fiber diameter
e	mass specific radiation extinction
g	anisotropy factor
m	complex index of refraction
m_A	mass per area
n	effective index of refraction within an insulation
$p(\Omega)$	scattering phase function at direction Ω
Q_a	relative absorption cross-section
Q_d	relative diffusion cross-section
Q_e	relative extinction cross-section
Q_{geom}	relative geometric cross-section
Q_s	relative scattering cross-section
R_∞	directional–hemispherical reflection of an optically thick sample
T	directional–hemispherical transmission
T_{d-d}	directional–directional transmission
T_r	mean radiation temperature within an insulation
x	dimensionless size parameter.

Greek symbols

θ	scattering angle
λ	total conductivity
λ_g	gas conductivity
λ_r	radiative conductivity
Λ	wavelength
ρ	insulation density
ρ_0	material density
σ	Stefan–Boltzmann constant
τ_0	optical thickness of sample
ϕ	fiber tilt angle
ω_0'	single scattering albedo
ω_0	albedo for diffusing radiation
Ω	solid angle.

scattering theory and infrared optical transmission and reflection measurements.

2.1. *Measurement of total conductivity* λ

As in low density insulations the solid conduction via the fibers usually can be neglected, the radiative conductivity λ_r is determined by subtracting the known air conductivity λ_g from the measured total conductivity λ:

$$\lambda_r \simeq \lambda - \lambda_g. \tag{2}$$

From equation (1) the specific extinction e is derived. Vice versa the total conductivity at temperature T_r may be calculated, if air conductivity λ_g, the density ρ of the insulation and the specific extinction e are known. If the insulation is not optically thick more complicated expressions for the radiative heat loss have to be used [8].

2.2. *Mie scattering theory*

Electro-magnetic scattering theory allows to calculate the relative cross-sections of fibers as a function of the fiber diameter d, the incoming wavelength Λ, the tilt angle ϕ of the fiber and the complex index of refraction m [2]. It can be shown that the relative radiation pressure cross-section is equivalent to the effective relative cross-section Q_d of diffusing radiation in a disperse medium, if forward scattering is accounted for in a correct way [9, 10]. The radiative diffusion cross-section Q_d is related to the extinction cross-section Q_e by

$$Q_d = (Q_a + Q_s) \cdot (1-g) = Q_e \cdot (1-g), \tag{3}$$

where Q_a is the absorption and Q_s the scattering cross-section. The anisotropy factor g is defined by

$$g = \frac{1}{4\pi} \int_\Omega p(\Omega) \cos(\theta)\, d\Omega, \tag{4}$$

where θ denotes the angle between the scattered and incoming radiation and Ω stands for the solid angle. The scattering phase function $p(\Omega)$ has been normalized to the single scattering albedo $\omega_0' = Q_s/Q_e$:

$$\frac{1}{4\pi} \int_\Omega p(\Omega)\, d\Omega = \omega_0'. \tag{5}$$

If, for example, the fiber lies perpendicular to the incoming radiation (tilt angle $\phi = 0$) the diffusion cross-section can be calculated according to

$$Q_d(x, m, \phi = 0) = \frac{1}{x} Re\left[a_0 + b_0 + 2 \cdot \sum_{n=1}^{\infty} a_n(1 - a_{n-1}^*) + b_n(1 - b_{n-1}^*) \right] \tag{6}$$

with $x = \pi \cdot d/\Lambda$: size parameter, m: complex index of refraction at wavelength Λ, $a_i(m, x)$, $b_i(m, x)$ complex scattering coefficients as defined in Bohren and Huffman [2] and *: symbol for conjugate complex.

The wavelength dependent cross-section $Q_d(\Lambda)$ finally has to be averaged over the thermal radiation spectrum at temperature T_r by using the Rosseland mean function [11].

