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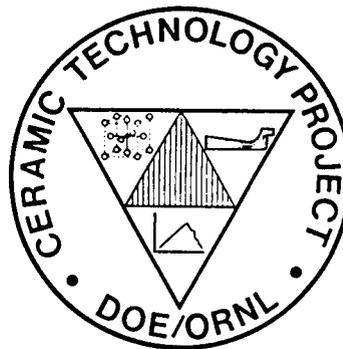
**MARTIN MARIETTA**

## **Effect of Translucence of Engineering Ceramics on Heat Transfer in Diesel Engines**

**Final Report**

**S. Wahiduzzaman  
T. Morel**

**CERAMIC TECHNOLOGY FOR  
ADVANCED HEAT ENGINES**



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MARTIN MARIETTA ENERGY SYSTEMS, INC.  
FOR THE UNITED STATES  
DEPARTMENT OF ENERGY**

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**EFFECT OF TRANSLUCENCE OF ENGINEERING CERAMICS  
ON HEAT TRANSFER IN DIESEL ENGINES**

**S. Wahiduzzaman and Thomas Morel**

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**FINAL REPORT**

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## FINAL REPORT

### EFFECT OF TRANSLUCENCE OF ENGINEERING CERAMICS ON HEAT TRANSFER IN DIESEL ENGINES

#### PART II

#### EXPERIMENTS ON RADIATIVE PROPERTIES OF CERAMICS

S. Wahiduzzaman and T. Morel

#### ABSTRACT

This report describes the experimental portion of a broader study undertaken to assess the effects of translucence of ceramic materials used as thermal barrier coatings in diesel engines. In an earlier analytical work a parametric study was performed, varying several radiative properties over ranges typical of engineering ceramics, thereby identifying the most important radiative properties and their impact on in-cylinder heat transfer. In the current study these properties were experimentally determined for several specific zirconia coatings considered for thermal barrier applications in diesel engines.

The methodology of this study involved formulation of a model capable of describing radiative transfer through a semitransparent medium as a function of three independent model parameters, ie, absorption coefficient, scattering coefficient and refractive index. A set of radiative measurements was then fitted to this model by optimization of the above model parameters. The radiative measurements were comprised of hemispherical reflectance and transmittance spectra from samples of several different thicknesses of each ceramic under collimated (unidirectional) illumination. A four-flux model was required to extract accurate radiative properties from the measurements.

For the zirconia-based ceramics investigated in this study, it was concluded that for usual coating thicknesses (1.5-2.5 mm) these ceramics are optically thick and hence, are effective as radiative heat transfer barriers. These ceramics possess high scattering coefficients and low absorption coefficients causing them to be highly reflective (60-80%) in the spectral region where thermal radiation is important. As a consequence, only a small portion of the incident energy is captured by the coating system and transmitted to the metal substrate via conduction.

The convective heat flux, which is driven by the temperature differential between the gas and the ceramic-gas interface, was found to be highly dependent on the location where radiative energy is absorbed. If a significant amount of energy is absorbed near the surface, as is the case for ceramics investigated, surface temperature is raised causing convective heat flux to be decreased. On the other hand, for ceramics that are transparent the radiative energy is absorbed at the ceramic/metal interface

and consequently, the gas/ceramic interface remains relatively cool promoting convective heat transfer.

The performance of the investigated ceramics and the mechanism of heat transfer were found to depend on surface condition, specifically on soot deposition. If the surface fouling is small, the ceramics behaved as described above, i.e., as optically thick materials characterized by high scattering coefficient and low absorption coefficient. In this case, the energy transport is primarily due to convective heat transfer since only a small fraction of the incident radiant energy can be trapped by the coating. On the other hand, if the coating surface is totally covered by soot, its performance will be similar to that of a clean optically thick material characterized by small scattering coefficient and high absorption coefficient. In this case, there will be a significant increase in radiation heat transfer absorbed at the surface with an associated decrease in convective heat transfer.

Although the coatings investigated in this study were found to be optically thick at or near nominal thicknesses (2.5 mm), this is by no means representative of all engineering ceramics. For example, based on available literature data, monolithic ceramics have been found to be more transparent than the ceramics investigated in this study and, consequently, are less effective as radiation heat transfer barriers.

Based on the results, it is concluded that ceramics with high scattering and low absorption coefficients are the most suitable for engine applications where in-cylinder surfaces can be expected to remain clean. If surfaces are expected to be sooted an opaque ceramic characterized by either high scattering coefficient and/or high absorption coefficient should be chosen in order to bring about a substantial decrease in convective heat flux. Thus, to insure the optimum thermal barrier operation for either clean or heavily sooted surfaces, a ceramic material with high scattering coefficient provides the best choice.

#### INTRODUCTION

Ceramic materials are being developed for application as heat barrier materials in insulated diesel engines. Their purpose is to reduce the heat transfer rate from the gases to the walls, with the twin benefits of increased engine efficiency and of lower heat rejection to the coolant.

The idea of a heat barrier layer is to create a path of high resistance to heat transfer, and this is accomplished by using high temperature materials which have low thermal conductivity. Certain ceramics, e.g. zirconia, combine these properties with the potential for low material cost, which accounts for the interest in their development for this application. Computer simulations of in-cylinder heat transfer processes and of the heat conduction through the structural components of the engine indicate that ceramics can indeed provide very substantial reductions in engine heat transfer, translating into important thermal efficiency gains, reduced heat rejection to coolant and increased exhaust energy availability.<sup>1</sup>

However, it is increasingly being realized that some of the ceramics proposed for diesel engine thermal barriers are partially transparent in the spectral region where most of the thermal energy associated with the combustion process is concentrated (Liebert<sup>2,3</sup> and Makino et al<sup>4</sup>). This causes concern about the effectiveness of such ceramic layers in reducing heat transfer in insulated diesel engines, where it is known that radiation can account for a significant portion of the total heat transfer.<sup>6</sup>

The gas-to-wall heat transfer in diesel engines is produced by convection from cylinder gases and by radiation from the soot-laden burning zone. In the high output turbocharged engines typically used in highway truck applications, the heat radiation accounts for some 20 percent of the total heat transfer from gases to walls at the rated engine conditions. When the combustion chamber is insulated, the convective component of the heat transfer is selectively reduced. The proportion of heat transfer that is due to radiation then rises to over 50 percent, thereby becoming a much more significant fraction of<sup>5</sup> the overall heat rejection and a more important design consideration.

In all simulations of diesel engine heat transfer processes reported to date, the assumption has been made that the absorption coefficient of the ceramic is very large so that all of the incident radiation is deposited at the gas/ceramic interface -- i.e., the radiation has not been allowed to penetrate into the interior. This is a carryover from studies of metallic engines where this treatment provides a fairly accurate description of the actual process. However, since some ceramics of engineering interest are known to be translucent, especially at infrared radiation wavelengths, this approximation may be quite incorrect.

The work carried out on this contract is divided into two parts. A previous report<sup>7</sup> described the results of the first part, which concerned an analytical study of the radiation process within engineering ceramics, and showed how heat transfer is affected by the radiation properties of ceramics (absorption coefficient, scattering coefficient, index of refraction and surface reflectivities). This report presents the results of the second part of the work concentrating on an experimental determination of the radiative properties of some specific ceramics, and then predicting their effectiveness as heat barrier materials when used in a diesel engine environment.

There were three principal aspects to work described in this report. The first was the selection and implementation of an appropriate methodology to determine the radiative properties of the ceramic materials. The second was the use of the method to determine the radiative properties of several existing coatings that have been developed for diesel engine application. The last was an analysis of the heat transfer through a ceramic-coated wall of a diesel engine, including all three heat transfer mechanisms -- convection, radiation and conduction -- and through it an assessment of the effectiveness of these specific ceramics as heat barrier materials.

## METHODOLOGY

The principal parameters required to describe radiative transport in a semitransparent medium are the absorption coefficient ( $k$ ), scattering coefficient ( $s$ ), refractive index ( $n$ ) and scattering phase function ( $p$ ). For many ceramic materials in which scattering can be assumed to be isotropic, the first three parameters are adequate to describe radiative transport, and in this work we concentrate on these three. An additional assumption, implicit in this description, is that the interfacial reflectivities are known or can be determined using Fresnel's equation. The latter approach is chosen for this study which requires that the interfaces are optically smooth.

The approach adopted here to determine these parameters for a given material is to take radiative measurements of several samples of that material fashioned into thin coupons of different thicknesses. A model describing the radiative transfer in semi-transparent media can be then fitted to the acquired data to determine the above radiative properties. The parameter inversion process involves matching the measured reflectance and transmittance data of a set of coupons of different thicknesses with the model computed values. An objective function based on a sum of squares of normalized differences between measured and computed reflectance and transmittance values can be minimized to obtain the values of  $n$ ,  $k$  and  $s$  which give the best correspondence between measurement and model predictions.

The approach discussed above was implemented using measurements obtained at two different locations (wavelengths of 0. to 2.5  $\mu\text{m}$  at NBS\* and 2.0 to 20.0 at Willey Corporation\*\*). In both laboratories hemispherical reflectance and transmittance spectra were measured under collimated illumination and consequently include contributions of the collimated flux in addition to the diffuse flux which can be created from the collimated flux undergoing multiple scattering. This necessitates the use of a radiation model capable of describing decay of the collimated flux into a diffuse flux due to multiple scattering unless the incident radiation is diffuse to begin with. A four flux model as opposed to a two flux model is better equipped to handle the problem at hand. Initially a two flux model was used for parameter inversion and was soon discovered to be inadequate at low scattering levels and was discarded in favor of a four flux model. However, for the sake of completeness both models will be discussed below and a comparison of the model performances will be presented in a later section.

---

\*Center for Radiation Research, National Measurement Laboratory, National Bureau of Standards, Gaithersburg, Maryland 20899.

\*\*Willey Corporation, P.O. Box 670, Melbourne, Florida 32901.

#### FOUR FLUX MODEL

The model chosen in this study is the four-flux model, which predicts specular and diffuse reflectivity and transmissivity for a planar medium illuminated by diffuse and/or collimated flux. The parameters  $k$ ,  $s$ ,  $n$  and the material thickness are the only inputs to the model. The model (Ishimaru<sup>5</sup> and Makino and Kunitomo<sup>4</sup>) assumes that the radiation transport in a cold (non-emitting) medium can be described by four fluxes. Two of these are a diffuse forward flux and a diffuse backward flux, both of which remain diffuse upon interfacial reflection and scattering. The other two fluxes are a collimated forward and a collimated backward flux which undergo isotropic scattering, thereby contributing equally to the forward and backward diffuse fluxes. The collimated fluxes are assumed to remain collimated after specular (mirror-like) interfacial reflections. Under these assumptions the governing equations and associated boundary conditions are (Figure 1):

governing equations:

$$\frac{dD^+}{dx} = -(2k+s) D^+ + sD^- + \frac{s}{2} C^+ + \frac{s}{2} C^- \quad (1)$$

$$-\frac{dD^-}{dx} = -(2k+s) D^- + sD^+ + \frac{s}{2} C^- + \frac{s}{2} C^+ \quad (2)$$

$$\frac{dC^+}{dx} = -(k+s) C^+ \quad (3)$$

$$-\frac{dC^-}{dx} = -(k+s) C^- \quad (4)$$

boundary conditions:

$$D^+(0) = (1-R_{do}) F_d + R_{di} D^-(0) \quad (5)$$

$$D^-(d) = R_{di} D^+(d) \quad (6)$$

$$C^+(0) = (1-R_{co}) F_c + R_{ci} C^-(0) \quad (7)$$

$$C^-(d) = R_{ci} C^+(d) \quad (8)$$

where 0 and d denotes front and back surface, respectively.

A general solution to the above set of equations is:

$$D^+(x) = \alpha_1 A e^{-\tau} + \alpha_2 B e^{\tau} + \alpha_3 (1+\beta) e^{-2\beta\tau} + \alpha_4 (1-\beta) e^{2\beta\tau} \quad (9)$$

$$D^-(x) = \alpha_1 B e^{-\tau} + \alpha_2 A e^{\tau} + \alpha_3 (1-\beta) e^{-2\beta\tau} + \alpha_4 (1+\beta) e^{2\beta\tau} \quad (10)$$

$$C^+(x) = \alpha_1 e^{-\tau} \quad (11)$$

$$C^-(x) = \alpha_2 e^{\tau} \quad (12)$$

where

- A =  $3\omega/(8\beta^2-2)$
- B =  $A/3$
- $C^+$  = collimated forward flux
- $C^-$  = collimated backward flux
- d = depth of the layer
- $D^+$  = diffuse forward flux
- $D^-$  = diffuse backward flux
- $F_d$  = diffuse incident flux
- $F_c$  = collimated incident flux
- k = absorption coefficient
- $R_{di}$  = diffuse interfacial internal reflection
- $R_{do}$  = diffuse interfacial external reflection
- $R_{ci}$  = collimated interfacial internal reflection
- $R_{co}$  = collimated interfacial external reflection
- s = scattering coefficient
- $\beta = \sqrt{1-\omega}$
- $\lambda$  = wavelength
- $\omega = s/(s+k)$  = albedo
- E =  $s+k$  = extinction coefficient
- $\tau = (k+s)x$  = optical depth at x
- $\tau_d = (k+s)d$  = optical thickness of the translucent layer

The coefficients  $\alpha_1$ ,  $\alpha_2$ ,  $\alpha_3$  and  $\alpha_4$  are determined by solving the following simultaneous equations obtained using the boundary conditions:

$$\begin{bmatrix}
1 & -R_{ci} & 0 & 0 \\
-R_{ci}e^{-\tau d} & e^{\tau d} & 0 & 0 \\
(A-R_{di}B)e & (B-R_{di}A)e & (1+\beta) & (1-\beta) \\
(B-R_{di}A)e^{-\tau d} & (A-R_{di}B)e^{\tau d} & -(1-\beta)R_{di} & -(1+\beta)R_{di} \\
& & [(1-\beta) & [(1+\beta) \\
& & -(1+\beta)R_{di}]e^{-2\tau d} & -(1-\beta)R_{di}]e^{2\tau d}
\end{bmatrix}
\begin{bmatrix}
\alpha_1 \\
\alpha_2 \\
\alpha_3 \\
\alpha_4
\end{bmatrix}
-
\begin{bmatrix}
(1-R_{co})F_c \\
0 \\
(1-R_{do})F_d \\
0
\end{bmatrix}
\quad (13)$$

Then collimated and diffused reflected fluxes at the inside front surface can be calculated after substitution of Eqs. (10) and (12) in the following equations:

$$S_c = R_{co} F_c + (1-R_{ci})C^-(0) \quad (14)$$

$$S_d = R_{do} F_d + (1-R_{di})D^-(0) \quad (15)$$

respectively.

Similarly, the collimated and diffused transmitted fluxes at the back surface of the ceramic are obtained after substituting Eqs. (9) and (11) in the following equations:

$$H_c = (1-R_{ci}) C^+(d) \quad (16)$$

$$H_d = (1-R_{di}) D^+(d) \quad (17)$$

From these fluxes reflectance and transmittance can be appropriately defined as follows:

$$R_h = (S_c + H_c)/(F_d + F_c) \quad (18)$$

$$T_h = (S_d + H_d)/(F_d + F_c) \quad (19)$$

## TWO FLUX MODEL

Under purely diffuse illumination, only two diffuse fluxes are necessary to describe radiative transport in a planar medium provided assumptions previously made are still valid. It was assumed that a collimated flux transforms into diffuse forward and backward fluxes by undergoing isotropic scattering. If the scattering level is high this conversion will happen early and a simpler two (diffuse) flux model can then be used as a further approximation of the radiative transport in such media. The flux equations and the boundary conditions for the model can be written as:

$$\frac{dD^+}{dx} = - (2k+s)D^+ + sD^- \quad (20)$$

$$- \frac{dD^-}{dx} = - (2k+s)D^- + sD^+ \quad (21)$$

The accompanying boundary conditions are:

$$D^+(0) = (1-R_{do})F_d + R_{di}D^-(0) \quad (22)$$

$$D^-(d) = R_{di}D^+(d) \quad (23)$$

It should be noted here that the above equations are identical to those obtained from the four flux model under purely diffuse illumination.

The general solution to the above equations is:

$$D^+ = \alpha_3(1+\beta)e^{-2\beta r} + \alpha_4(1-\beta)e^{2\beta r}$$

$$D^- = \alpha_3(1-\beta)e^{-2\beta r} + \alpha_4(1+\beta)e^{2\beta r}$$

The coefficients  $\alpha_3$  and  $\alpha_4$  are then determined from the boundary conditions as follows:

$$\begin{bmatrix} (1+\beta) - (1-\beta)R_{di} & (1-\beta) - (1+\beta)R_{di} \\ [(1-\beta) - (1+\beta)R_{di}]e^{-2\beta\tau_d} & [(1+\beta) - (1-\beta)R_{di}]e^{2\beta\tau_d} \end{bmatrix} \begin{bmatrix} \alpha_3 \\ \alpha_4 \end{bmatrix} = \begin{bmatrix} (1-R_{do})F_d \\ 0 \end{bmatrix} \quad (24)$$

After solution one obtains

$$\alpha_3 = \frac{F_d(1-R_{do})Ue^{2\beta\tau_d}}{U^2e^{2\beta\tau_d} - V^2e^{-2\beta\tau_d}} \quad (25)$$

$$\alpha_4 = \frac{-F_d(1-R_{do})Ve^{-2\beta\tau_d}}{U^2e^{2\beta\tau_d} - V^2e^{-2\beta\tau_d}} \quad (26)$$

where

$$U = (1+\beta) - R_{di}(1-\beta)$$

$$V = (1-\beta) - R_{di}(1+\beta)$$

The reflected and transmitted fluxes are:

$$S_d = R_{do}F_d + (1-R_{di})D^-(0) \quad (27)$$

$$H_d = (1-R_{di})D^+(d) \quad (28)$$

Then the reflectance is given by:

$$\begin{aligned} R_d &= S_d/F_d \\ &= R_{do} + (1-R_{do})(1-R_{di}) \frac{(1-\beta)Ue^{2\beta\tau_d} - (1+\beta)Ve^{-2\beta\tau_d}}{U^2e^{2\beta\tau_d} - V^2e^{-2\beta\tau_d}} \end{aligned} \quad (29)$$

and the transmittance is given by:

$$T_d = H_d/F_d$$

$$= (1-R_{do})(1-R_{di}) \frac{(1+\beta)U - (1-\beta)V}{U_e^{2-2\beta r_d} - V_e^{2-2\beta r_d}} \quad (30)$$

It should be noted here that the reflectance and transmittance in equations 18, 19, 29 and 30 are spectral quantities (i.e., have unique values for each wavelength).

