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Int. J. Heat Mass Transfer. Vol. 28, No. 7, pp. 1408–1411, 1985
Printed in Great Britain
0017-9310/85 $3.00 + 0.00
Pergamon Press Ltd.

Can thermal conductivity, λ, and extinction coefficient, E, be measured simultaneously?

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(*Received* 24 *October* 1984)

NOMENCLATURE

A, A_Λ	absorption coefficient at wavelength Λ [m^{-1}]
c_P	specific heat [J kg^{-1} K^{-1}]
D_0	total thickness of sample [m]
d	thickness of a layer in an extinction measurement [m^{-1}]
E, E_Λ	extinction coefficient at wavelength Λ [m^{-1}]
$\bar{E}, \bar{E}^*$	thickness-averaged value of E or E^*
E^*	effective value of the extinction coefficient (E corrected for anisotropic scattering)
I'_Λ	source function including scattered and remitted directional intensities at wavelength Λ [W m^{-2} μm^{-1} sr^{-1}]
i'_Λ	directional intensity at wavelength Λ [W m^{-2} μm^{-1} sr^{-1}]
K	K integral, equation (8b)
n	real part (effective value) of complex refractive index of porous medium
$p(\theta)$	scattering phase function at angle θ
$\dot{q}, \dot{q}_{sc}, \dot{q}_{rad}$	total, solid conductive and radiative heat flow [W m^{-2}]
r	fiber radius [μm]
S, S_Λ	scattering coefficient at wavelength Λ [m^{-1}]
T, T'	temperature [K]
T_1, T_2	temperatures of hot and cold wall [K]
T^{*3}	third power of radiative temperature, $(T_1^2+T_2^2)(T_1+T_2)$ [K^3]
t	time [s].

Greek symbols

Λ	wavelength [μm]
$\lambda, \lambda_{sc}, \lambda_{rad}$	total, solid conductive and radiative conductivity [W m^{-1} K^{-1}]
$\bar{\lambda}, \bar{\lambda}_{sc}$	thickness-averaged values of λ or λ_{sc}
μ	cos θ of scattering angle θ
$\bar{\mu}$	weighted mean of μ with respect to scattering phase function
ρ	density [kg m^{-3}]
σ	Stefan–Boltzmann constant, 5.669×10^{-8} [W m^{-2} K^{-4}]
θ	scattering angle
$\tau, \tau_\Lambda, \tau^*_\Lambda$	optical thickness at wavelength Λ of a layer (a star denotes the integration variable)
τ_0	total optical thickness, ED_0
Ω, Ω_Λ	albedo of single scattering, at wavelength Λ
$d\omega$	solid angle element.

THIS QUESTION was the subject of an invited statement prepared for a workshop on the effect of radiation on thermal transport properties held at the 9th European Thermophysics Conference, Manchester, U.K., September 1984. Following the intentions of the workshop, this problem will be discussed primarily from an experimental point of view. Two introductory remarks on a very elementary level will be made first: total thermal conductivity, λ, is a *calorimetric*, i.e. an *integral* quantity, which can be defined, e.g. by Fourier's equation:

$$\rho \cdot c_P \cdot \frac{\partial T}{\partial t} = \nabla \cdot \lambda \nabla T. \quad (1)$$

It is obvious that equation (1) specifies λ everywhere in a solid, liquid or gas only if a gradient ∇T of the temperature $T(x)$ exists within the whole volume of interest. Let us assume that the matter occupying this volume is in part transparent. A total thermal conductivity exists only if *all* existing heat transfer modes (radiative transport by absorption/re-emission and scattering, solid or liquid or gaseous heat transfer) can be described by a specific radiative, solid, liquid or gaseous conductivity. A radiative conductivity can be defined only if the diffusion model of radiative transfer applies, i.e. for a medium of a high optical thickness, τ_0. Thus the existence of the total λ is strictly speaking, coupled to an optically thick medium.

The extinction coefficient, E_Λ, on the other hand, is an *optical, spectral* quantity and is usually defined by Beer's law for a wavelength, Λ,

$$i'_\Lambda(\tau_\Lambda) = i'_\Lambda(0) \cdot e^{-\tau_\Lambda} = i'_\Lambda(0) \cdot e^{-E_\Lambda d} \quad (2)$$

where i' denotes the intensity in a particular direction.