In Fig. 1 the relative diffusion cross-section Q_d of randomly oriented polyester (PES), polypropylen (PP) and aluminum (Al) fibers as function of the fiber diameter d are depicted. They have been calculated from the corresponding complex index of refraction [3, 12, 13]. Above diameters of 10 μm both the cross-section of the dielectric and metallic fibers are almost

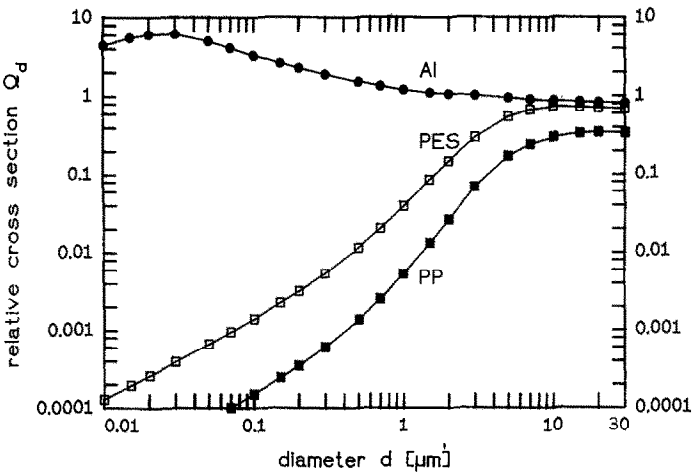

FIG. 1. Relative diffusion cross-section Q_d of randomly oriented aluminum (Al), polyester (PES) and polypropylen (PP) fibers as function of fiber diameter d for the thermal radiation spectrum at $T_r = 300$ K.

constant and near the geometric value of a randomly oriented, opaque fiber, where $Q(\phi) = \cos\phi$:

$$Q_{\text{geom}} = \int_{\phi=0}^{\pi/2} \cos\phi d(\sin\phi) = \frac{\pi}{4} \approx 0.785. \quad (7)$$

For smaller diameters, however, the relative cross-sections of polyester and polypropylen fibers decrease to very small values—in contrast to Al-fibers, where the relative cross-section even increases up to a value of $Q_d = 6$.

The specific extinction for diffusing radiation e can be related to the relative diffusion cross-section Q_d by

$$e = \frac{4}{\pi}\frac{1}{\rho_0}\frac{Q_d}{d}, \quad (8)$$

where ρ_0 denotes the density of the solid fibers. Figure 2 shows the theoretically calculated specific extinction e of PES and PP fibers and of Al-coated polypropylen fibers randomly oriented in space. It is assumed that the Al-coating is thick enough that the thermal radiation cannot penetrate the metallic layer. For ρ_0 we use the solid density of polypropylen (900 kg m^{-3}) neglecting the mass of the coating. In contrast to the non-coated dielectric PP and PES fibers the specific extinction e of the metallized fibers does not show a maximum and steadily increases with smaller fiber diameters.

2.3. *Reflection and transmission measurements*

Within certain limits the radiative transfer in cold, non-emitting samples is governed by the same cross-section Q_d as the diffusing thermal radiation in thermal insulations [10]. The diffusion cross-section Q_d already takes into account anisotropic forward scattering (equation (3)). In the radiation transfer theory Q_d is invariant against scaling and thus only the case of isotropic scattering must be considered.

Radiation transfer theory of an isotropic scattering medium provides a relation between directional–hemispherical reflection R_∞ of an optically thick

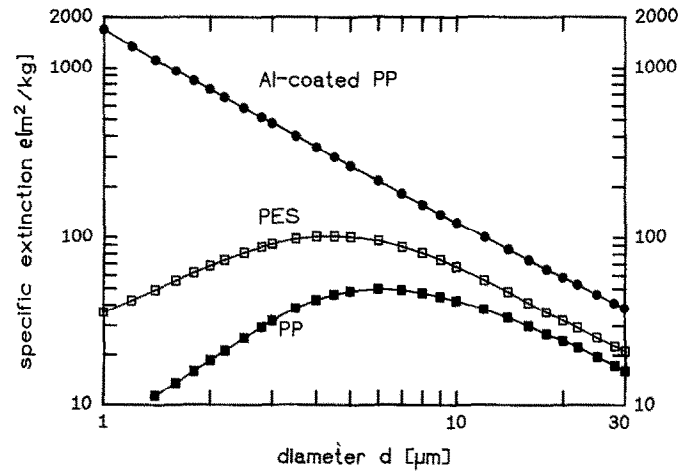

FIG. 2. Specific extinction e of randomly oriented Al-coated polypropylen (PP), non-coated polyester (PES) and polypropylen fibers as a function of fiber diameter d (Rosseland mean at $T_r = 300$ K).

sample and the scaled albedo ω_0. In the three flux approximation of Kaganer [14] the scaled albedo can be calculated according to

$$\omega_0 = \frac{5R_\infty \cdot (R_\infty + 4)}{(2R_\infty + 3) \cdot (4R_\infty + 1)}. \quad (9)$$