The essence of the method involves minimizing the differences between the computed reflectance and transmittance spectra using these equations with the measured spectra by suitable choice of model parameter in order to obtain spectral distributions for these parameters.

## EXPERIMENTAL DATA AND THEIR ANALYSIS

### RADIATIVE MEASUREMENTS

Four different ceramics were available for radiative parameter inversion. These included plasma sprayed zirconia (PSZ) (13% Yttria stabilized) from Plasma Technics\*, PSZ and TZP (Tridiagonal Zirconia Polycrystals) from Oak Ridge National Laboratory\*\* and PSZ (24% Ceria stabilized) from Caterpillar\*\*\*. The success of the above procedure, i.e. obtaining the radiative properties of a given ceramic, depends on acquiring a set of experimental data which are independent of each other. This requires measurement of reflectances and transmittances at sufficiently distinct optical depths. Hence, it is desirable to select thicknesses of samples such that an optically thin, an optically thick and an intermediate optical thickness are included in the set of samples. However, without a priori information, which was the case in the present study, one can only guess at the best thicknesses for this purpose. Based on feasibility of fabrication of the thinnest samples, mechanical strength, and literature values of properties of similar materials, three to four thicknesses were specified and fabricated for each of the four ceramics investigated in this study. All of these samples are labeled and described in the Table I.

---

\*Plasma Technics Inc., 1143 West Newport Center Drive, Deerfield Beach, Florida 33442.

\*\*Oak Ridge National Laboratory, P. O. Box 2008, Oak Ridge, Tennessee 37831.

\*\*\*Caterpillar Inc., Technical Center, P.O. Box 1875, Peoria, Illinois 61656-1875.

The original plans were to obtain normal incidence diffuse reflectance for all thirteen samples. The transmittance measurements were planned only for nine samples, excluding the thickest sample of each set which were expected to be too thick to allow any transmission. The first set of measurements was made over a wavelength band spanning 2  $\mu\text{m}$  to 20  $\mu\text{m}$  at Willey Corporation, a private firm specializing in radiometry. In this wavelength range meaningful transmittance measurement could be obtained only for seven samples, because only the thinnest sample of the PSZ samples from Caterpillar (set D) transmitted enough energy to obtain reliable transmittance data.

The measurements at lower wavelength, extending from 0.25 to 2.5  $\mu\text{m}$ , were conducted at the National Bureau of Standards. Again for some of the samples (A2, D11, D12) poor transmittance results were obtained owing to low signal-to-noise ratio. In addition, sample A1 was found to be broken during transit and no transmittance measurement could be obtained from this sample. The matrix of radiative measurements carried out by Willey Corporation and NBS are summarized in Table II.

#### ANALYSIS OF CONSISTENCY OF THE EXPERIMENTAL DATA

In this study, the emphasis of the analysis was placed on the wavelength range from the visible (0.4  $\mu\text{m}$ ) to 5  $\mu\text{m}$  where thermal radiative energy is important in diesel engine applications. (Note that the peak radiant intensity of a blackbody occurs at 2.9  $\mu\text{m}$  at 1000K and at 1.45  $\mu\text{m}$  at 2000K.) This wavelength band contains the overlap region (2.0  $\mu\text{m}$  to 2.5  $\mu\text{m}$ ) where data was obtained at both laboratories. The agreement between the data in the overlap range was examined from the two sources and it was found satisfactory. (It should be noted that the data from Willey Corporation exhibited noise inherent to the measuring instrument<sup>8</sup> and had to be smoothed.) The composite plots of data belonging to both sources are shown in Figures 2 to 5. (The unsmoothed data from Willey Corporation is presented in the Appendix.)

The available data were carefully scrutinized to determine their suitability for radiative parameter inversion. The following observations of unusual trends were made:

- (1) Sample sets B and C from Oak Ridge exhibited very similar reflectance and transmittance spectra despite different composition (Figures 3 and 4). This similarity was evident in the data obtained at both Willey and NBS. It was concluded that the two sets are radiatively identical.
- (2) The qualitative trend of the reflectance data of sample C9 was markedly different than those of thinner samples of the same material (C7 and C8). The differences include a) lower reflectance values below 0.5  $\mu\text{m}$ , b) relatively constant reflectance values between 0.5  $\mu\text{m}$  and 2.5  $\mu\text{m}$  and c) most importantly, a dip in the reflectance values around 2.75  $\mu\text{m}$  (Figure 4b). These characteristic differences suggest that the

sample C9 may have been contaminated during its fabrication. Hence it was necessary to exclude the data from this sample from parameter inversion analysis for set C. A similar conclusion can be made for sample B4 as evidenced in Figure 3a.

- (3) Reflectance spectra for samples belonging to set D exhibited an inconsistent trend with thickness. The spectra of samples D10 and D13 possessed characteristics which are absent in the spectra for samples D11 and D12 (Figure 5b). These characteristics include a dip at  $2 \mu\text{m}$  and high reflectance values beyond  $3 \mu\text{m}$ . Since transmittance data were available for sample D10, it was decided that only data from samples D10 and D13 would be used for parameter inversion for set D. The choice D11 and D13 would only provide two sets of measurements as opposed to three sets of measurements for the choice made.

After careful consideration of the above factors, consistent sets of data from the two laboratories were merged. Since data obtained at Willey Corporation are less reliable in the overlap region ( $2 \mu\text{m}$  to  $2.5 \mu\text{m}$ ), the data from NBS were given higher weight in this band during merger. Figures 6 to 8 show merged data for sample sets B, C and D. It can again be noted here, allowing for the measurement uncertainties, sets B and C possess identical spectra.

Unfortunately, the data for sample A is too sparse for parameter inversion. It can be seen in Figure 2(b) that reflectance is nearly identical for all three samples (A1, A2, A3). (The spectrum A1 presented here was measured from a damaged sample and as such NBS has not certified this data.) This signifies that even at the smallest thickness ( $0.51 \text{ mm}$ ) the ceramic is optically thick and therefore the samples do not produce sufficiently independent information required for parameter inversion. It should be noted that transmittance spectrum of A1 is unreliable since it was measured from a damaged sample. Despite the deficiencies in the measurements, some conclusions can still be drawn for this material and will be discussed in a later section.

#### PARAMETER INVERSION

As discussed in the previous section, for sets B and/or C we have two reflectance and two transmittance spectra that are consistent within each set. For the set D, there are only two reflectance and one transmittance spectra that have consistency. Thus, for both sets, the number of spectra available for parameter inversion is rather small. It was found that it is a difficult, if not impossible, task to extract three independent radiative parameters from such a sparse input data set. Initial analysis showed that of the three chosen parameters ( $n$ ,  $k$ ,  $s$ ), the refractive index  $n$  is the least sensitive. Upon analysis, one observes that the refractive index affects radiative transport in two ways; through interfacial reflectivities (Fresnel's reflectivity) and through the self-emission term. Since the ceramics studied here possess high scattering coefficients, interfacial reflectivities have little contribution to layer reflectances. In addition, reflectance and transmittance of the samples were measured at

room temperature, consequently, the self-emission term is negligible in comparison to other fluxes and may be dropped from the transport equation. Under these circumstances, it was decided to keep the refractive index fixed at 1.58 and to determine only the remaining two parameters (s and k) through the parameter inversion procedure. This value of refractive index is based on previous observations and trends observed during current data analysis. It was reported in previous investigations (e.g. Makino and Kunitomo<sup>4</sup>), that refractive index remains nearly constant from 0.5  $\mu\text{m}$  to 5  $\mu\text{m}$ . Its value is expected to be between 1.2 to 2.5. In yet another study (Matthews<sup>9</sup>) involving a similar nonlinear parameter inversion technique, the refractive index of zirconia was estimated to be 1.6. Thus the value of 1.58 chosen for this study is expected to be close to the actual value of the parameter. The total uncertainty on the remaining parameters introduced by this range in refractive index is of the order of 25% and 35% for scattering and absorption coefficient, respectively.

With the above restrictions, the parameter inversion model was applied to data obtained for sample sets B and C. The results are shown in Figures 9 and 10. As expected, two samples exhibited nearly identical radiative properties. The scattering coefficient is found to be high in the visible band, and it steadily decreases as wavelength is increased. On the other hand, absorption coefficient is relatively small and remains constant except in the visible band (0.4-0.7  $\mu\text{m}$ ), and beyond 4  $\mu\text{m}$  where it starts to increase very sharply. Figures 11a and 11b show model-predicted reflectance and transmittance for sample set C using the k and s values deduced from the data. It can be observed that the predicted reflectances and transmittance are in good agreement with the experiment except for the 10 mm sample (Figure 4b). It may be recalled that the measured reflectance spectrum for this sample was excluded from the data analysis owing to its inconsistent characteristics.

Analysis was also performed on the data belonging to set D. The inverted parameters are shown in Figures 12a and 12b. The predicted reflectance and transmittance are shown in Figure 13a. There is good agreement between measured (Figure 13b) and predicted (Figure 13a) transmittance values, however, the predicted reflectances for the two different thicknesses are nearly identical to the measured reflectance of the thinnest sample (D10). If an absorptance spectrum is computed for this sample from the measured reflectance and transmittance (Absorptance = 1 - Transmittance - Reflectance) spectra, it can be readily observed that for much of the spectrum the optical depth of the sample is high. Hence, there is no reason to believe that for higher thickness reflectance will be markedly different, contrary to the trends suggested by the data. Rather, it is likely that the apparent differences in the measured reflectance spectra are due to variations in surface preparations.

The scattering coefficients for set D possess the same qualitative trend as those of set B and C except their values are about twice as high. On the other hand, the absorption spectrum shows a distinctive peak at 3  $\mu\text{m}$  corresponding to the valleys in the reflectance and transmittance spectra. In general, set D is more opaque than B and C due to its higher scattering and absorption coefficients.

It is worth noting here that the radiative parameters obtained in this study are derived from room temperature measurements. However, in a previous study Makino and Kunitomo<sup>4</sup> did not observe any major dependence of radiative properties on temperature for zirconia and alumina. Hence, it is believed that the parameter values reported here will be valid for diesel engine applications where temperatures (1000 K) are not too dissimilar to the range investigated in the referenced study (~300 K to 700 K).

#### FOUR FLUX MODEL VS. TWO FLUX MODEL

The parameter inversion procedure discussed in the previous section utilizes a four flux radiation model. However, initially a two flux radiation model was used before it was discarded in favor of the four flux model. The experimental measurements described earlier were made under collimated (near normal beam) illumination. The two flux model is inadequate for describing propagation of collimated flux through a semi-transparent medium. However, if the fraction of incident energy that penetrated the medium is properly calculated (using directional reflectivities), the two flux model may still be valid for a medium where collimated flux undergoes quick transformation into diffuse flux due to high scattering level. On the other hand, for samples having small optical depths where the extinction coefficient is of the order of unity the contribution of the internal collimated flux on total transmittance and reflectance will be comparable to the contribution from the internal diffuse flux. Hence, in this case it is necessary to solve for both collimated and diffuse fluxes simultaneously, which the two flux model cannot accomplish.

In order to remove this weakness in the originally planned radiative parameter inversion model, replacement of the the two flux model by a four flux model was essential as it was confirmed by comparisons of parameters evaluated by both models. (Both of these models have been described in previous sections.) Initial results obtained using the original two-flux inversion model revealed that for ceramics under study the scattering level tapers off at higher wavelength. It seemed likely that the validity of two-flux model will break down at these high wavelengths. The revised model utilizing a four-flux model was applied to the available experimental data. The radiative parameters thus deduced were compared against the parameters from the two-flux model. Figures 14a and 14b show a comparison of radiative parameters obtained using both models for ceramic D. It was observed that the two-flux model underpredicted the scattering coefficient by as much as 50% at higher wavelengths, whereas the absorption coefficient remained almost unchanged.

#### RECOMMENDATION FOR FUTURE MEASUREMENTS

It is evident that the proper choice of the sample thicknesses is an essential prerequisite for successful parameter inversion. However, prior knowledge regarding optical thickness is required to make such choices. Hence the recommended procedure would be to make preliminary transmittance

measurements to obtain some knowledge about the opacity of the sample, or making a larger number of samples (at least 5) of various thicknesses to increase the likelihood of capturing the right sample thickness range. The recommended set of optical thicknesses is  $kL = 0.5, 1.0, 1.5, 2.0$  and  $2.5$  which assures that meaningful values of transmittance and reflectance can be attained. For example if a material has an extinction coefficient of  $10,000 \text{ m}^{-1}$  in the wavelength region of interest, the required samples will have thicknesses of  $0.05 \text{ mm}$  to  $0.25 \text{ mm}$ . If samples that thin cannot be fabricated, a thicker set of samples may be substituted, but a lower accuracy will result.

As discussed earlier, the refractive index is the least sensitive of the chosen parameters. In order to extract this parameter, one needs measurement techniques which possess stronger dependence. One approach could be a separate measurement of the diffuse and directional reflectance and transmittance (in the present case the sum of the diffuse and directional values was measured). However, this would require greater attention to surface preparation; the sample surfaces should be smooth, parallel and consistent from sample to sample. Another approach would measure polarized components of this reflectance.

Further, the number of samples should be as large as possible. A large number of samples is often necessary (eight for Makino<sup>10</sup>) to obtain reliably at least three linearly independent measurements. As for the data reduction method, it is recommended that it be based on the four-flux radiation model (or a higher order model) which has been shown here to be better capable of dealing with radiative properties of typical engineering ceramics than the two-flux model.

## ANALYSIS OF TRANSLUCENCE EFFECTS ON HEAT TRANSFER

### APPROACH

The general scheme of evaluating the heat transfer in diesel engines involves the solution of the energy equation (heat transfer) within the wall under appropriate gas side and coolant side boundary conditions. In the present study the cylinder walls were assumed to be composed of two layers: an insulating ceramic wall facing the combustion gas and a metallic substrate. The energy equation is a one-dimensional conduction equation with an additional source term representing radiation energy absorbed/emitted within the medium. This energy equation was solved for two distinct regions: a semitransparent region within the ceramic, and a fully opaque region within the substrate. A two-flux radiative transport equation was solved to compute the radiative heat flux vector. Since the radiation process in a diesel engine has no collimated component, both the two-flux model and four-flux yield the same result. However, equations 1, 2, 20 and 21 do not contain a self emission term. This term was included since the wall temperature of ceramics in diesel engine applications is expected to be in a region where self emission begins to be important. The resulting equation is given in a previous report (Wahiduzzaman and Morel<sup>7</sup>).

The gas side boundary conditions were established by computer simulation of in-cylinder processes taking place in a heavy duty truck diesel engine operating at rated conditions using IRIS engine design analysis code<sup>11</sup>. This provides cyclic histories of radiation/convection temperatures, heat transfer coefficients and the products of soot volume fractions and radiation path length. These histories are shown in Figures 15a, 15b and 15c. The coolant side boundary condition consisted of a fixed coolant temperature (640K) and a very high heat transfer coefficient to impose a temperature of about 650K at the ceramic/metal interface. Details of the implementation of the energy equation and the boundary conditions were given in an earlier report describing the analytical phase of the study (Wahiduzzaman and Morel<sup>7</sup>). It is worth noting here that during the part of the cycle when no radiating combustion gas is present, it was assumed that the volumetric and surface emission outward from the ceramic is exactly balanced by the energy received from the other walls. This is equivalent to the assumption of equal radiosity for all walls.

For typical radiation temperatures encountered during combustion ( $T_{\text{gas}} = 1800\text{-}2500\text{K}$ ), the fraction of total radiation energy existing beyond  $5 \mu\text{m}^{\text{gas}}$  is small (5-11%). At the same time, it was also evident from the experimental results that the ceramics under study become highly opaque beyond this wavelength. Hence, the ceramic layer can be assumed to have an opaque band from  $5 \mu\text{m}$  to  $\infty$ , where the absorption and emission of radiation energy beyond  $5 \mu\text{m}$  is accounted for as a surface phenomenon. For the rest of the wavelength domain, the absorption and scattering coefficient spectra were discretized into a number of bands having constant radiative properties within each band. The number of bands was carefully chosen so as to preserve a satisfactory match with the original spectra, without imposing an excessive computational burden in the subsequent analysis. The band

model representation of absorption and scattering coefficient spectra is summarized in the following tables.

#### Ceramic B and C

$\lambda$ ( $\mu\text{m}$ )	0.0-0.5	0.5-1.0	1.0-1.5	1.5-2.5	2.5-4.0	4.0-5.0
k (1/m)	150.0	35.0	20.0	20.0	20.0	112.0
s (1/m)	35000.	27400.	10700.	7200.	6200.	4600.

#### Ceramic D

$\lambda$ ( $\mu\text{m}$ )	0-1.5	1.0-1.5	1.5-2.75	2.75-3.5	3.5-5.0
k (1/m)	710.0	300.0	10.0	1570.0	14.0
s (1/m)	45200.	45200.	30600.	14300.	20400.

The thermophysical properties (conductivity and heat capacity) of the ceramic samples were not available and their determination was beyond the scope of the present study. It was shown earlier (Wahiduzzaman and Morel<sup>6</sup>) that thermal conductivity is an important parameter influencing the relative sensitivity to translucence. Presently, in the absence of such data, nominal values of 0.8 W/mK and  $2.91 \times 10^6$  J/Km<sup>3</sup>, pertaining to Plasma Technics plasma sprayed PSZ, were used for thermal conductivity and volumetric heat capacity, respectively, of all the materials considered in this study.

Heat transfer analysis was carried out for several thicknesses of ceramic coatings ranging from 0.04 mm to 5.0 mm, where 2.5 mm (~0.100 inches) is chosen as the nominal thickness for most of the calculation to be discussed later on. The grid points in the ceramic and the substrate were generated in such a manner that the grid spacing was compact near the gas-ceramic and ceramic-substrate interfaces. The smallest elements were located at either side of the ceramic-substrate interface and at the gas-ceramic interface, and they were 0.015 mm (0.0006 inch) thick. There were twenty-one grid points in the insulating ceramic layer.