If we consider an experimental device for the measurement of E as schematically described in Fig. 1, care has to be taken that no scattered or absorbed/re-emitted radiation falls on the detector. Otherwise application of equation (2) for determination of E is invalid, and the solution of the complete equation of transfer including a source function I'_Λ for

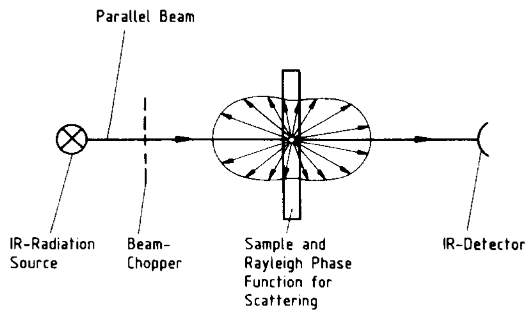


FIG. 1. Standard experimental set-up for the determination of the extinction coefficient (Rayleigh's phase function is given for illustration of an anisotropically scattering medium).

scattered and remitted intensities in the same direction

$$i'_\Lambda(\tau_\Lambda) = i'_\Lambda(0)\,e^{-\tau_\Lambda} + \int_0^{\tau_\Lambda} I'_\Lambda(\tau^*_\Lambda)\,e^{[-(\tau_\Lambda-\tau^*_\Lambda)]}\,d\tau^*_\Lambda \tag{3}$$

has to be investigated (see, e.g. [1]).

Thus, in a calorimetric measurement of λ, performed, for example, in a flat plate device schematically described in Fig. 2, two problems arise if a simultaneous determination of E is desired:

(1) If an infrared detector is tentatively integrated in the cold wall at temperature T_2 and if the sample fills the space between walls 1 and 2 completely except for a narrow gap in front of the detector, a considerable amount of scattered and absorbed/re-emitted radiation will be seen by the detector. As a consequence, an apparent extinction will be measured that is smaller than the true value. Therefore, determination of E is excluded whereas measurement of λ is possible if a gradient ∇T

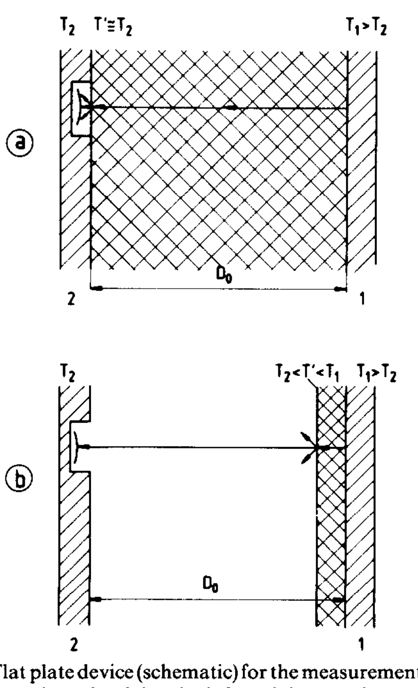


FIG. 2. Flat plate device (schematic) for the measurement of the total thermal conductivity. An infrared detector is tentatively integrated in the cold wall (2) Case ⓐ: medium fills space between walls 1 and 2 completely; case ⓑ: medium consists of a thin layer.

exists in the sample. If the sample consists of a thin layer of which its left boundary is located sufficiently far from the detector, the solid angle of the system target/detector is so small that only a negligible amount of scattered and reemitted radiation falls on the detector. Thus the detector registrates only the direct beam with intensity $i'_\Lambda(\tau_\Lambda)$ in equation (2). The measurement of λ is, however, impossible since a gradient ∇T does not exist anywhere in space between walls 1 and 2.

(2) The detector, if not equipped with spectral filters, registers an integral $\bar{E}$ value of E_Λ since walls 1 and 2 emit and reflect and the sample re-emits a distribution of radiation, which follows approximately a Planck radiation law if optical quantities like coefficients of thermal emission and refractive indices are dependent on wavelength.

In view of these difficulties, a relation between λ and E will be considered which instead of a *simultaneous* measurement of λ and E, allows an extraction of $\bar{E}$ *from* λ-measurements: following the diffusion model of radiative transfer, and assuming for the moment that the solid conduction part, λ_{sc}, of the total λ and E are independent of temperature and that the scattering phase function is isotropic, the total heat flow, $\dot{q}$, is given by

$$\dot{q} = \dot{q}_{sc} + \dot{q}_{rad}$$
$$= -\left[\lambda_{sc}(x) + \frac{16\sigma n^2}{3E(x)}\,T^3(x)\right]\frac{dT}{dx}(x). \tag{4}$$