The scaled albedo for diffusion ω_0 is related to the single scattering albedo ω_0' of equation (5) by

$$\omega_0 = \frac{\omega_0' - g}{1 - g}. \quad (10)$$

Furthermore the directional–hemispherical transmission T depends on both the effective albedo ω_0 and the optical thickness for diffusion $\tau_0 = e \cdot m_A$ (m_A: mass per unit area). According to Kaganer [14] in the three flux approximation for a cold, isotropically scattering medium one gets the following relation:

$$T(\omega_0(R_\infty), \tau_0) = A \cdot \left[(e^{-\tau_0} - e^{-k\tau_0}) - B \cdot \frac{\left(\frac{e^{-\tau_0}}{5} - Be^{-k\tau_0}\right) \cdot (e^{k\tau_0} - e^{-k\tau_0})}{e^{k\tau_0} - (A/5)e^{-k\tau_0}} \right] + e^{-\tau_0} \quad (11)$$

with

$$A = \frac{\frac{3}{4}\omega_0}{1 - \frac{8}{5}\omega_0}, \quad B = \frac{1 - \frac{2}{3}k}{1 + \frac{2}{3}k}, \quad k = \frac{3}{2}\sqrt{\frac{1 - \omega_0}{1 - \omega_0/4}}$$

or

$$A = \frac{\frac{5}{4}R_\infty(4 + R_\infty)}{1 - 6R_\infty}, \quad B = \frac{5R_\infty}{4 + R_\infty} \quad \text{and} \quad k = \frac{1 - R_\infty}{2/3 + R_\infty}.$$

Hence from two directional–hemispherical measurements of reflection R_∞ and transmission T by using an integrating sphere one can derive the optical thickness τ_0 of diffusing radiation and, with the known mass per area m_A of the sample, the specific extinction e.

Table 1. Data of samples, mass per area per layer 4 g m^{-2}

Sample	Al-coating thickness [nm] Side 1	Side 2
A	0	0
B	20	0
C	80	0
D	100	0
E	100	100

In directional–directional transmission experiments (see Fig. 3) the transmission $T_{d\text{-}d}$ varies exponentially with optical thickness τ_0':

$$T_{d\,d} = \exp(-\tau_0') = \exp(-e' \cdot m_A). \quad (12)$$

The specific extinction e' is proportional to the extinction cross-section Q_e, which due to the effect of forward scattering usually is not the correct parameter in describing diffusing radiation. Only for purely absorbing particles or in the case of isotropic scattering are the relations $Q_d = Q_e$ and $e = e'$ valid.

In our experiments we used a FTIR-Fourier spectrometer in the wavelength range from 2.3 up to 40 μm. Directional–hemispherical measurements were performed with a gold-coated, integrating sphere from Labsphere with a diameter of 7.5 cm. We deposited different amounts of Al on single layers of PP-fleece by evaporation of aluminum. The data of the prepared samples are summarized in Table 1.

3. RESULTS

Figure 4 shows the measured reflection R_∞ of an optically thick sample of non-coated PP-layers (sample

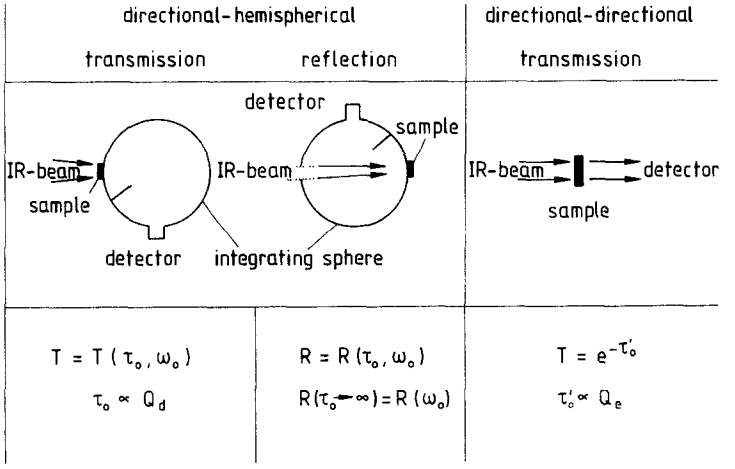

FIG. 3. Experimental set-up for directional–directional transmission and directional–hemispherical transmission/reflection measurements.