#### EFFECTIVENESS OF INVESTIGATED CERAMICS AS HEAT BARRIER MATERIALS

The effectiveness of the ceramics under study to act as a barrier to thermal radiation was investigated by subjecting these and three idealized (limiting-case) ceramics to identical boundary conditions. The results of these simulations are summarized in Table III. The idealized ceramics were either completely transparent or completely opaque. The opaque limiting cases were of two types, ie, ceramics characterized either by high scattering coefficient ( $>10^7$  m<sup>-1</sup>) and negligible absorption coefficient (denoted as opaque (s) in Table III) or by high absorption coefficient ( $>10^7$  m<sup>-1</sup>) and negligible scattering coefficient (denoted as opaque (a) in

Table III). All five ceramics had a thickness of 2.5 mm, and the nominal thermophysical properties discussed above. The resulting temporal history of various components of the total heat flux for ceramics C and D is shown in Figures 16a and 16b. The solid curve represents the net radiation flux just inside the ceramic at node #1. The dashed line represents the amount of this flux transmitted to the ceramic/metal interface. The dotted line represents the radiation flux comprising the reflected and emitted radiation from the ceramic coating to the gas. The remaining line represents the convective heat flux to the ceramic coating.

It may be noted that the total flux for the clean C case is only one quarter of the metal case. This is due to the much higher surface temperature which reduces the convective heat transfer. On the other hand the swing is about three times as high, and that is due to the lower value of the product  $\rho ck$ .

It can be seen from Table III that the effectiveness of the two actual ceramic coatings (each 2.5 mm thick) as a barrier to total heat transfer is about the same. However, the ratio of the radiative heat flux to convective heat flux is different. For the more opaque ceramic (D), much of the radiation is absorbed near the surface, raising the mean surface temperature. This lowers the effective temperature difference between the combustion gas and the surface, and the convective heat flux is reduced. It is interesting to note that the radiative heat flux through ceramic C is less than that with ceramic D, despite the fact that C is more transparent than D. This is a consequence of differences in reflectance between the two materials. Since both materials have a high scattering coefficient, the one which has a lower absorption coefficient scatters more energy out of the coating before it can be absorbed. Hence ceramic C has a higher total reflectance than ceramic D. Consequently, less net radiation energy is absorbed in ceramic C than D.

If the thicknesses of the samples are sufficiently reduced this trend can be reversed, since for the ceramic C more of the radiation energy will reach the ceramic-substrate interface and be absorbed there before being scattered back out. Figures 17a and 17b show distributions of radiative and total heat fluxes as a function of layer thickness, and show that this reversal happens at around a thickness of 1.25 mm. In either case, however, the heat flux decreases with increasing layer thickness. It may be noted that the total heat fluxes for the two ceramics appear to not converge as the coating thickness is reduced towards zero. That is not really so, because in the limit the two curves must coincide. However, at even very small thicknesses, e.g., 0.05 mm for these specific ceramics, the product of the scattering coefficient and the layer thickness is large enough to produce significant scattering which augments the reflected component of radiation heat flux. Since the scattering coefficient of ceramic D is greater than that of ceramic C its net radiative flux is smaller and this persists to even quite small thicknesses.

In terms of the total heat flux, it appears from Table III that the ceramics under study are much closer to both opaque limiting cases than to the transparent limiting case. However, the mechanism of the heat transfer

process is closer to the opaque(s) limit; in this opaque limit most of the heat transfer is due to the convective flux, since very little radiative flux can penetrate the coating due to high reflectance caused by its high scattering and low absorption coefficients. In ceramics C and D a large fraction of incident radiant energy will be reflected away for the same reason. Consequently, a smaller fraction of the radiation energy can be trapped by the coating system and the surface temperature only increases moderately due to absorption of this radiation energy. This in turn will cause a small reduction of the convective heat flux from the maximum value obtainable in the fully transparent case. For the opaque (a) limiting case much of the incident radiation will be absorbed at the surface raising its temperature which will cause a larger reduction in the convective heat flux at the cost of an increased radiative heat transfer.

In the case of the ideal transparent case, both radiative and convective heat fluxes are high. Since there is no scattering, a very small portion of the incoming radiative energy is reflected (due to interfacial Fresnel's reflectivity) and most of the energy is absorbed at the ceramic-substrate interface. This leaves the gas-ceramic interface at a relatively lower temperature, which increases the convective heat transfer.

#### EFFECT OF SURFACE CONDITIONS

In the preceding analysis it is implicitly assumed that the surface of the ceramic thermal barrier is the same as that of the samples obtained for our analysis. Further it is assumed that it remains free of soot and other deposits and that its surface layer does not become impregnated with foreign materials and/or discolored.

These are very significant assumptions and one needs to understand how do the results obtained here translate to situations where the surface condition is different from that found on the supplied test samples. In an operating diesel engine it is expected that surface fouling will occur and degree of which will depend on the particular engine and the operating condition. This surface fouling could be due to absorption of moisture/fuel by the coating and/or deposition of solid particulates in the ceramic surface. The absorption of solid and liquid particulates can be expected to affect the thermophysical and radiative properties of the coating. The effect of this change is difficult to anticipate or model. In terms of radiative properties, the absorption of combustion products by the coating will tend to make the coating more radiatively opaque and the opaque (a) limit of the coating will provide an indication of its effect on heat transfer.

If the ceramic surface departs from being optically smooth ( $l/\lambda < 1$ , where  $l$  is rms value of surface roughness) more and more incident radiation will be trapped thereby reducing effective reflectance and increasing hemispherical emissivity. In general, this will tend to promote radiation heat transfer. It is difficult, if not impossible, to predict the effect of impregnated surface impurities. It will, most likely, be dependent on the nature of impurities. It can however be speculated that impurities will also alter the surface finish and make it optically rough and thereby will have a

similar impact on radiant heat transfer as surface roughness. However, definitive conclusions can only be made if reflectance measurements are made for the ceramic coatings before and after being exposed to the engine environment.

#### MODELING OF SOOT EFFECT

The effect of soot deposit is one that can be modeled. Let us consider a thin layer of soot deposited on the coating. Since this deposit layer will be composed mostly of carbonaceous material it will readily absorb radiative heat flux incident on it. This layer can be modeled as a thin coating of negligible thermal capacity (uniform temperature) and having a negligible thermal resistance between the layer and the ceramic coating. Under these assumptions, in order to compute radiative and convective heat flux, it is sufficient to modify the boundary condition imposed on the first ceramic node point. A modification of the boundary condition has to be constructed, which is capable of modeling any state of fouling between clean to completely soot-covered ceramic coating. In the first limiting case the ceramic is clean and the incident radiation penetrates directly into the ceramic without first being absorbed by intervening deposit layer. In the second limiting case the incident radiation is totally absorbed by the deposit and the deposit reradiates into the ceramic coating. Intermediate levels of fouling are represented by a fouling factor (G) which can take any value between 0 and 1. A value of one for the fouling factor represents fully sooted surface and a value of zero represent clean surface. The intermediate values can be thought of as a representation of soot layer with varying degrees of surface coverage. The detail of the requisite boundary conditions representing this physical situation is described below.

Radiation energy passing through at the gas-soot layer interface (node #1)

$$Q(0) = q_1 - q_2 - q_3 + q_4 \quad (31)$$

where

$$q_1 = \int_0^{\lambda_c} (1-R_{do})(1-G) q_i d\lambda \quad (31a)$$

$$q_2 = \int_0^{\lambda_c} GE_{b\lambda} d\lambda \quad (31b)$$

$$q_3 = \int_0^{\lambda_c} (1-R_{di})(1-G)F^-(0)d\lambda \quad (31c)$$

$$q_4 = \int_{\lambda_c}^{\infty} (\alpha + G(1-\alpha))(q_i - E_{b\lambda}) d\lambda \quad (31d)$$

$E_{b\lambda}$  - blackbody emissive power

$G$  - fouling factor

$q_i$  - incident radiation

$\lambda$  - wavelength

$\lambda_c$  - 5  $\mu\text{m}$  - cutoff wavelength for transparent/opaque bands

$\alpha$  - surface absorptance in the opaque band

The first term of equation (31) represents the fraction of the incident energy within the transparent band of the ceramic (0 to  $\lambda_c$ ) that will be either absorbed or pass through the gas/ceramic interface. The second term represents emission of the soot deposit back to the radiating gas. The contribution of this term will be present only if there is a soot layer on the surface. The third term represents emitted and scattered radiation from within the coating escaping to the gas. Here again if the surface is fully sooted no radiation will escape and consequently this term will vanish. The final term of the equation (31) is the fraction of the incident energy within the opaque band ( $\lambda_c$  to  $\infty$ ) that is absorbed at the surface. This term is composed of the difference between the net incident energy (incident energy - reflected energy) and the emitted energy from the surface.

This deposited energy at the first node will be transported to the interior node by either conduction or radiation. The radiation flux at node #1 is given by

$$q_r(0) = F^+(0) - F^-(0) \quad (32)$$

where

$$F^+(0) = q_5 + q_6 + q_7 \quad (33)$$

$$q_5 = \int_0^{\lambda_c} (1-R_{do})(1-G)q_i d\lambda \quad (33a)$$

$$q_6 = \int_0^{\lambda_c} G n_i^2 E_{b\lambda} d\lambda \quad (33b)$$

$$q_7 = \int_0^{\lambda_c} R_{di} F^-(0) (1-G) d\lambda \quad (33c)$$

$F^+(0)$  - toward diffuse flux at node 1 (pointing to the interior)

$F^-(0)$  - backward diffuse flux at node 1 (pointing to the exterior)

The first term in equation (33) is the fraction of the incident radiation within the transparent band that passes through the gas ceramic interface. (Note that this term is different from  $q_1$  inasmuch as it does not include the radiation absorbed at the surface.) The second term is the radiation energy within the transparent band ( $0-\lambda_c$ ) that is emitted by the surface into the coating. The final term is the portion of the backward flux that reflected back into the coating by the front interface. (This is the complementary term of  $q_3$ .)

The conduction flux at node #1 is the difference between the total flux passing through the surface and the radiation flux at that node

$$q_c(0) = Q(0) - q_r(0) + h_g (T_g - T_w) \quad (36)$$

The other boundary conditions (e.g. at ceramic/metal interface and coolant/metal interface were given in a previous report (Wahiduzzaman and Morel<sup>6</sup>) and are only summarized here. The continuity condition for temperature at the ceramic/metal substrate is given by

$$T_c(d) = T_m(d) \quad (37)$$

where  $T_c$  and  $T_m$  are ceramic side and metal side temperatures respectively. The radiative flux boundary condition at ceramic/metal interface is

$$\int_0^{\lambda_c} F^-(d) d\lambda = \int_0^{\lambda_c} (\rho_s F^+(d) + \epsilon_s E_{b\lambda}) d\lambda \quad (38)$$

and at the gas/ceramic interface it is

$$\int_0^{\lambda_c} F^+(0) d\lambda = \int_0^{\lambda_c} (R_{di} (1-G) F^-(0) + (1-R_{do}) (1-G) q_1) d\lambda \quad (39)$$

This model was checked by setting it to zero to obtain clean ceramic results. It should be noted here that the above integrations are performed over several discrete bands, as discussed in a previous section.

The results obtained with this model are presented in Table III. With G set to unity (fully sooted case) the results labeled "sooted" were generated. It can be seen that the presence of the soot layer produced a variety of changes. The surface temperature was increased, one big reason being the elimination of surface reflectivity by the presence of black soot and the resultant higher radiative flux. A larger change is seen in the surface swing, this being caused by the absorption right at the surface rather than sub-surface. The convective flux was reduced due to the higher surface temperature. Finally, the total heat flux was generally slightly increased, except for the fully transparent case where a 10% reduction was seen due to the reduction in convective flux. It is very interesting to see that even though the soot reduced the differences in total heat flux between the fully transparent and the fully opaque cases from 70 to 49 kW/m<sup>2</sup>, there remained a substantial difference between them. This indicates that if the ceramic surface temperature is high, exceeding 1000K, the time-averaged radiation produced by the soot layer with its high emissivity, approaches and can exceed that produced by the much hotter diesel flame, which however persists for only a short fraction of time. It is also interesting to note that for the two opaque limits soot deposit produces identical results. This is because in the opaque(s) case soot emitted radiation is reabsorbed by the soot layer after being scattered back to the soot/ceramic interface. This is very similar to the situation existing in opaque (a) case where soot radiation is quickly absorbed by the coating close to the soot/ceramic interface.

The distributions of the radiative, convective and total heat fluxes computed for different levels of fouling on a 2.5 mm ceramic (type C) coating are given in Figure 18. As the fouling level is increased, the radiative transport increases sharply, accompanied by a decrease in the convective heat flux. This decrease in convective heat flux is due to increase in surface temperature caused by the absorption of radiative energy at the surface. As expected, at the highest fouling level ceramic C behaves more like the sooted opaque limiting case rather than the transparent one (Table III). This is again due to absorption of radiative energy at or very near the surface as in opaque limiting case. There is a small increase in total heat flux with increasing surface fouling, indicating that although the mechanism of the heat transfer changes with surface fouling, its impact on total heat transfer is moderate at least for the boundary conditions used in this study.

## DISCUSSION

The radiative properties of several existing ceramic coating materials were determined experimentally. This involved measuring reflectance and transmittance spectra of the ceramic samples of different thicknesses. A model describing the radiative transport process in semitransparent media was then fitted to the measured data sets. An optimization technique was used to adjust the radiative parameters until the best fit to the measured data was obtained.

The model describes the radiative transport within a medium using three parameters, namely scattering coefficient, absorption coefficient and

refractive index. This involves some implicit assumptions about interfacial reflectivities and phase function for the material. It was assumed that the interfacial reflectivities can be approximated by Fresnel's equations which is strictly valid for optically smooth surfaces ( $l/\lambda < 1$ , where  $l$  is the rms value of the surface roughness). It was determined a posteriori that accurate determination of the surface reflectivities was not critical in view of the high scattering coefficients of the studied ceramics. Another assumption regards the phase function. Since the grains of the ceramic are densely packed and randomly oriented, there will not be any preferred direction of scattering. Consequently, a uniform scattering phase function was used. In addition, a fixed value of refractive index, as discussed earlier, was used for all parameter inversion. This constraint had to be imposed due to the small number of independent data points available, keeping in mind that the results are relatively insensitive to this parameter (Wahiduzzaman and Morel<sup>7</sup>).

In order to perform a systematic error analysis on the inverted parameter, the variability of the measured quantities (reflectance and transmittance spectra) are needed. This in turn depends on the repeatability of the sample preparation and uncertainty associated with the measuring instrument. Unfortunately, such study was beyond the scope of this program, since the number of samples of each material used in this study was limited and no repeat measurements with identical samples were made. Nevertheless, based on the assumptions involved in the technique and an expected variability in measured data, an assessment can be made of the uncertainty in the values of the inverted parameters. For errors of  $\pm 5\%$  in transmittance,  $\pm 5\%$  in reflectance and  $\pm 25\%$  in refractive index, maximum error over the entire spectral range was found to be of the order of 53% for scattering coefficients and 40% for absorption coefficient. Although this uncertainty appears to be large, the calculated heat transfer is affected only moderately over this range. Thus despite the uncertainty in the properties, the basic results of this work, as discussed below, should be unchanged.

The ceramic samples used in this study were found to be highly reflective (reflectance on the order of 0.6-0.8) and optically thick even at thicknesses of the order of 2 mm. In particular, the reflectance spectrum of the 0.51 mm sample (A1) of PSZ obtained from Plasma Technics was nearly identical to the corresponding spectra belonging to 1.092 and 5.74 mm samples (A2 and A3) indicating that even the thinnest sample is optically thick. It should be noted here that the level of reflectances of the material A are of same order as for materials B and C, and thus the scattering coefficients are probably of similar magnitude. However, the ceramics B and C are less optically dense than the ceramic A, based on the fact that their reflectances display dependence on thickness between 0.5 and 1 mm not seen in ceramic A. Thus one may make the indirect inference that the ceramic A has much higher absorption coefficients than ceramics B and C.

Ceramics B and C (PSZ and TZP from Oak Ridge Laboratory) were found to be radiatively identical. The reflectance and transmittance spectra of both materials were nearly identical and consequently, inverted properties were

also the same for both materials. The absorption coefficients of ceramics B, C and D were found to be much smaller than the scattering coefficients (high scattering albedo). Material B and C possessed lower scattering and absorption coefficients in comparison to material D and hence, were optically less dense than D. In any event, a nominal thickness of 2 mm of any of the studied ceramics would be sufficient to prevent any appreciable radiation energy from reaching the metal substrate.

The radiative parameters of the materials studied here exhibited both qualitative and quantitative agreement with previous measurements (Makino et al<sup>4</sup>) made on a number of monolithic ceramics including zirconia (85% ZrO<sub>2</sub> by weight, with Y<sub>2</sub>O<sub>3</sub>, SiO<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>). The scattering coefficients obtained in that study displayed similar decreasing trends at higher wavelengths, although the values of the coefficients were lower (12000 m<sup>-1</sup> at shorter wavelengths). The absorption coefficients were of similar order of magnitude as those found here and the spectrum contained sharp rise around 4 μm, similar to that evidenced in the spectrum of ceramic C. The lower scattering coefficient found for the monolithic ceramics by Makino et al can be attributed primarily to the difference in microstructure of the two ceramics. To quantify the differences between the present coatings and Makino's monolithics, an analysis was carried out of the heat barrier effectiveness of a monolithic ceramic 2.5 mm thick subject to the same boundary conditions as done for the coatings in Table III. The results showed that for Makino's monolithic ceramics the radiation heat transfer was larger by 30% and the total heat transfer was 8% higher. This shows that these ceramics were less optically opaque and thus less effective as barriers to heat radiation than the present coatings (at the same thickness and assuming the same thermal conductivity and heat capacity).

A heat transfer analysis based on these radiative parameters showed that at the larger thicknesses (above 1.25 mm) materials C and B are more effective against radiative heat transfer than material D of same thickness. At lower thicknesses the situation is reversed. This trend is a consequence of reflectivity of the coatings which can be substantially influenced by the ceramic/metal interface depending on the coating thickness.

In general, the studied ceramics exhibited similar characteristics in terms of preventing radiative heat transfer. The scattering albedo of these ceramics were found to be large, causing them to be fairly reflective (60-80%) and optically thick at conventional coating thicknesses (1 to 2 mm). As a result, most of the radiation that penetrates the coating is absorbed within the coating as opposed to at ceramic/metal interface. Since a significant portion of this absorption occurs at or near the surface, the surface temperature is increased. This has a beneficial effect of reducing the convective component of the total heat transfer.