Here σ is the Stefan–Boltzmann constant and n denotes the effective value of the refractive index (real part) of the porous medium. If equation (4) is integrated, λ can be extracted as an average value over the whole thickness D_0 of the sample as

$$\bar{\lambda} = \frac{\dot{q}D_0}{T_1 - T_2} = \overline{\lambda_{sc}} + \frac{4\sigma n^2}{3\bar{E}}\,\frac{T_1^4 - T_2^4}{T_1 - T_2}$$
$$= \overline{\lambda_{sc}} + \frac{4\sigma n^2}{3\bar{E}}\,T^{*3}. \tag{5}$$

If $\bar{\lambda}$ is plotted vs the radiation temperature, T^{*3}, $\overline{\lambda_{sc}}$ can be extracted from the intercept and $\bar{E}$ from the slope of the resulting linear relation $(\bar{\lambda}, T^{*3})$. This procedure has been successfully applied in cryophysics as well as in high temperature measurements of $\bar{\lambda}$ (see e.g. [3]) for a determination of solid conduction and radiative contribution to the total heat flow.

However, only in few exceptional cases is scattering completely isotropic. It is also to be expected that λ_{sc} and E are in reality not constant but depend on temperature.

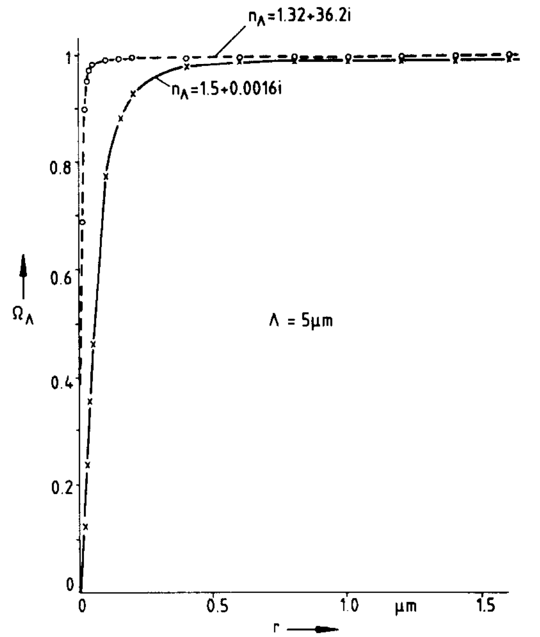
It would be desirable if the simple diffusion model could be applied also in the case of strongly anisotropically scattering media. For example, Fig. 3 shows the albedo

$$\Omega_\Lambda = \frac{S_\Lambda}{S_\Lambda + A_\Lambda} = \frac{S_\Lambda}{E_\Lambda} \tag{6}$$

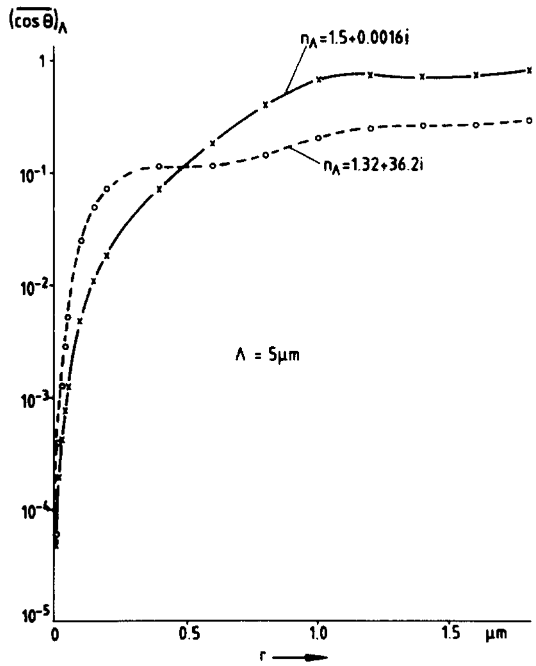
of single scattering, Ω, for glass fibers and metal fibers (silver) in the case of perpendicular incidence of unpolarized radiation at the wavelength $\Lambda = 5\ \mu m$ (S denotes the scattering and A the absorption coefficient). This calculation was performed using a computer program for evaluation of relative scattering and absorption cross sections from Mie theory following the recommendations made in [4]. Only for very small values of the fiber radius, r, is Ω small, that is, absorption dominates. We can calculate an average value of the cosine of the scattering angle, $\overline{\cos\theta}$, by its weighted mean with respect to the scattering phase function, $p(\theta)$,

$$\overline{\cos\theta} = \frac{\int \cos\theta \cdot p(\theta)\,d\omega}{\int p(\theta)\,d\omega} = \bar{\mu} \tag{7}$$