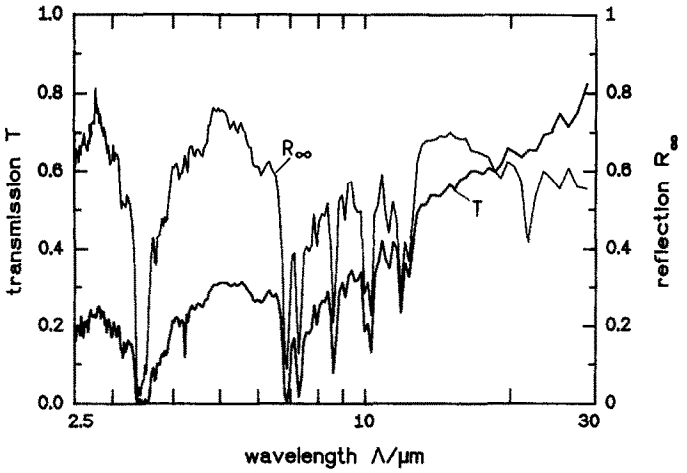

FIG. 4. Measured reflection R_∞ of an optically thick sample of PP-fleece layers and directional–hemispherical transmission T of seven layers of PP-fleece as function of wavelength Λ.

A) together with the corresponding transmission T of seven PP-layers as function of wavelength. From these data the specific extinction of the PP-fibers can be extracted. In Fig. 5 the latter is compared with the calculated specific extinction via Mie theory assuming a uniform distribution of three fiber diameters 2.0, 2.5 and 3.0 μm. Experimental and calculated values show a very good agreement up to wavelengths of 15 μm. Due to the small fiber thicknesses the extinction drops to low values beyond wavelengths of 15 μm, where the Rayleigh scattering theory for fibers predicts a $1/\Lambda^3$ behaviour for the scattering efficiency. As the thermal radiation spectrum for room temperature peaks at 10 μm and is still quite significant above 20 μm, the calculated overall mean of the spectral extinction values only amounts to $e \approx 30\ \mathrm{m^2\ kg^{-1}}$. Natural downs for comparison yield a higher specific extinction of at least $80\ \mathrm{m^2\ kg^{-1}}$ [1, 4].

From the reflection data according to equation (9) the effective albedo ω_0 for diffusion can also be calculated. The results are shown in Fig. 6 for both the uncoated (sample A) and the Al-coated PP-microfiber fleece (sample D). The albedo of the coated sample is close to 0.9 almost over the whole wavelength range, as is typical for metal fibers. In contrast the albedo of the pure PP sample A drops to low values within the absorption bands. This demonstrates that the Al-coating covers the non-coated PP-fibers efficiently. Outside of the absorption bands the albedo of the non-coated fibers reaches values beyond $\omega_0 = 0.98$; thus absorption here accounts for less than 2% of the total extinction.

The transmission T of one layer of coated PP-microfiber fleece varies with the thickness of the deposited Al-layer (Fig. 7). Only for Al thicknesses of at least 80 nm does the infrared transmission of a single PP-layer become lower than 20%.

From the transmission and reflection properties according to equations (9) and (12) the specific extinction for the coated PP-fleeces have been calculated (Fig. 8). The influence of the deposited Al-thickness is clearly recognizable. For both sample C and the double-coated sample E the specific extinction varies

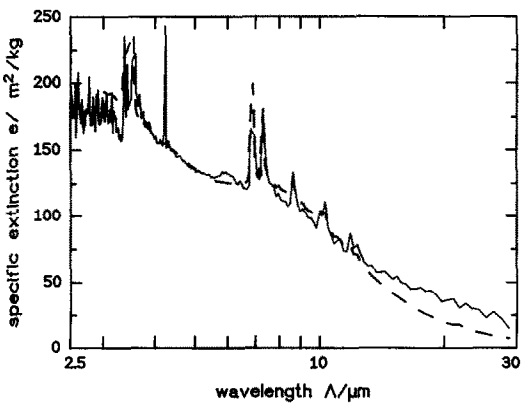

FIG. 5. Specific extinction e of non-coated PP-fibers vs wavelength Λ derived from transmission and reflection measurements (full line) compared with calculations according to Mie theory (dashed line).