In an operating engine, the coating surface will lose a lot of its reflectivity due to carbonaceous deposits. In such a case the incident radiant energy will be readily absorbed by the deposit layer causing the surface temperature to go up even higher. This will further reduce the convective heat flux and partially compensate for the increased radiant heat transfer at the surface (less reflected radiation). On the other

hand, the radiation of the surface deposit layer towards the interior of the ceramic increases rapidly with the increasing surface temperature and at high enough temperature could exceed the radiation from the flame (because it has high emissivity and acts all the time, whereas the gas radiates only some 5-7 percent of the time). It should be mentioned that when the surface temperature becomes sufficiently high the soot particles in the deposit layer will tend to burn off. Thus the ceramic will tend to return to its original clean state, probably before the point of excessive surface radiation is reached.

Based on the present results and the above observations, one can speculate on the ideal thermal barrier for engine applications. To prevent radiant heat transfer, the coating should be highly reflective, for example through a combination of high scattering coefficient ( $>20000 \text{ m}^{-1}$ ) and low absorption coefficient. If a homogeneous ceramic satisfying these conditions could not be found, one could contemplate a composite ceramic, or one having a thin reflective and opaque metallic coating on the surface. However, if significant soot deposition occurs at the coating surface a large portion of the incident radiation energy will be trapped by the deposit layer causing a substantial increase in radiation heat transfer. As mentioned previously, this will be associated with an increase in surface temperature. The level of this increase will depend on the location where the energy reradiated by the deposit layer is absorbed. If the coating is opaque it will be absorbed at or near the interface and the increase in surface temperature will be large which will in turn cause a large reduction in convective heat flux. This opacity could be achieved, as evidenced in Table III, either through high scattering coefficient or high absorption coefficient or a combination of both. In the case of transparent ceramic the reradiated energy will be absorbed at the coating/metal interface, leaving the surface at a lower temperature (Table III) and consequently the reduction in the convective heat flux will be small. Hence, to insure the best thermal barrier for either clean or heavily sooted surfaces, a ceramic material with high scattering and low absorption coefficient provides the best choice. To reduce the convective heat transfer relatively independently of the radiant heat transfer, one can follow the conventional approach of choosing a material having low thermal conductivity and as large as practicable coating thickness.

#### CONCLUSIONS

- 1) An experimental study was carried out to determine the radiative properties of four ceramics; A (PSZ, 13% yttria stabilized), B (PSZ), C (TZP) and D (PSZ, 24% ceria stabilized). Ceramics B and C were found to possess high scattering coefficient (in excess of  $30000 \text{ m}^{-1}$  in short wavelengths) and low absorption coefficients. Ceramic D had even higher scattering coefficients (in excess of  $40000 \text{ m}^{-1}$  at short wavelengths) and moderate absorption coefficients ( $2000 \text{ m}^{-1}$  at  $3 \mu\text{m}$ ). Although parameter inversion was not possible for material A, a comparison of its reflection spectra with corresponding spectra of other ceramics tested indicates it to be the most optically dense material in the group.

2. In terms of overall radiation effect, these coatings were found to be highly reflective (reflectance around 0.6-0.8) due to their high scattering coefficients. They also were found optically thick even at thicknesses of 2 mm, i.e., permitting only a small fraction of radiation to pass through to the substrate. Comparison to literature data on monolithic zirconia ceramics showed those to be more transparent and thus less effective as barriers to heat radiation.
3. When compared to the limiting cases of completely transparent and completely opaque, the total heat transfer through the studied ceramics for 2.5 mm thickness was substantially closer to either of the opaque limits than to the transparent limit. However, when the surface is clean, the mechanism of the heat transfer more closely approached the opaque(s) limit rather than the opaque(a) limit. On the other hand, for a sooted surface, both opaque limits converged into a single limit.
- 4) With regard to the most desirable ceramic properties for diesel engine applications, it was concluded that a highly scattering ceramic with low absorption coefficient would be the best heat barrier material for blocking thermal radiation. An alternative would be a composite ceramic coated with an opaque high reflectance surface layer. From the point of view of convective heat transfer, any scheme aimed at raising surface temperature will be effective in reducing convective heat transfer, and low thermal conductivity is an essential element in this respect. However, other factors such as volumetric efficiency, thermal stress, etc. have to be considered in selection of an appropriate insulation package.
- 5) In experimental determination of radiative properties of ceramics it is highly desirable to perform preliminary transmittance measurements to obtain some knowledge on the opacity of the material. This knowledge can then be used for proper selection of material thicknesses to obtain samples ranging from optically thin to optically thick. In order to extract the refractive index, alternate type of measurements are recommended. These may include separate measurements of diffuse and directional reflectance and transmittance, and/or measurement of polarized components of the reflectance.
- 6) Both four-flux and two-flux methods were used and it was concluded that the four-flux method is more appropriate for parameter inversion models than the simpler two-flux method. This method is better equipped to handle collimated illumination and has the ability to predict both diffuse and directional quantities. It was found that the two-flux model underpredicts scattering coefficient substantially when the scattering level is low.

## NOMENCLATURE\*

A	-	$3\omega/(8\beta^2-2)$
B	-	A/3
C <sup>+</sup>	-	collimated forward flux
C <sup>-</sup>	-	collimated backward flux
d	-	depth of the layer
D <sup>+</sup>	-	diffuse forward flux
D <sup>-</sup>	-	diffuse backward flux
E	-	s+k = extinction coefficient
E <sub>bλ</sub>	-	blackbody emissive power
F <sub>d</sub>	-	diffuse incident flux
F <sub>c</sub>	-	collimated incident flux
G	-	fouling factor
h <sub>g</sub>	-	heat transfer coefficient
k	-	absorption coefficient
n	-	refractive index
p	-	phase function
q <sub>i</sub>	-	incident radiation
R <sub>di</sub>	-	diffuse interfacial internal reflection
R <sub>do</sub>	-	diffuse interfacial external reflection
R <sub>ci</sub>	-	collimated interfacial internal reflection
R <sub>co</sub>	-	collimated interfacial external reflection
R <sub>d</sub>	-	diffuse reflectance
R <sub>h</sub>	-	hemispherical reflectance
S <sub>c</sub>	-	reflected collimated flux
S <sub>d</sub>	-	reflected diffuse flux
s	-	scattering coefficient
T <sub>c</sub>	-	ceramic side temperature at the interface
T <sub>d</sub>	-	diffuse transmittance
T <sub>g</sub>	-	gas temperature
T <sub>h</sub>	-	hemispherical transmittance
T <sub>m</sub>	-	metal side temperature at the interface

\*All variables are in S.I. units except where noted.

$T_w$  - surface temperature of the wall  
 $\alpha$  - surface absorptance in the opaque band  
 $\alpha_1$  - linear constant for flux equation  
 $\alpha_2$  - linear constant for flux equation  
 $\alpha_3$  - linear constant for flux equation  
 $\alpha_4$  - linear constant for flux equation  
 $\beta$  -  $\sqrt{1-\omega}$   
 $\lambda$  - wavelength  
 $\omega$  -  $s/(s+k)$  - albedo  
 $\tau$  -  $(k+s)x$  - optical depth at  $x$   
 $\tau_d$  -  $(k+s)d$  - optical thickness of the entire translucent layer  
 $\lambda$  - wavelength  
 $\lambda_c$  -  $5 \mu\text{m}$  - cutoff wavelength for transparent/opaque bands

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Table I. List of Ceramic Samples

<u>Set</u>	<u>Sample #</u>	<u>Material</u>	<u>Supplier</u>	<u>Thickness (mm)</u>
A	1	PSZ	Plasma Tech	0.51
A	2	PSZ	Plasma Tech	1.02
A	3	PSZ	Plasma Tech	5.74
B	4	TZP	Oak Ridge Lab	0.50
B	5	TZP	Oak Ridge Lab	1.00
B	6	TZP	Oak Ridge Lab	10.00
C	7	PSZ	Oak Ridge Lab	0.50
C	8	PSZ	Oak Ridge Lab	1.00
C	9	PSZ	Oak Ridge Lab	10.00
D	10	PSZ	Caterpillar	0.41
D	11	PSZ	Caterpillar	0.58
D	12	PSZ	Caterpillar	1.04
D	13	PSZ	Caterpillar	2.49

Table II. Matrix of Radiative Measurements

<u>Sample ID</u>	<u>Reflectance</u>		<u>Transmittance</u>	
	<2.5 $\mu\text{m}$	2-20 $\mu\text{m}$	<2.5 $\mu\text{m}$	2-20 $\mu\text{m}$
A1	✓	✓	s/u	✓
A2	✓	✓	l/s	✓
A3	✓	✓	n/r	n/r
B4	✓	✓	✓	✓
B5	✓	✓	✓	✓
B6	✓	✓	n/r	n/r
C7	✓	✓	✓	✓
C8	✓	✓	✓	✓
C9	✓	✓	n/r	n/r
D10	✓	✓	✓	✓
D11	✓	✓	l/s	l/s
D12	✓	✓	l/s	l/s
D13	✓	✓	n/r	n/r

Key: ✓ - measurement made; s/u - sample unavailable; n/r - measurement not requested; l/s - measurement attempted but failed due to low signal-to-noise ratio.

Table III. Effect of Ceramic Properties and Surface Fouling on Time-Averaged Heat Flux and Temperatures

<u>Ceramic</u>	<u>Surface Temp.</u>	<u>Indepth* Temp.</u>	<u>Surface Swing</u>	<u>Radiative Flux</u>	<u>Convective Flux</u>	<u>Total Flux</u>
	K		K	kW/m <sup>2</sup>	kW/m <sup>2</sup>	kW/m <sup>2</sup>
Metal baseline	701.9		25.2	86.5	316.2	402.8
Clean C	943.5	650.0	90.7	22.0	84.0	106.0
Clean D	953.7	649.5	90.4	25.8	75.2	101.0
Transparent	937.5	657.0	91.7	91.3	89.2	180.5
Opaque (a) #	995.2	650.0	116.6	82.2	28.1	110.3
Opaque (s) #	935.3	648.6	92.1	0.7	91.1	91.8
Sooted C	1001.4	651.0	122.3	97.4	20.6	118.0
Sooted D	1001.3	650.7	121.4	96.8	18.1	114.9
Sooted trans.	948.8	655.2	131.9	97.6	65.2	162.8
Sooted opaque (a)	1005.8	650.6	121.2	96.4	17.1	113.5
Sooted opaque (s)	1005.6	650.6	121.2	96.5	17.1	113.6

\*Temperature at the ceramic/metal interface, i.e. at a depth of 2.5 mm from the surface.

#Opaque (a) is the opaque limit attained by high absorption and negligible scattering.

#Opaque (s) is the opaque limit attained by high scattering and negligible absorption.

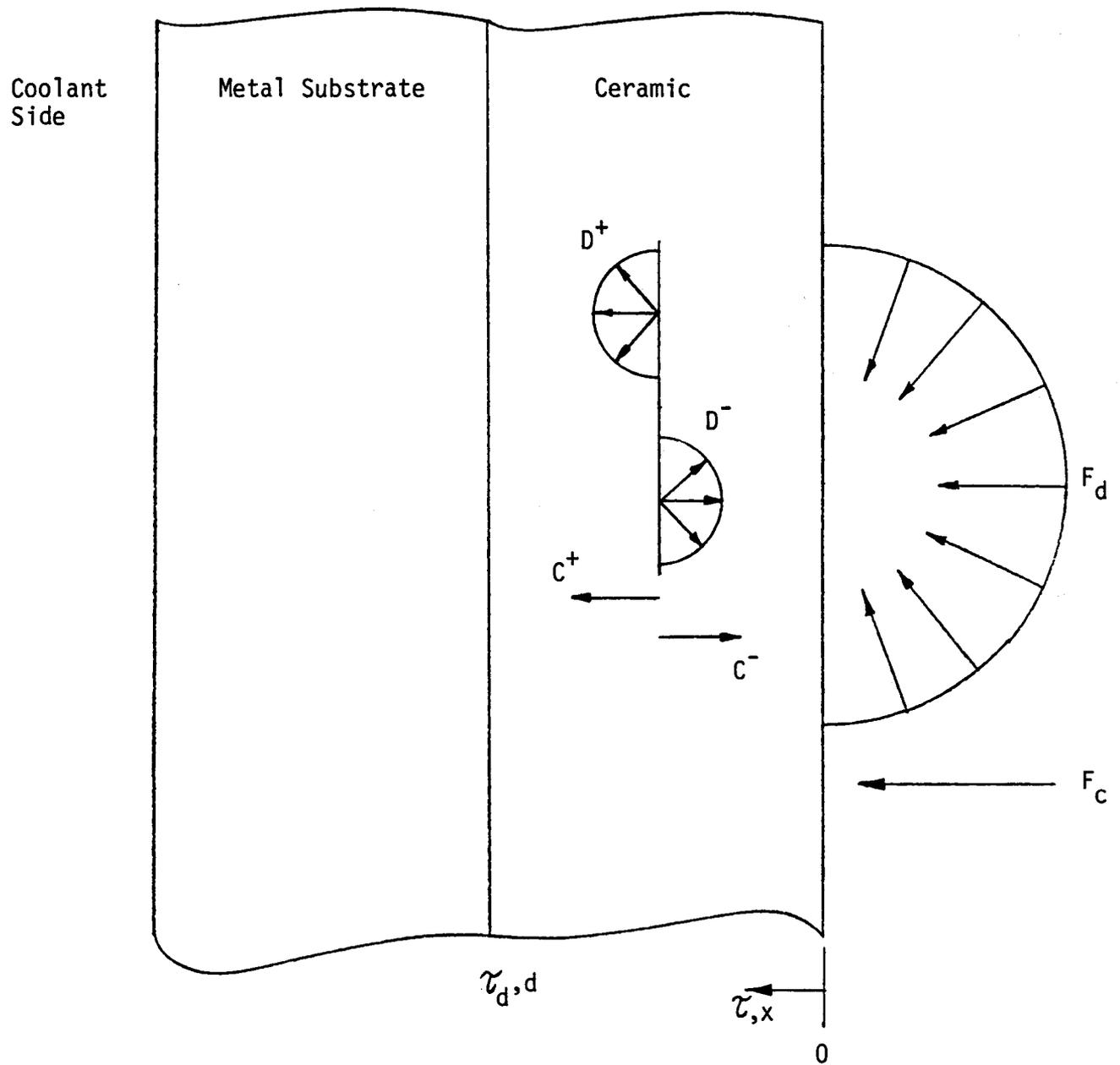


Figure 1. Physical model of the insulating coating.

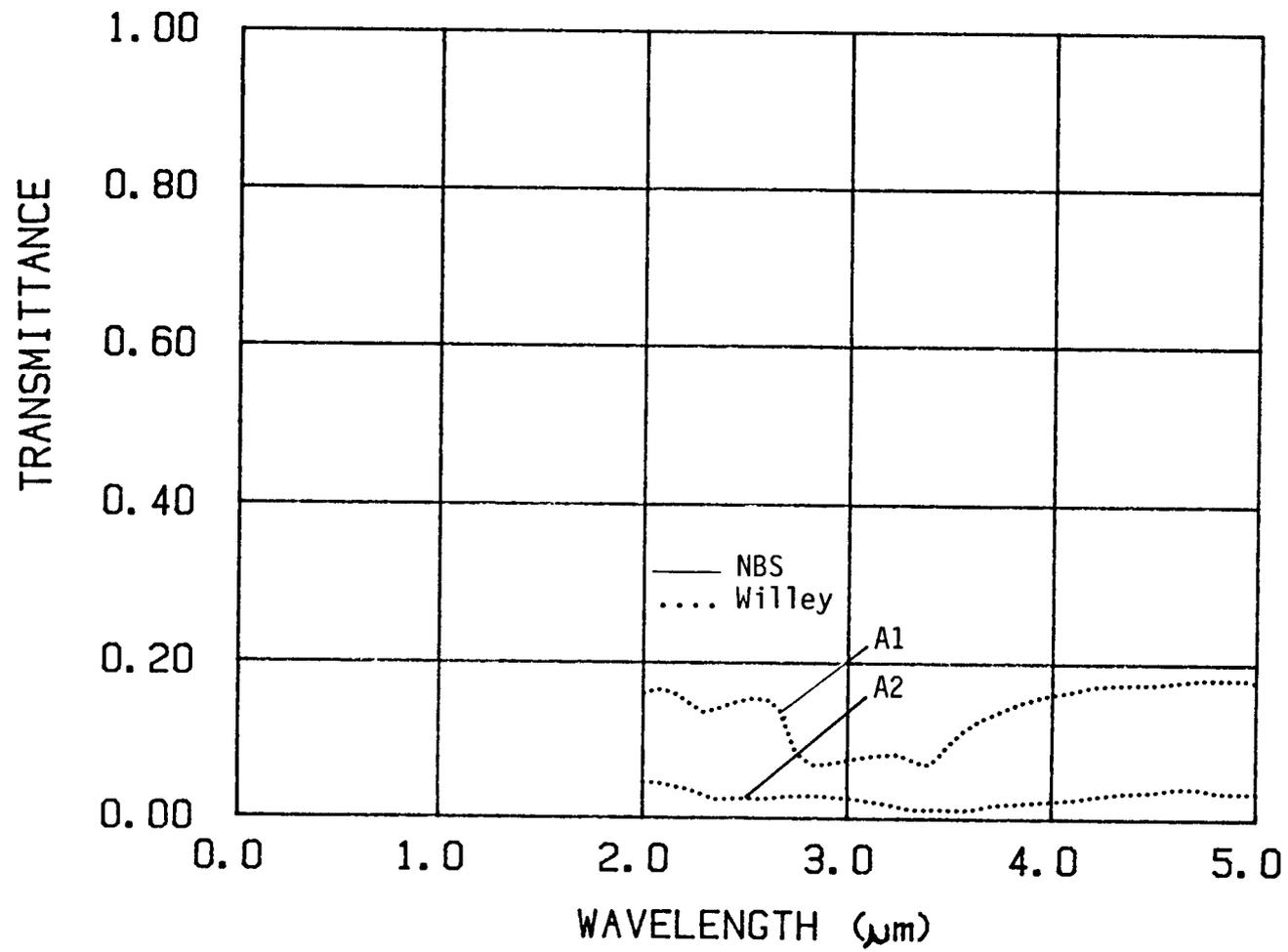


Figure 2(a). Transmittance spectra of material A (PSZ, 13 yttria stabilized) from Plasma Techniques.