(where $d\omega$ denotes the solid angle element). In the case of completely forward, isotropic or backward scattering, respectively, $\overline{\cos\theta}$ amounts to $+1$, 0, -1. Figure 4 shows $(\overline{\cos\theta})_\Lambda$ for glass and metal fibers using the same indices of



Fɪɢ. 3. Calculated spectral values of the albedo, Ω_Λ, of single scattering for perpendicular incidence of unpolarized radiation on infinite cylinders. The complex indices of refraction are for glass (crosses, solid curve) and for silver (Ag) (open circles, dashed curve) and are taken from [12] and [13], respectively. r denotes the fiber radius, Λ the wavelength.



Fɪɢ. 4. Calculated weighted mean $\overline{\cos\theta}$ of the cosine of the scattering angle θ using the phase function of scattering for perpendicular incidence of unpolarized radiation on infinite cylinders for its calculation. The calculation has been performed for glass fibers (crosses, solid curve) and for metal fibers (open circles, dashed curve) using the same indices of refraction as in Fig. 3. r denotes the fiber radius, Λ the wavelength.

refraction, wavelength etc. as in Fig. 3. Comparison of both figures shows that if scattering predominates, the angular distribution of infrared radiation scattered by fibers is obviously forward peaked. Since essentially the same observation can be made with spheres (except for very small metallic spheres where most of the radiation is backward scattered), isotropic scattering has to be considered not as the normal but as an *exceptional* case in the whole catalogue of scattering phenomena.

It is well known from reactor physics that the diffusion model is still applicable if a modified extinction coefficient, E^*, is used for anisotropically scattered neutrons: $E^* = E(1-\bar{\mu})$ [5]. The same relation can be found for scattered photons if the phase function is expanded into a series using Legendre polynomials and if, for example, a relation

$$\frac{\mathrm{d}K}{\mathrm{d}\tau} = \frac{1}{4\pi}\,[\dot{q}_{\mathrm{rad}}(1-\bar{\mu})] \qquad (8a)$$

between the K-integral [6]

$$K = \frac{1}{2}\int_{-1}^{1} i'(\tau,\mu)\mu^2\,\mathrm{d}\mu \qquad (8b)$$

and $\dot{q}_{\mathrm{rad}}$ in the case of pure scattering is exploited for calculation of $\dot{q}_{\mathrm{rad}}$ in terms of $\pi i'$ [7]. For the total integrated heat flow, we then have in strict accordance with equation (4):

$$\dot{q} = -\left[\lambda_{\mathrm{sc}}(x) + \frac{16\sigma n^2}{3E^x(x)}\,T^3(x)\right]\frac{\mathrm{d}T}{\mathrm{d}x}(x). \qquad (9)$$

From a plot $(\bar{\lambda}, T^{*3})$, $\bar{E}^*$ can thus be extracted in the same way as described above. For a determination of $\bar{E}$ from $\bar{E}^*$ and thus from λ, it is obvious that a knowledge of the scattering phase function $p(\theta)$ is indispensable.

To the authors' knowledge, this seems to be the only reliable way for a determination of E from steady-state calorimetric measurements if the medium is weakly or even strongly scattering.

If λ_{sc} and E are in reality not independent of temperature, analysis of local, i.e. temperature-resolved valves of λ, can be performed [8].

Comparison of the extracted $\bar{E}$-values with those following (a) from measured spectral values E_Λ which are temperature averaged using the Rosseland weight function or (b) from calculated extinction cross sections using Mie theory and the Rossland weight function shows very good agreement [8, 9].

In transient measurements of λ, the temperature distribution within the medium differs the more strongly from steady-state profiles the more the medium scatters radiation [10, 11]. For a strongly backward scattering porous medium, a heat pulse applied to wall 1 leads to a sudden increase of the medium temperature near the wall because heat flow by conduction or by stepwise absorption/re-emission is a 'slow' transport mechanism. Since a temperature drop $T_1 - T_2$ occurs within a very small section of the whole sample thickness, a determination of λ and thus of E^* and E can be largely in error. Strong forward scattering transfers a heat pulse directly and very quickly to the cold wall. Although scattered radiation travels in a diffusive manner, the usual radiative diffusion equation fails with regard to this part of the radiation field [10], and application of equation (5) (or its equivalent containing $\bar{E}^*$) for the simple extraction of $\bar{E}^*$ or $\bar{E}$ from measured λ as described above is not allowed.

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