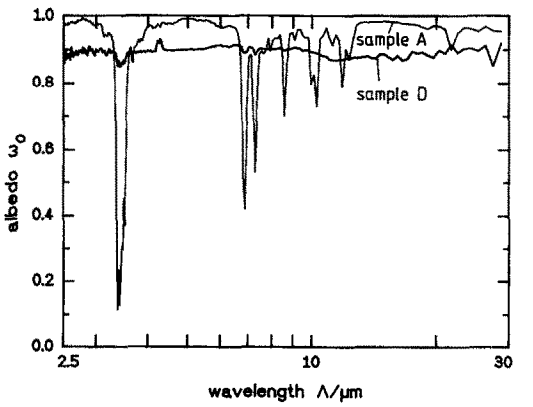

FIG. 6. Effective albedo ω_0 of non-coated PP-fibers (sample A) and Al-coated PP-fibers (sample D) vs wavelength Λ.

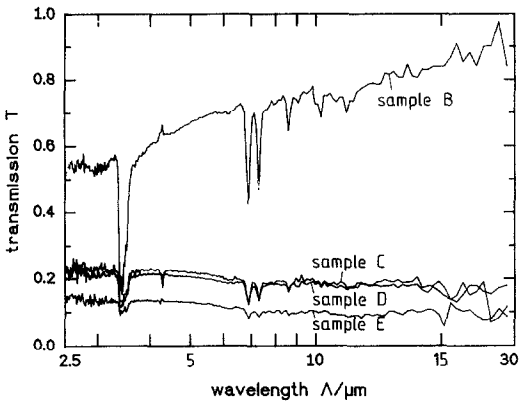

FIG. 7. Directional–hemispherical transmission T of one layer of Al-coated PP-fleece as function of wavelength; the parameter is the coating thickness: $B = 20$ nm, $C = 80$ nm, $D = 100$ nm, $E = 100$ nm on both sides of the fleece.

only smoothly with wavelength as it is expected for metal fibers. The mean extinction is in the order of 700 $m^2 kg^{-1}$ for sample C and 850 $m^2 kg^{-1}$ for sample E.

According to Fig. 2 Mie theory predicts a specific extinction between 500 and 800 $m^2 kg^{-1}$ for randomly oriented fibers between 2 and 3 μm diameter. For a fiber distribution with diameters of 2.0, 2.5 and 3.0 μm we get a good agreement between the measured spectral extinction values and the calculated ones, if we assume that 50% of the fibers lie in a plane perpendicularly oriented to the IR-beam (Fig. 8).

Though the Al-coating of sample E was applied on both sides, it did not enhance the extinction efficiency considerably compared to the single coating of samples C and D. Thus a single coating with 80 nm Al seems to be sufficient to cover the PP-fibers effectively.

4. CONCLUSIONS

We have shown that with metal coated PP-microfibers mass specific infrared extinction values can be reached, which are at least 10 times higher than for other non-coated dielectric fiber materials like polyester and PP-fibers. The infrared optical properties of coated and non-coated PP-fleeces described by Mie theory and experiments are in good agreement.

In order to make use of the large specific extinction of coated fibers one would have to distribute microfibers homogeneously within a coarser matrix structure. Low density insulations with metallized PP-microfibers dispersed as infrared opacifiers can be expected to have total conductivities around 0.030 $W m^{-1} K^{-1}$. A technical solution for the production of such an insulation system is not yet at hand.

However, the coated PP-layers may also be used simply as radiation shields within a thermal insulation. These low-weight shields are water vapor permeable and can be separated by layers of coarser fiber material. Thus a low density and at the same time a high thermal radiation extinction is provided.

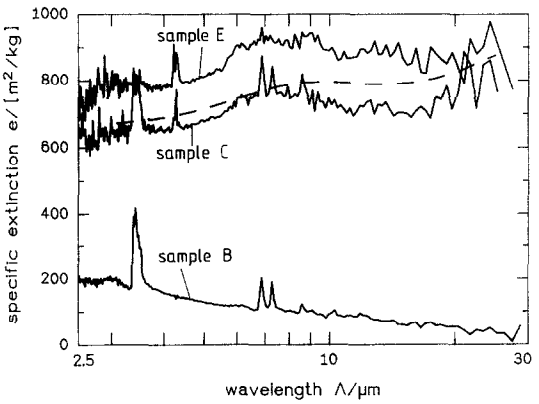

FIG. 8. Specific extinction e for samples B, C, E as derived from transmission and reflection measurement compared with Mie theory calculations (dashed line).

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