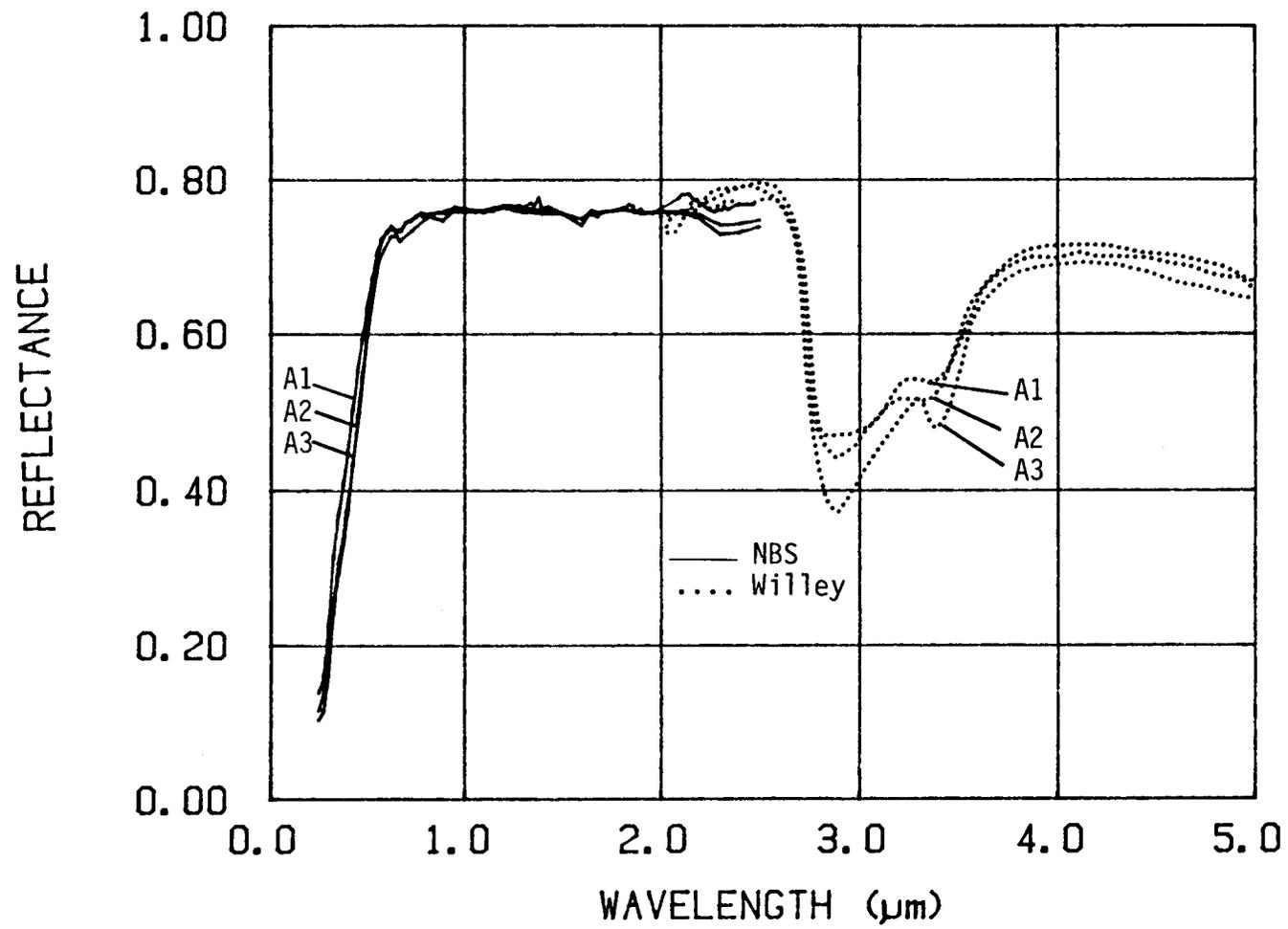


Figure 2(b). Reflectance spectra of material A.

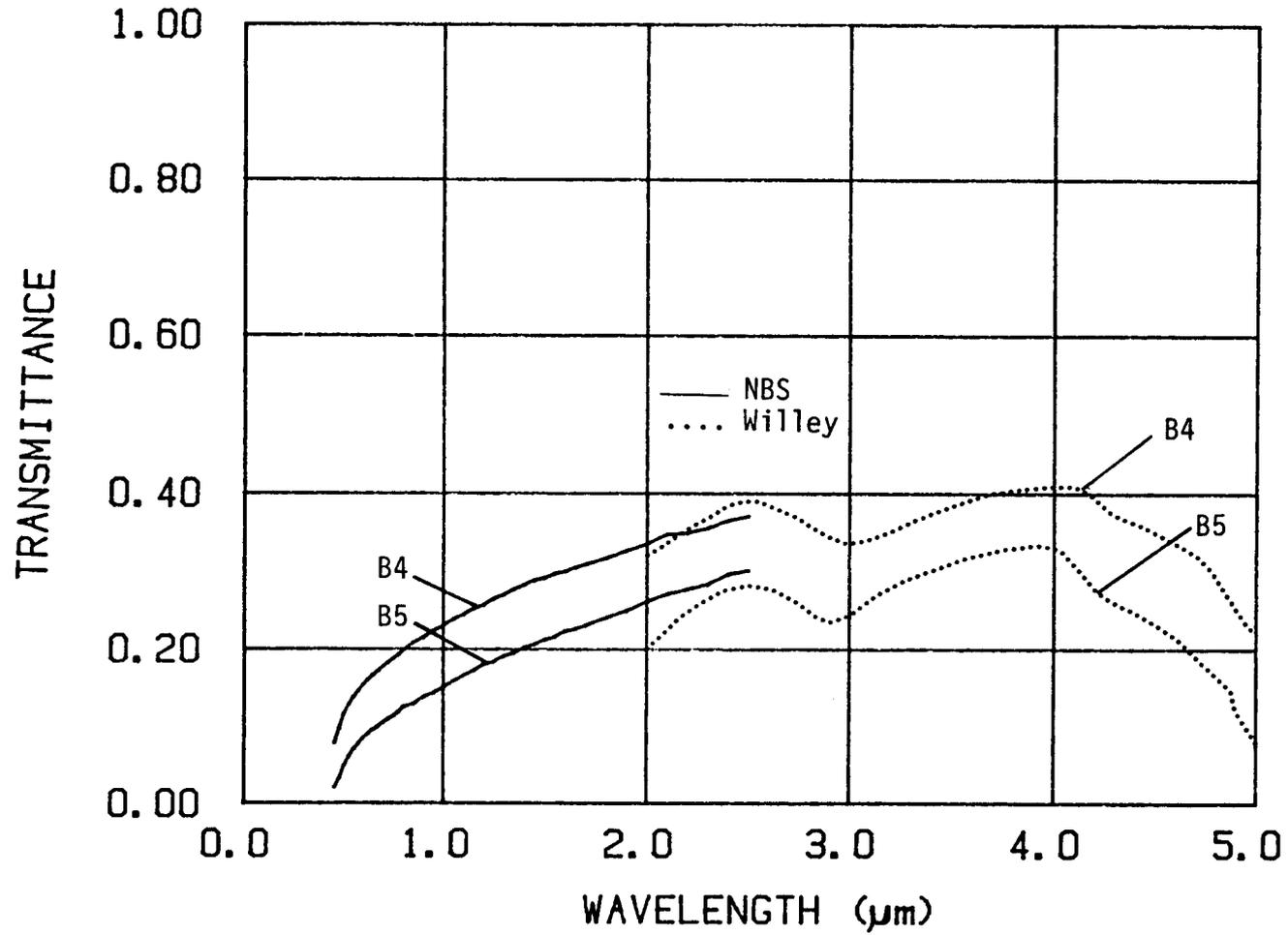


Figure 3(a). Transmittance spectra of material C (PSZ) from Oak Ridge National Laboratory.

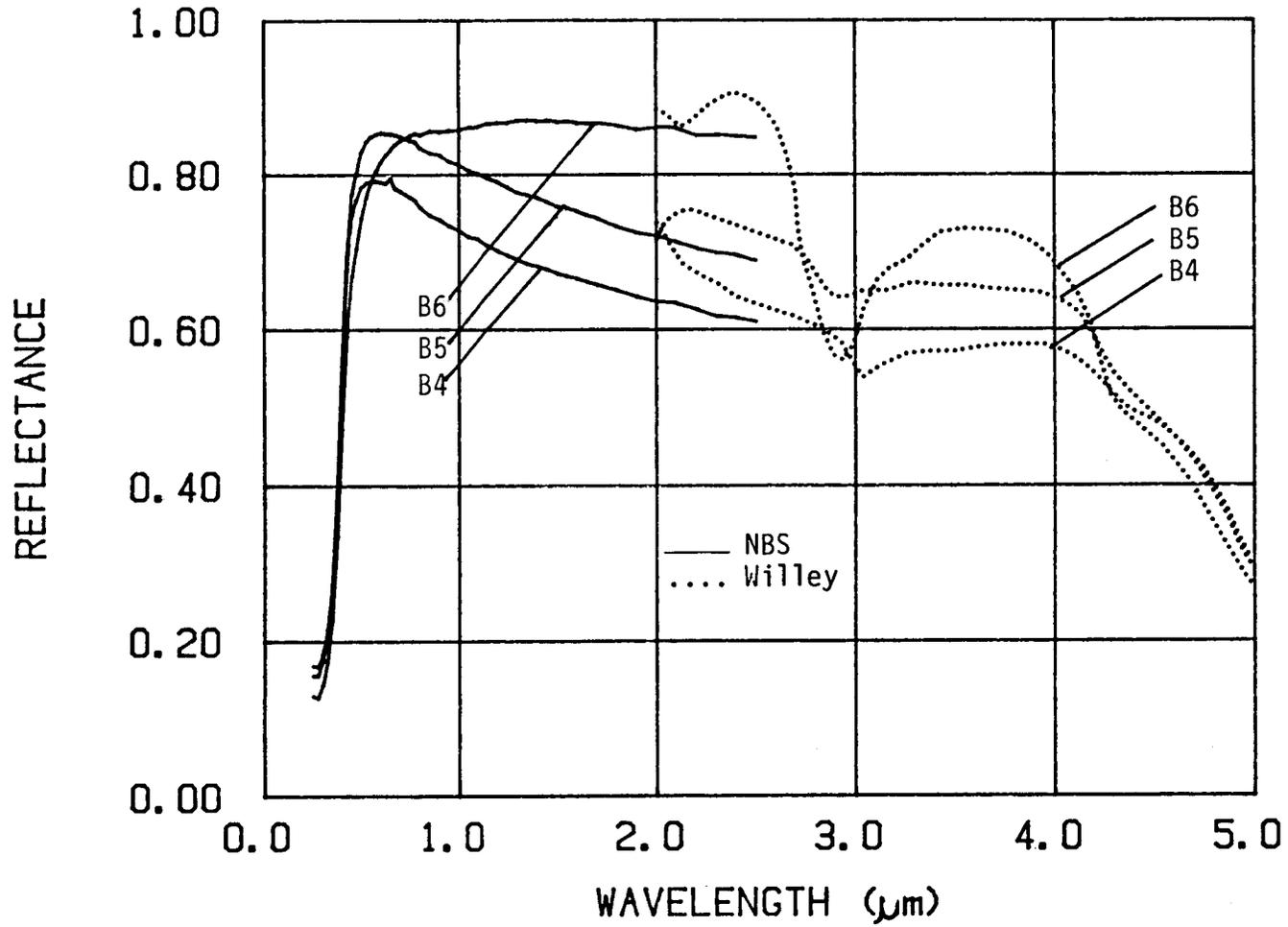


Figure 3(b). Reflectance spectra of material C.

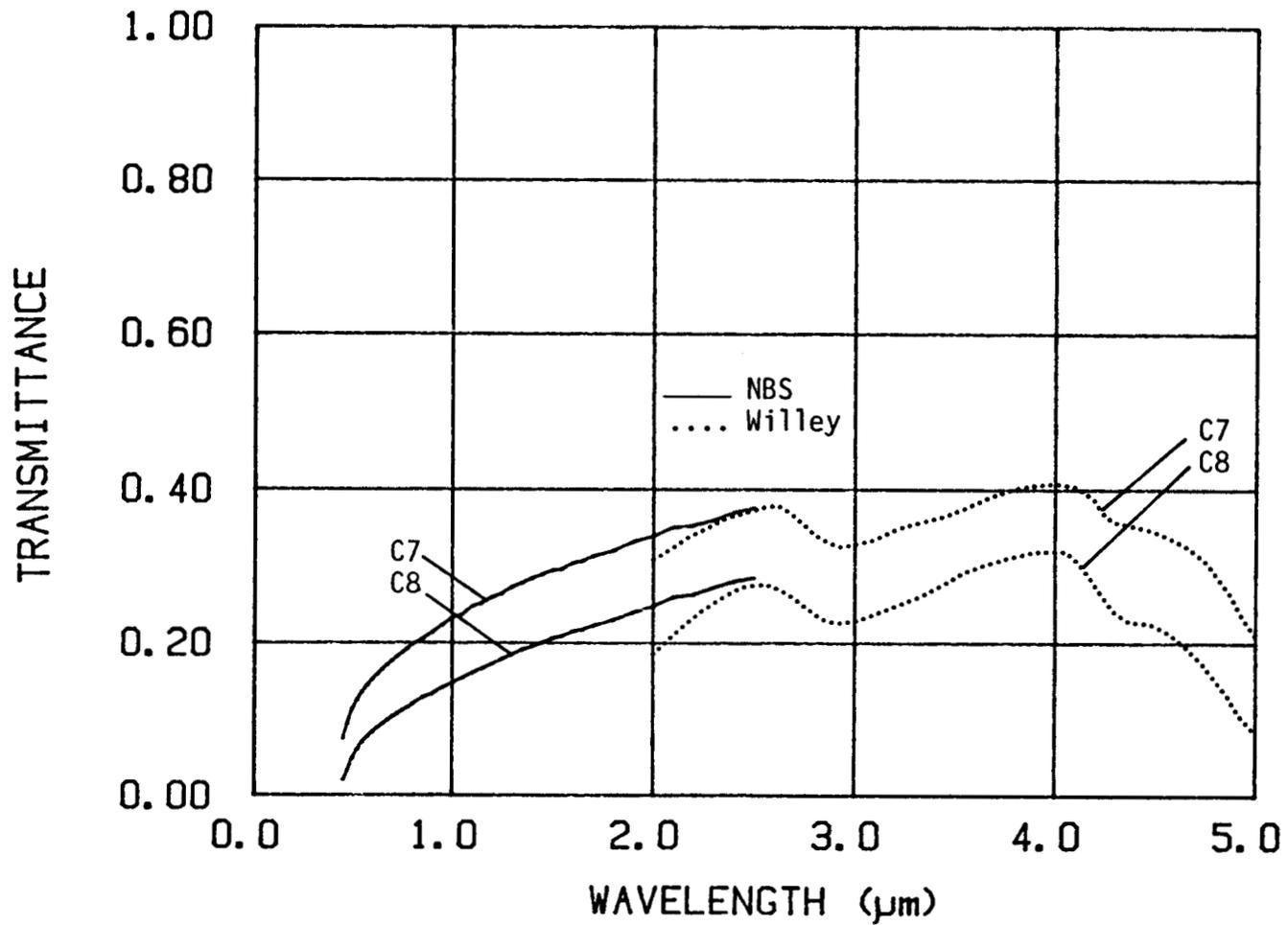


Figure 4(a). Transmittance spectra of material C (PSZ) from Oak Ridge National Laboratory.

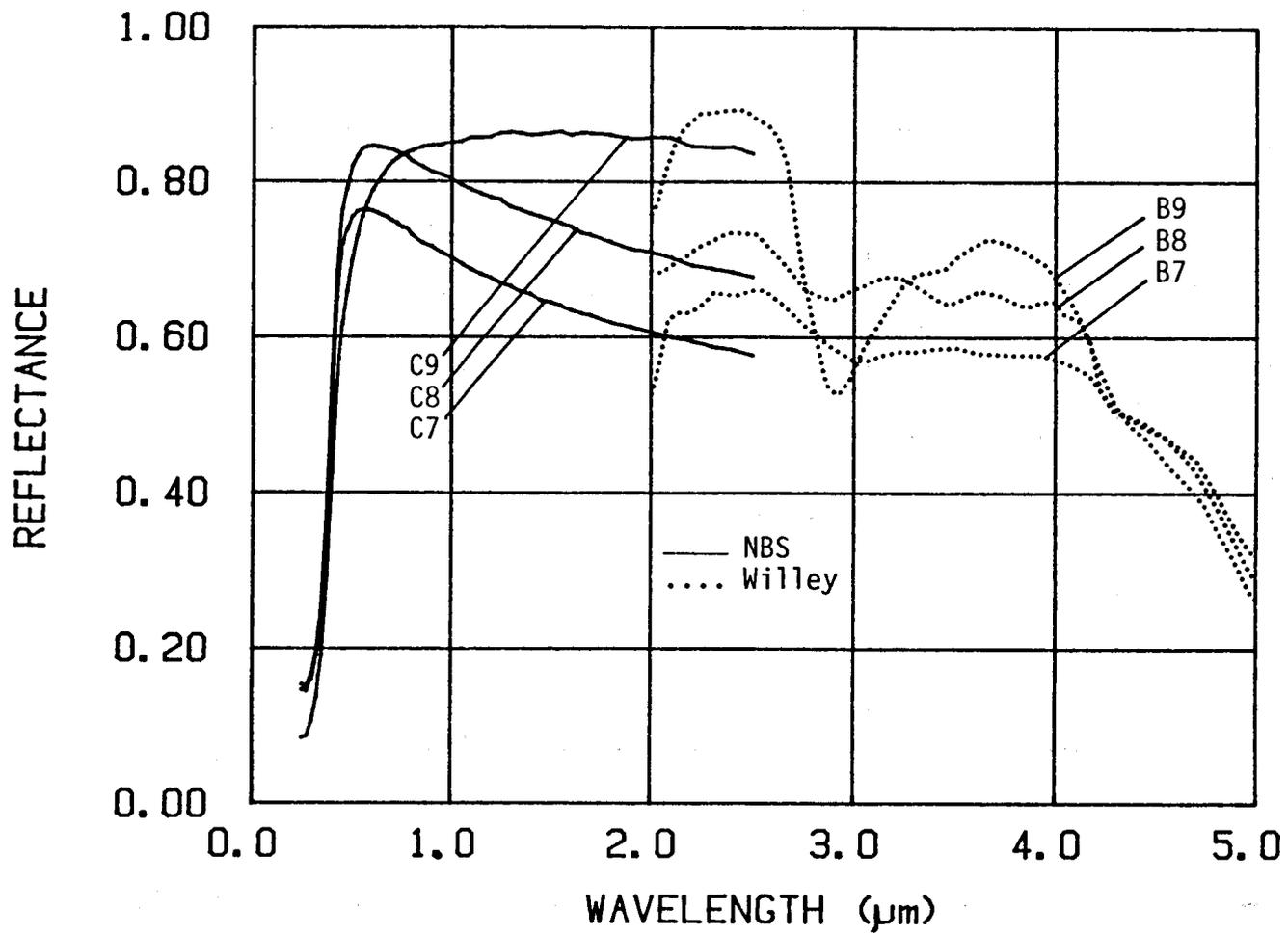


Figure 4(b). Reflectance spectra of material C.

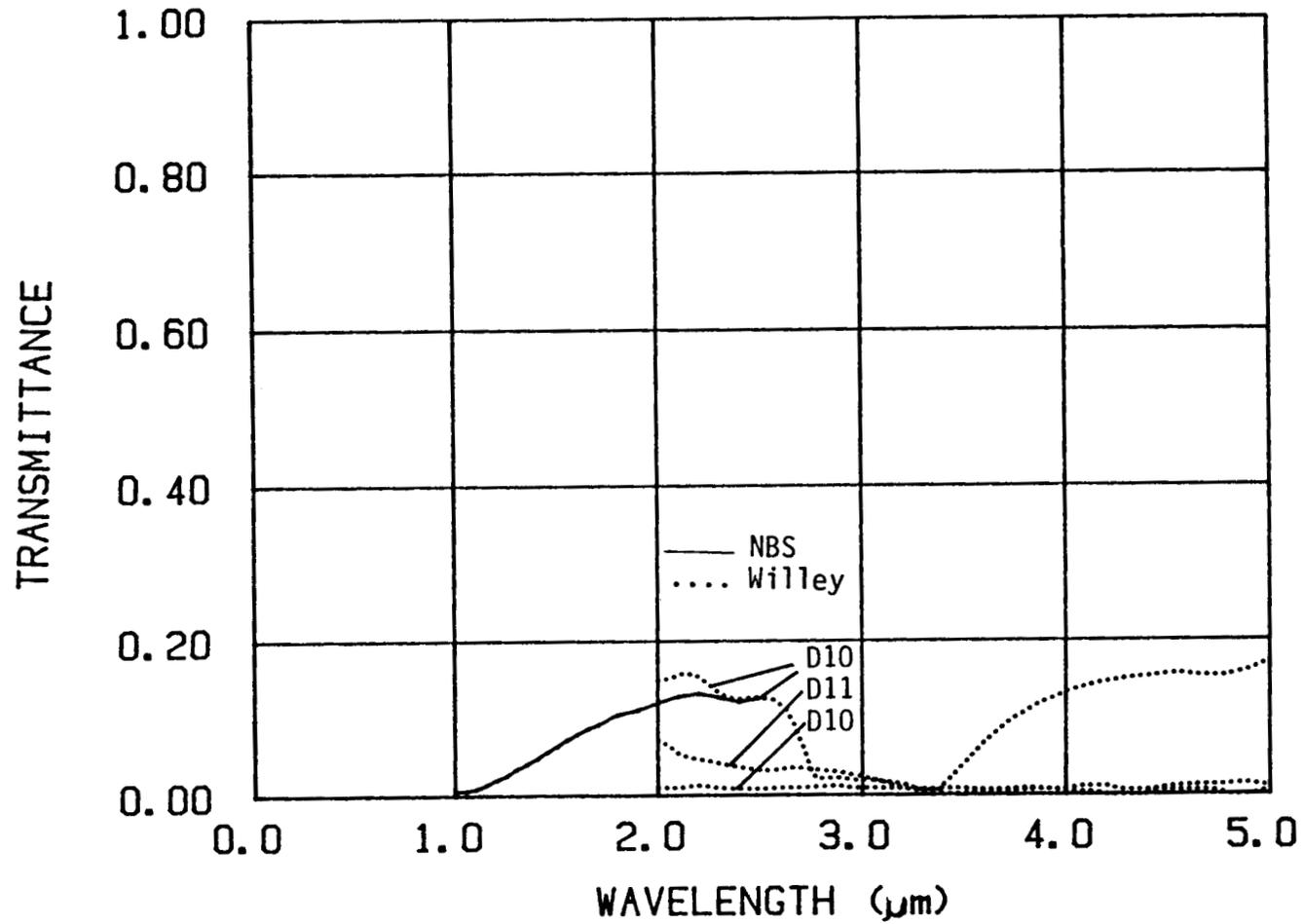


Figure 5(a). Transmittance spectra of material D (PSZ, 241 ceria stabilized) from Caterpillar.

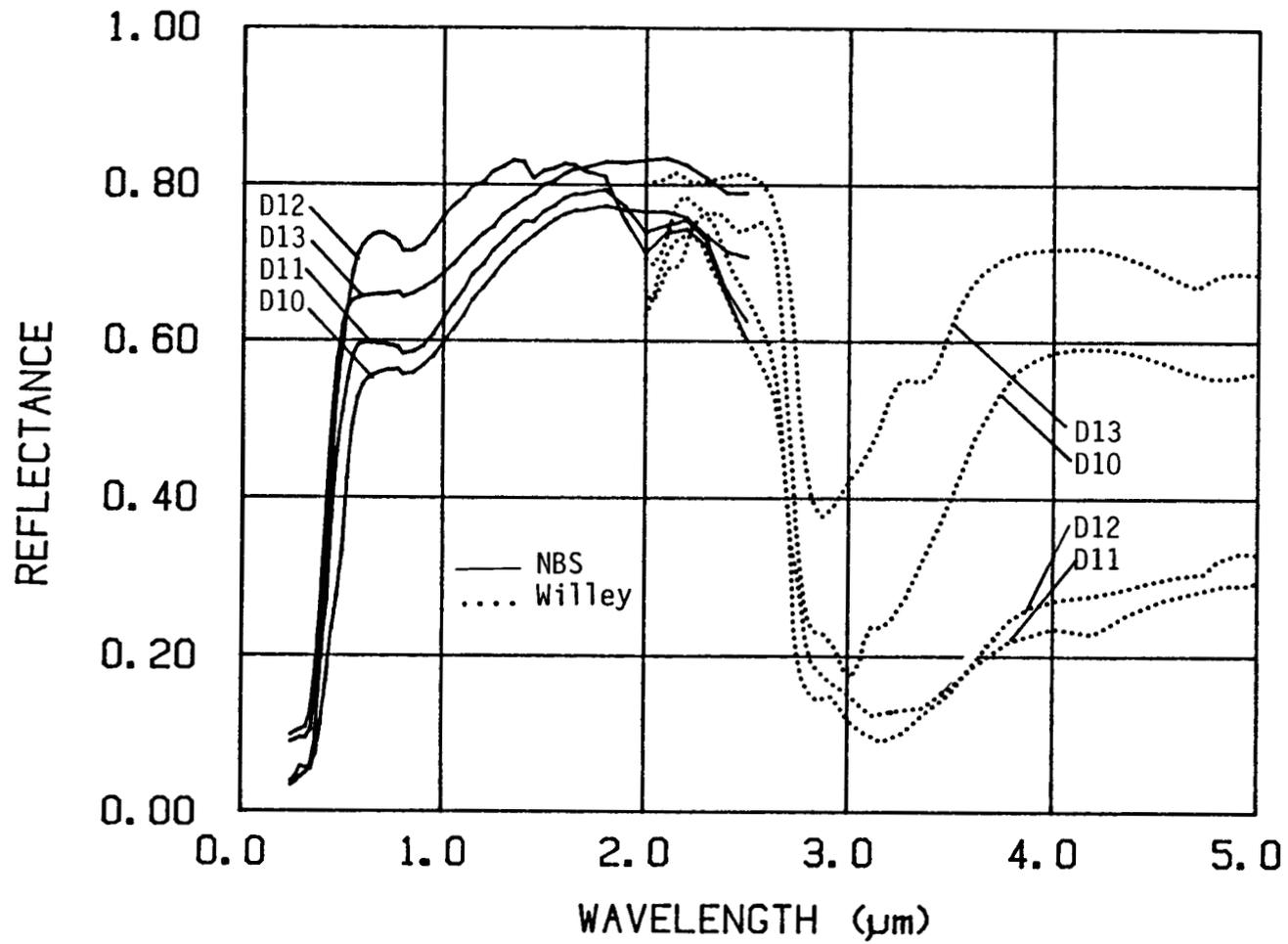


Figure 5(b). Reflectance spectra of material D.

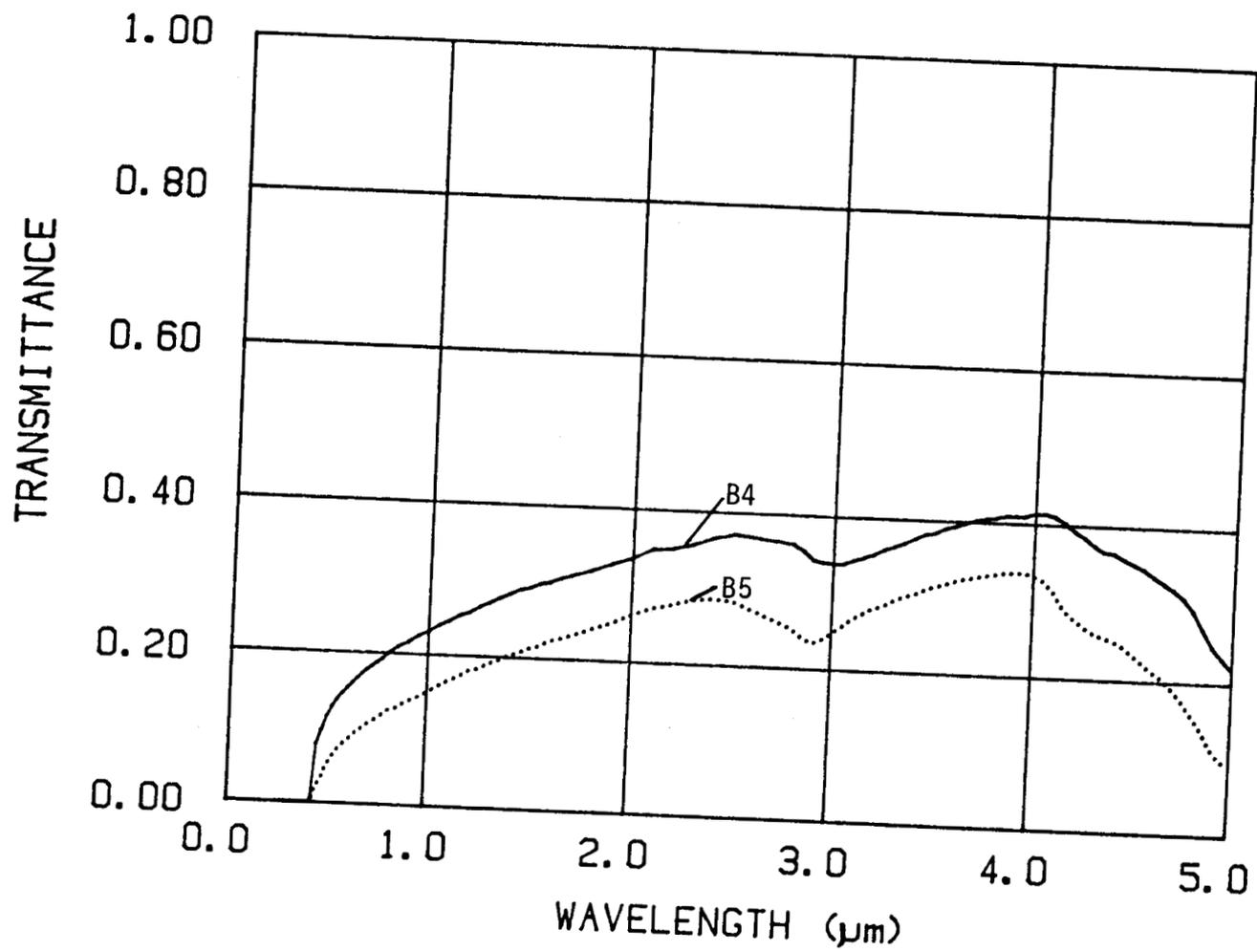


Figure 6(a). Merged transmittance spectra of material B used for parameter inversion.

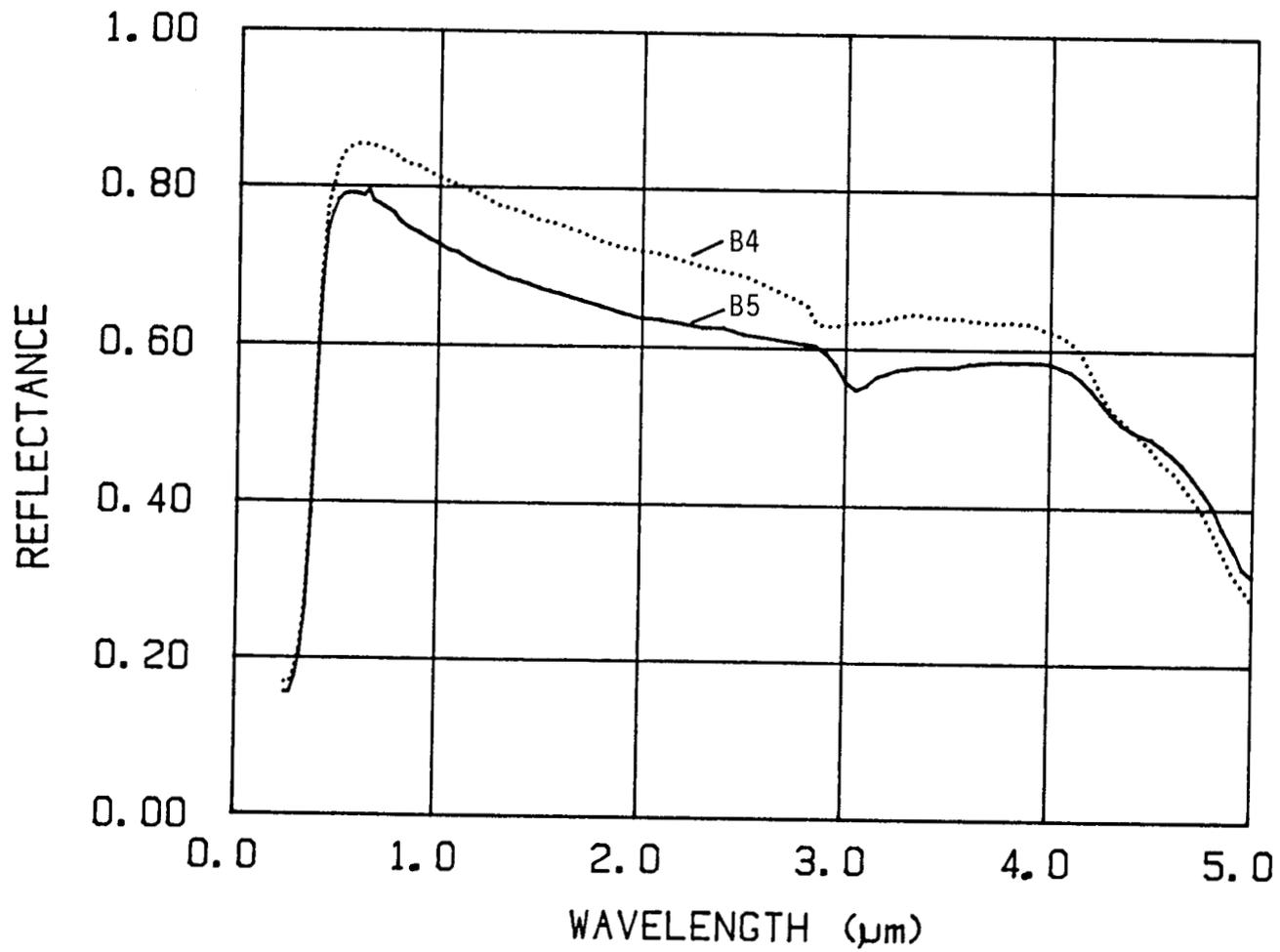


Figure 6(b). Merged reflectance spectra of material B used for parameter inversion.

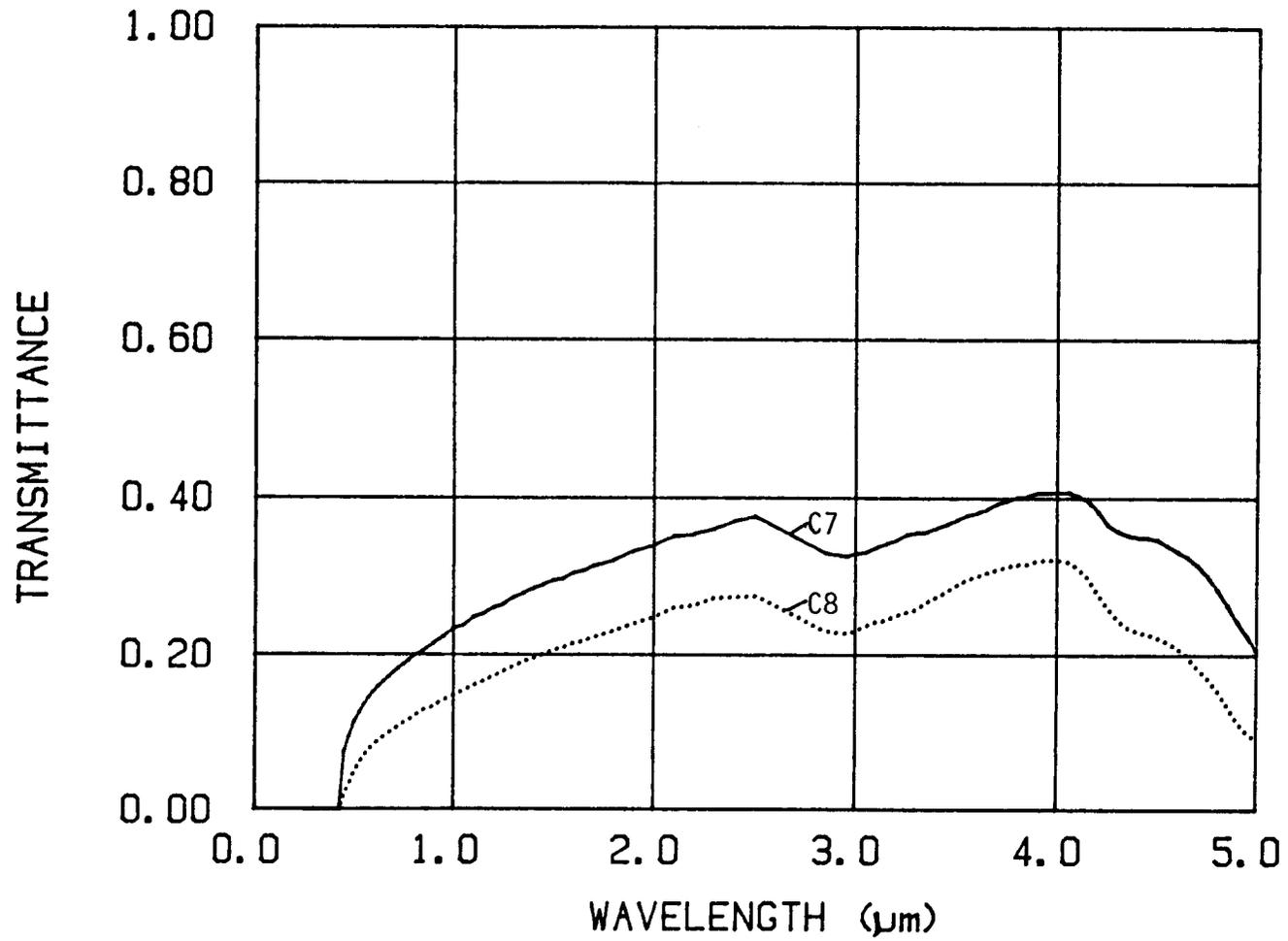


Figure 7(a). Merged transmittance spectra of material C used for parameter inversion.

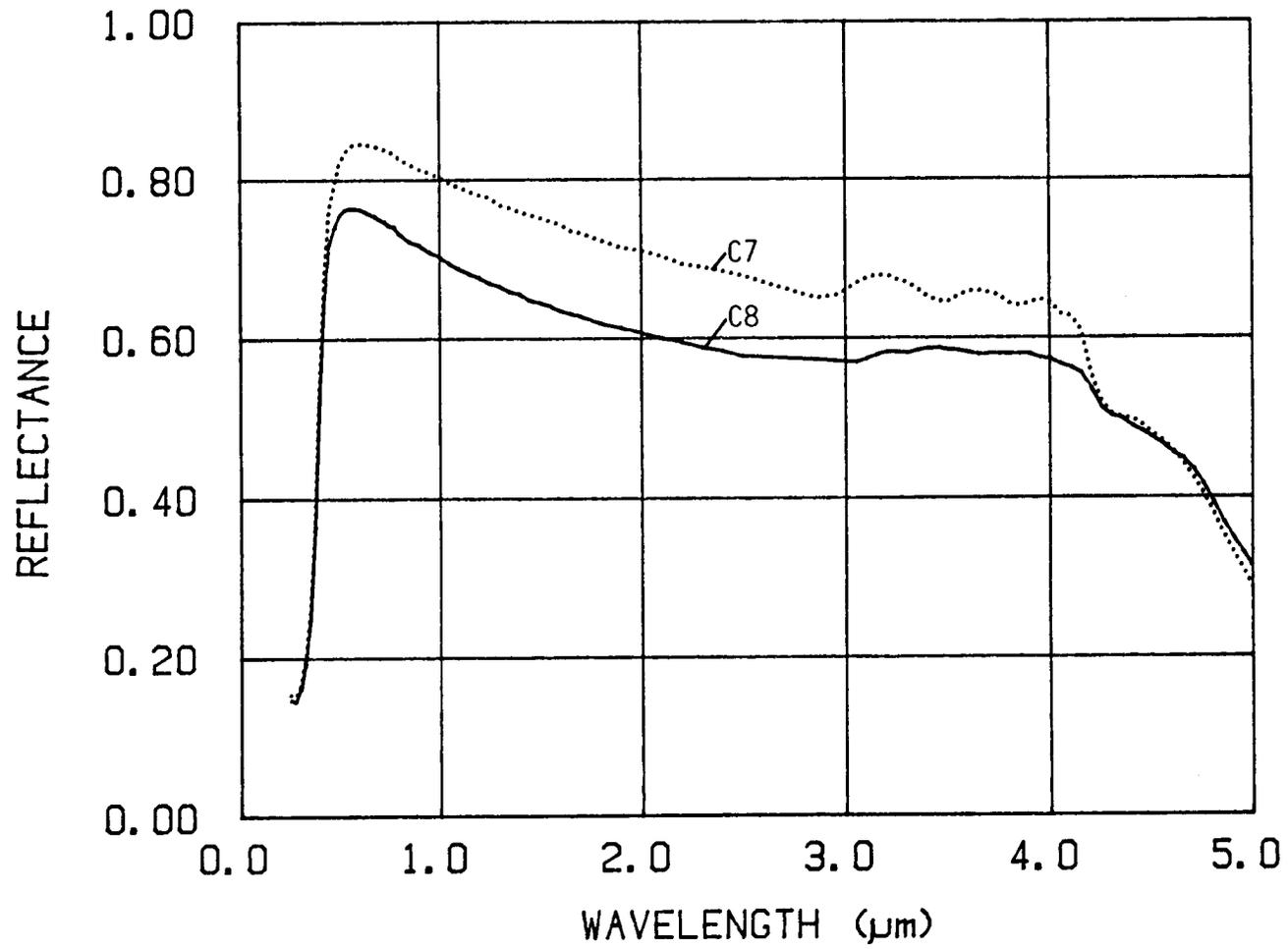


Figure 7(b). Merged reflectance spectra of material C used for parameter inversion.

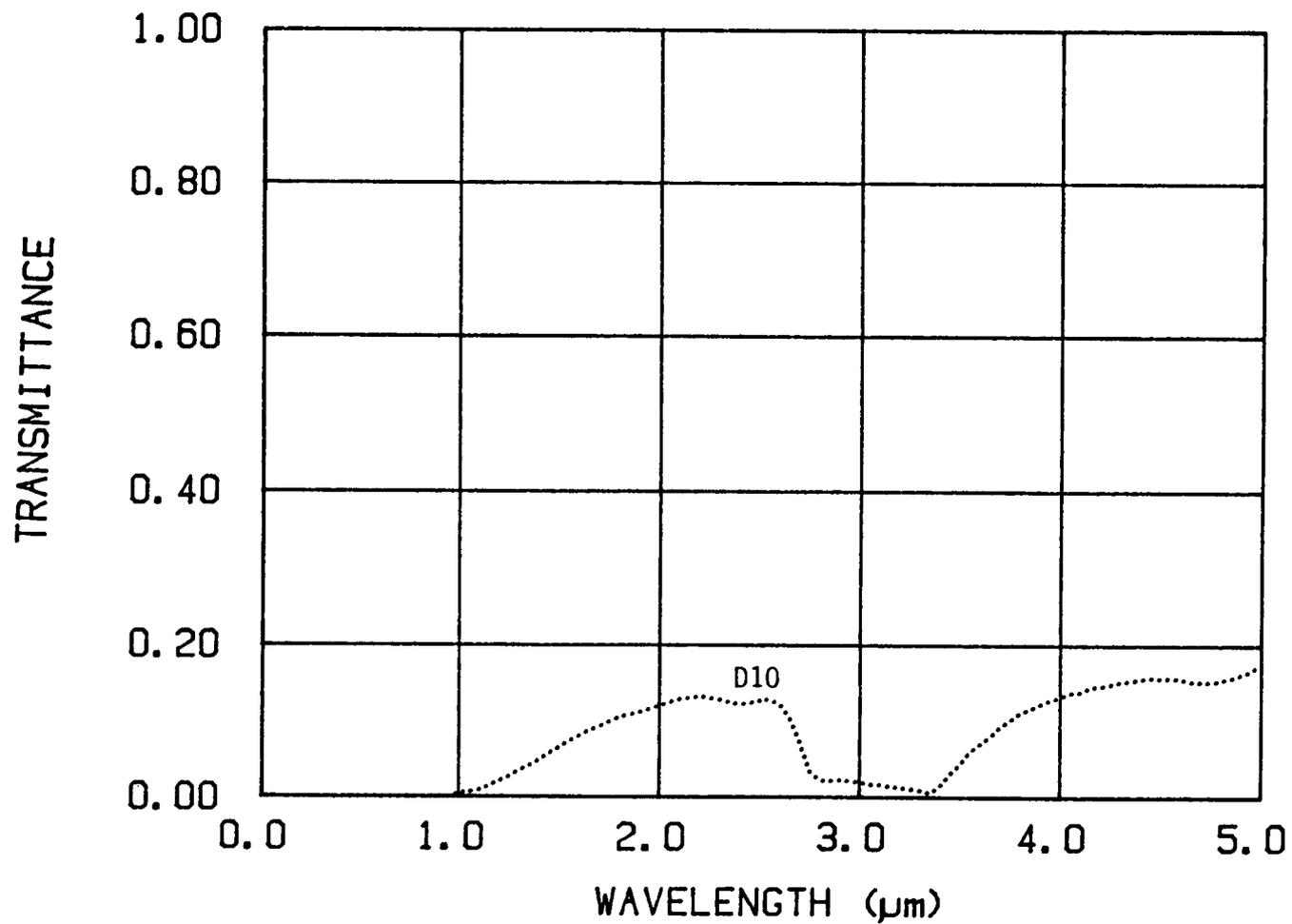


Figure 8(a). Inverted scattering coefficient spectrum of material B.

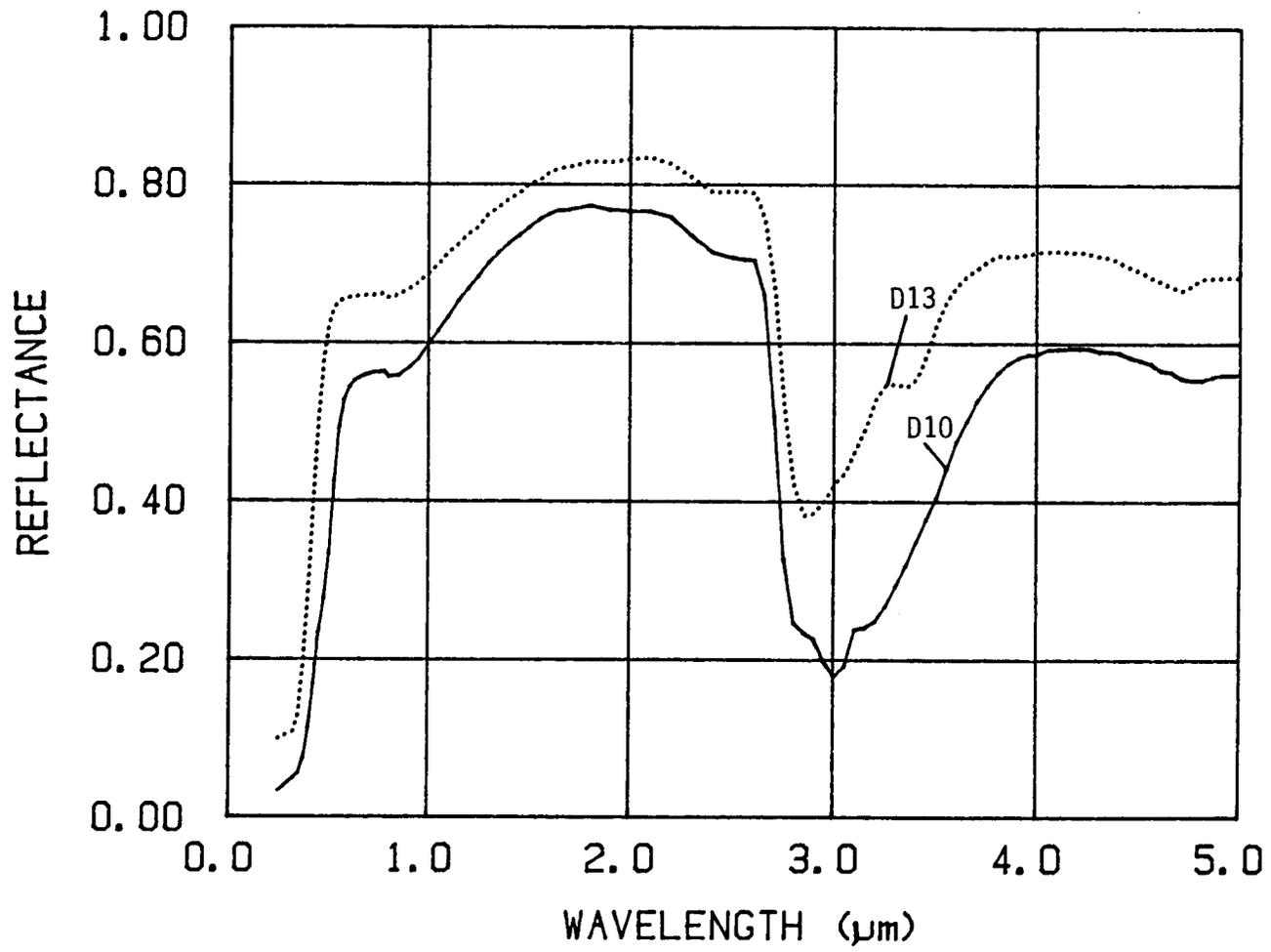


Figure 8(b). Inverted absorption coefficient spectrum of material B.

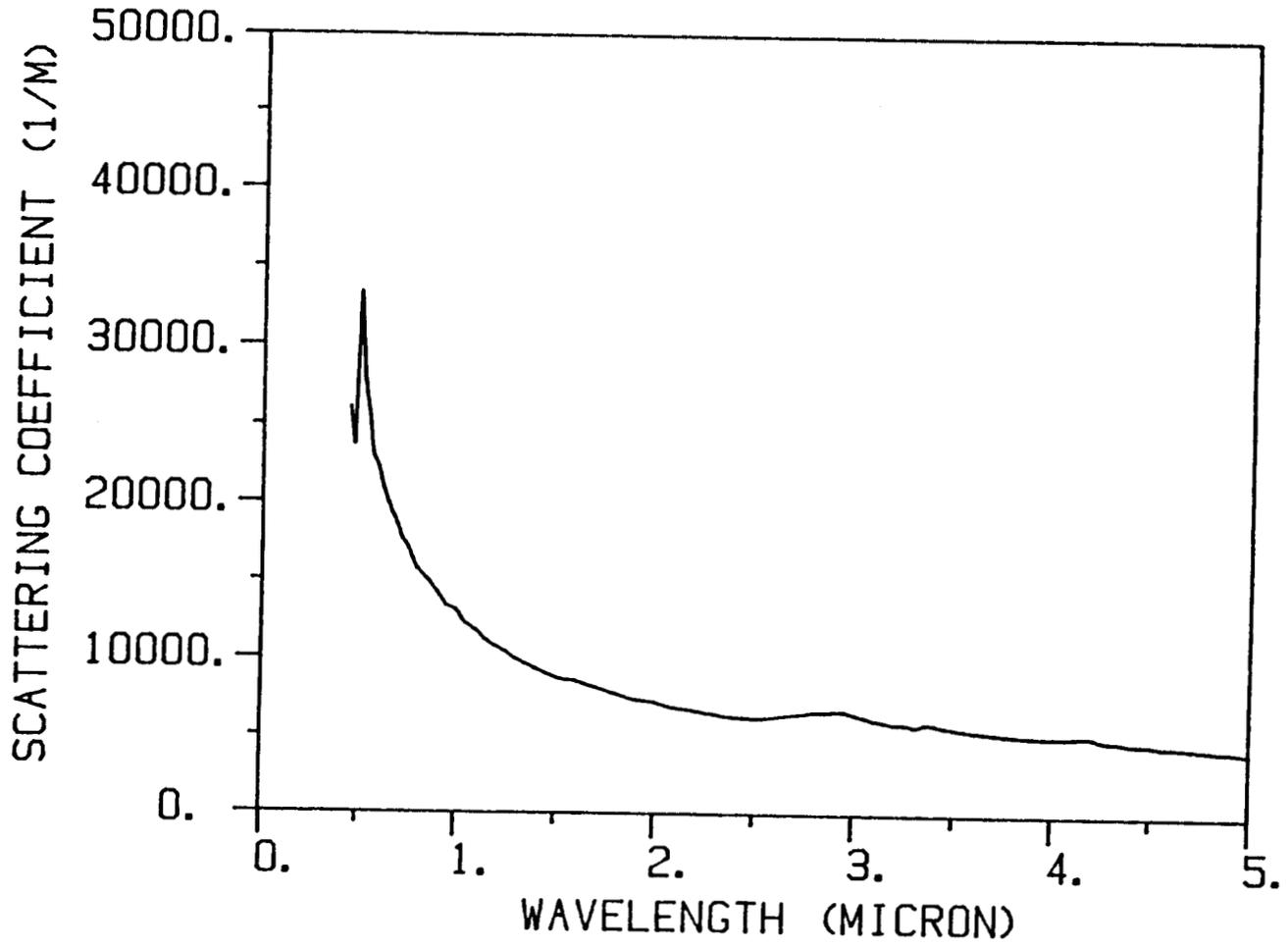


Figure 9(a). Inverted scattering coefficient spectrum of material B.

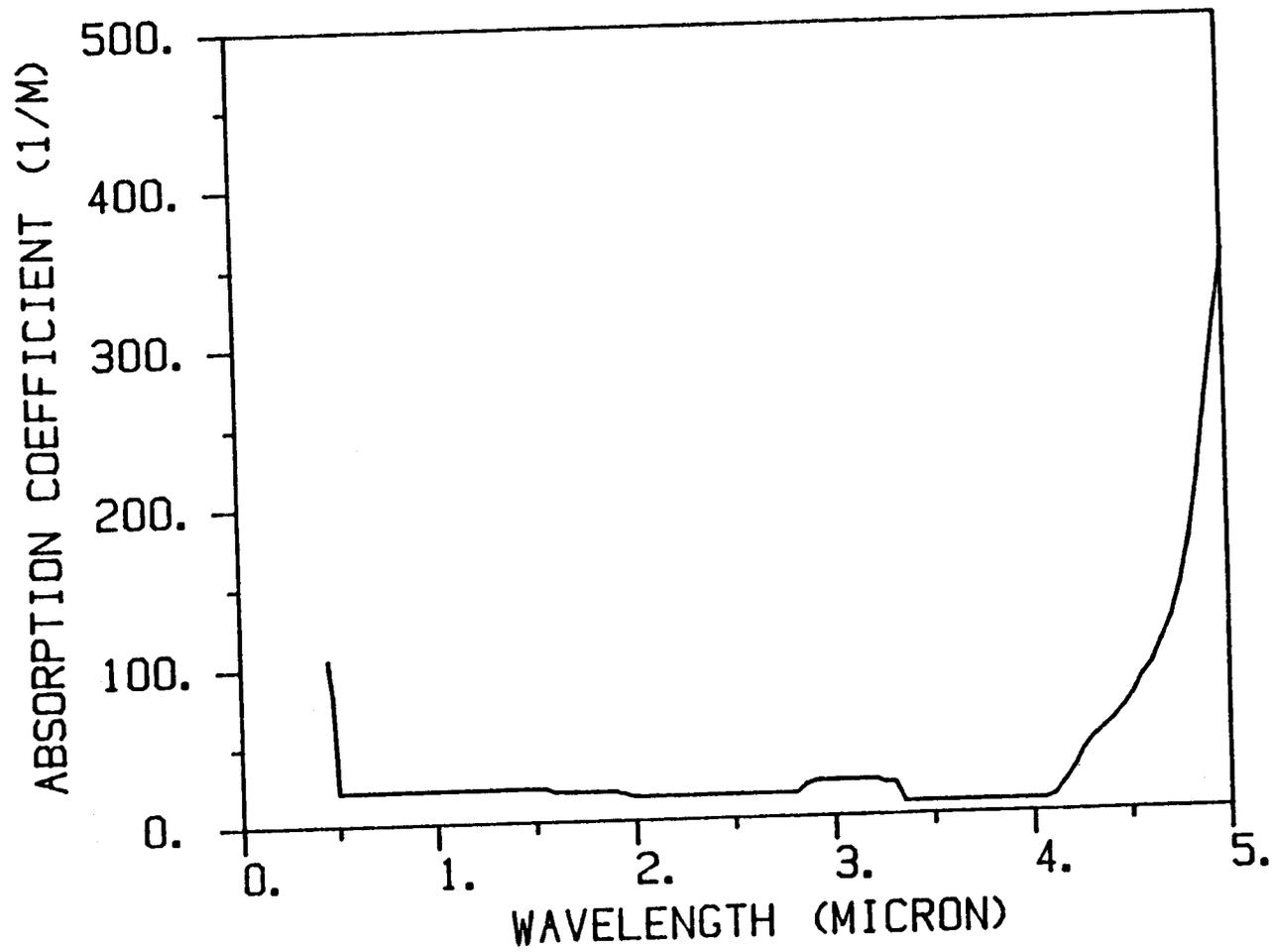


Figure 9(b). Inverted absorption coefficient spectrum of material B.

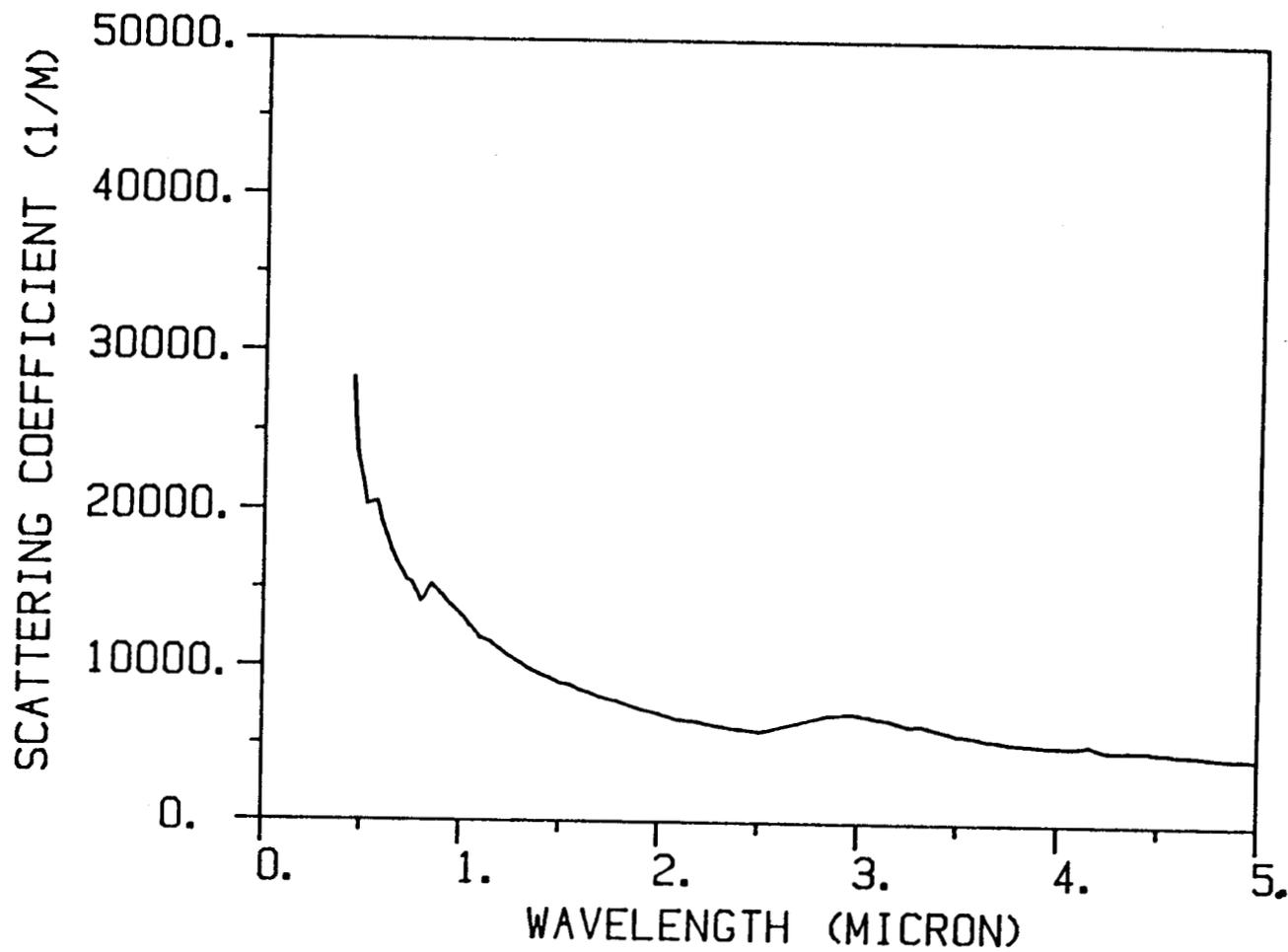


Figure 10 (a). Inverted scattering coefficient spectrum of material C.

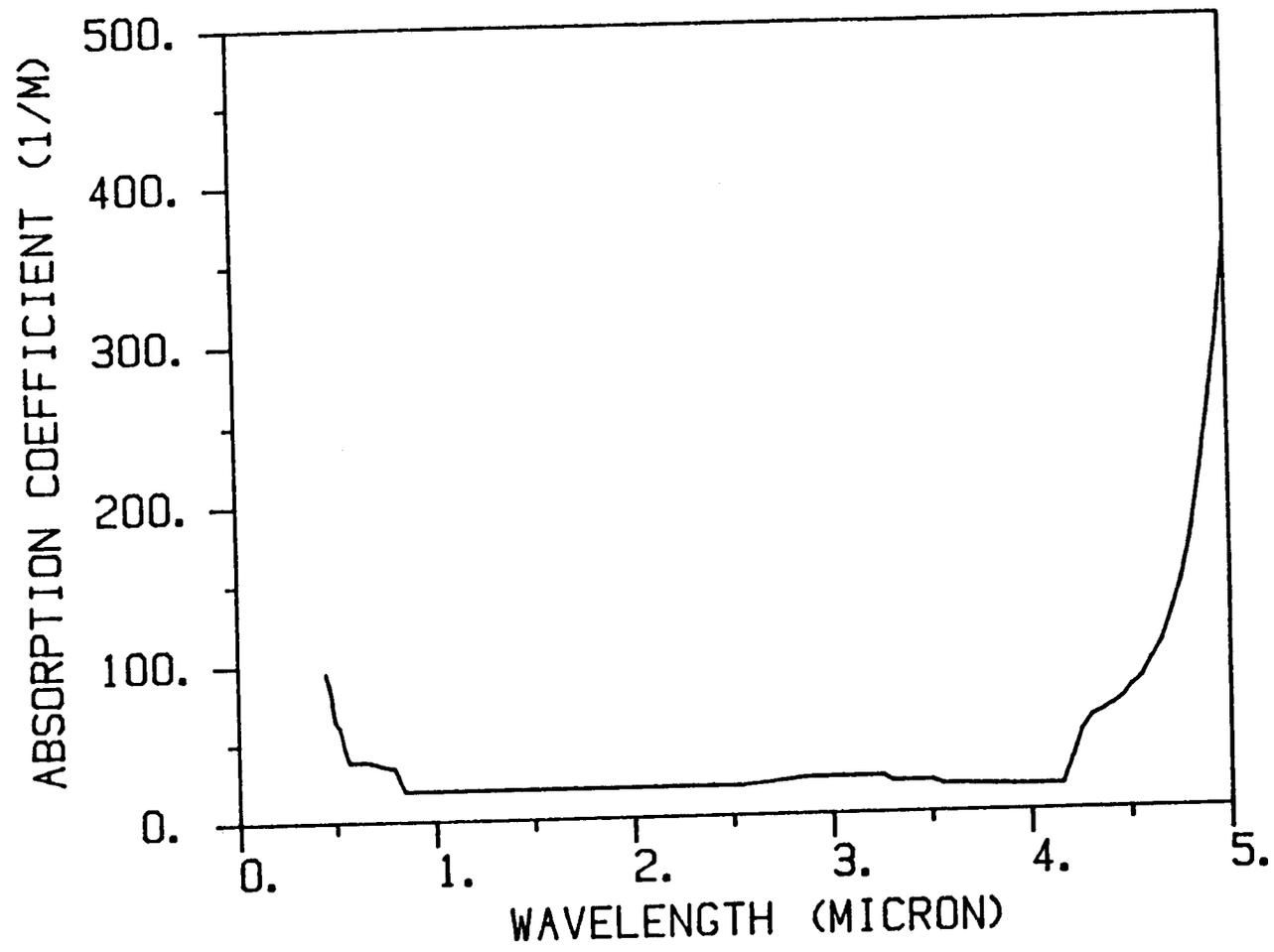


Figure 10 (b). Inverted absorption coefficient spectrum of material C.

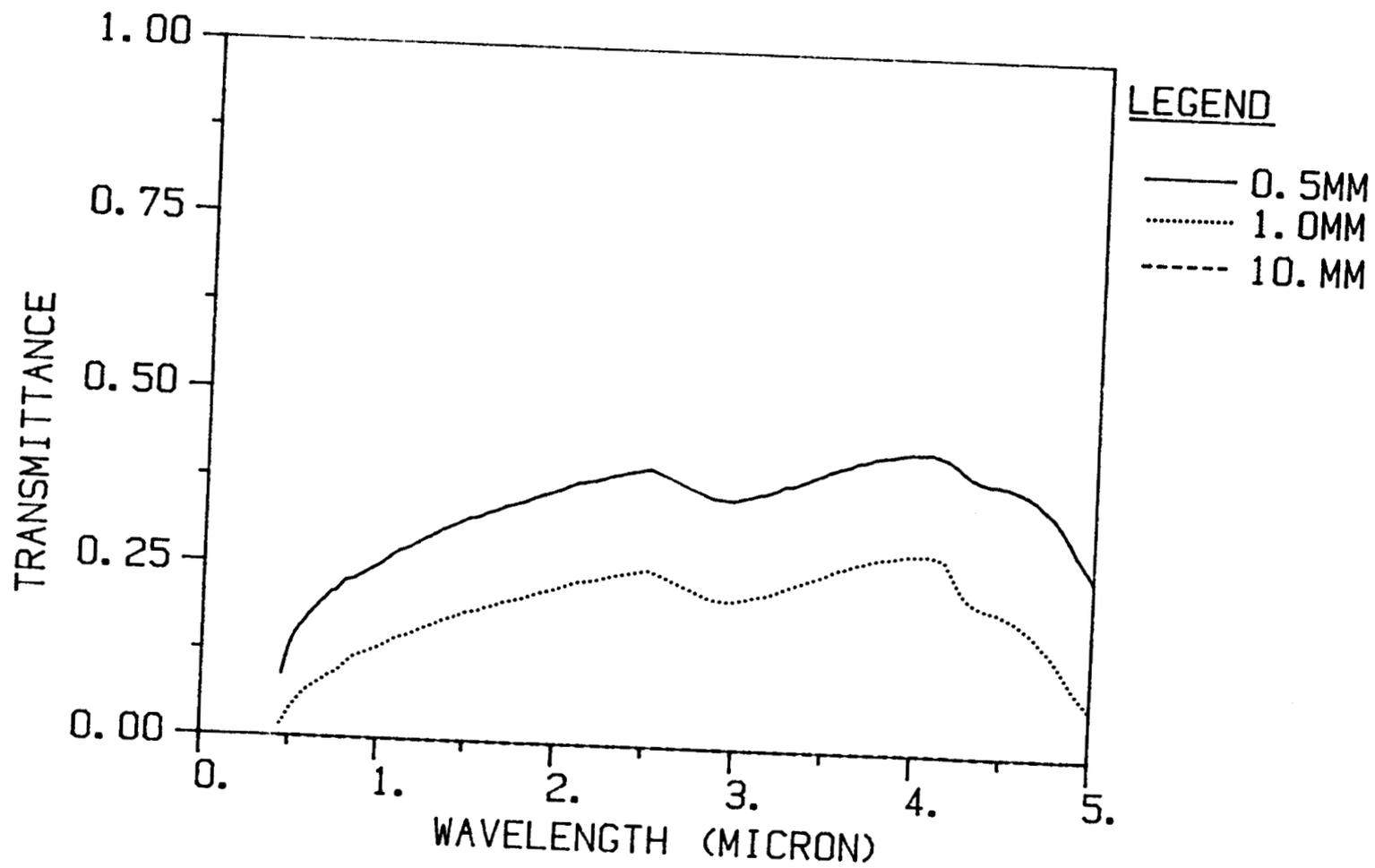


Figure 11(a). Calculated transmittance spectra of material C.

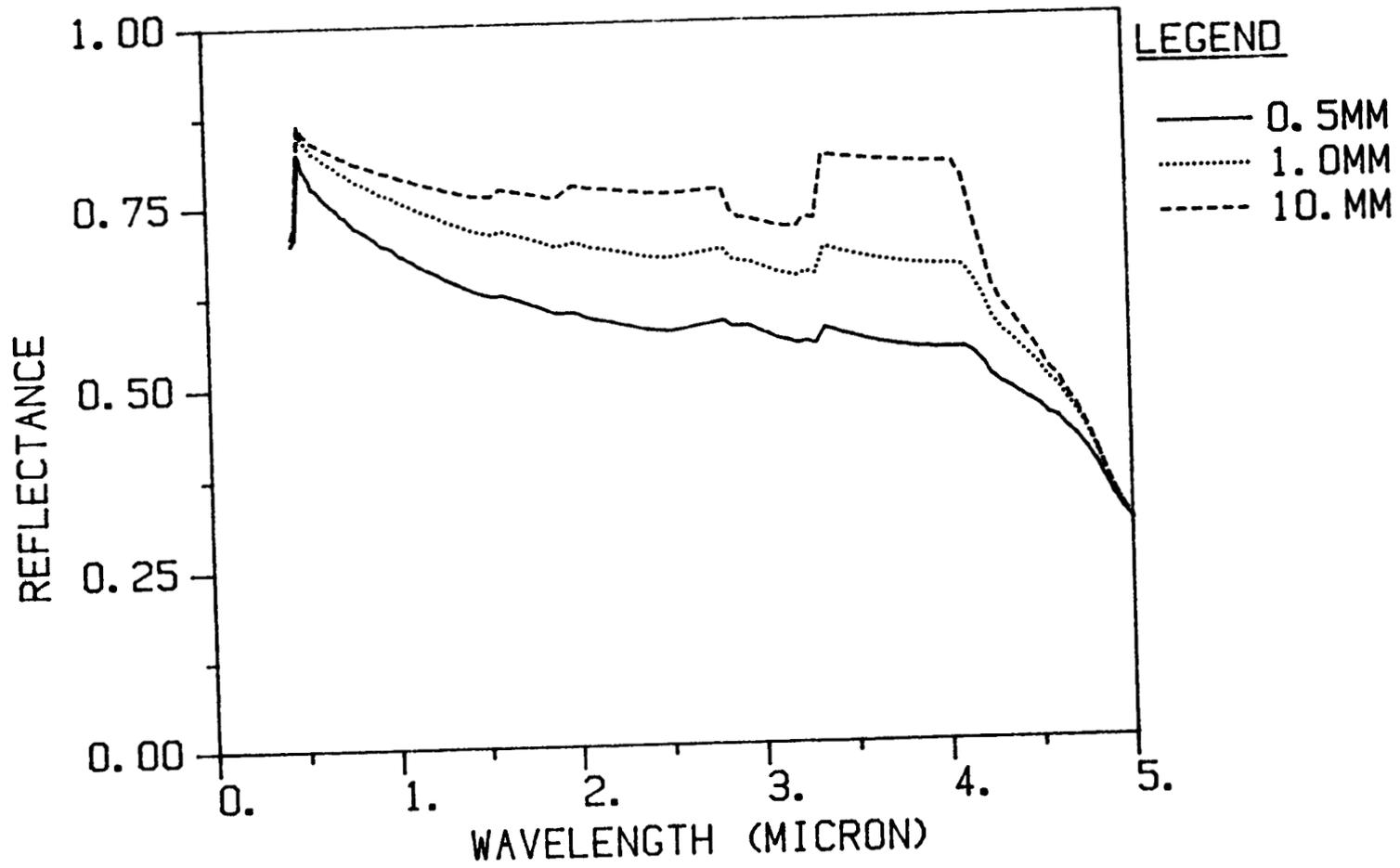


Figure 11(b). Calculated reflectance spectra of material C.

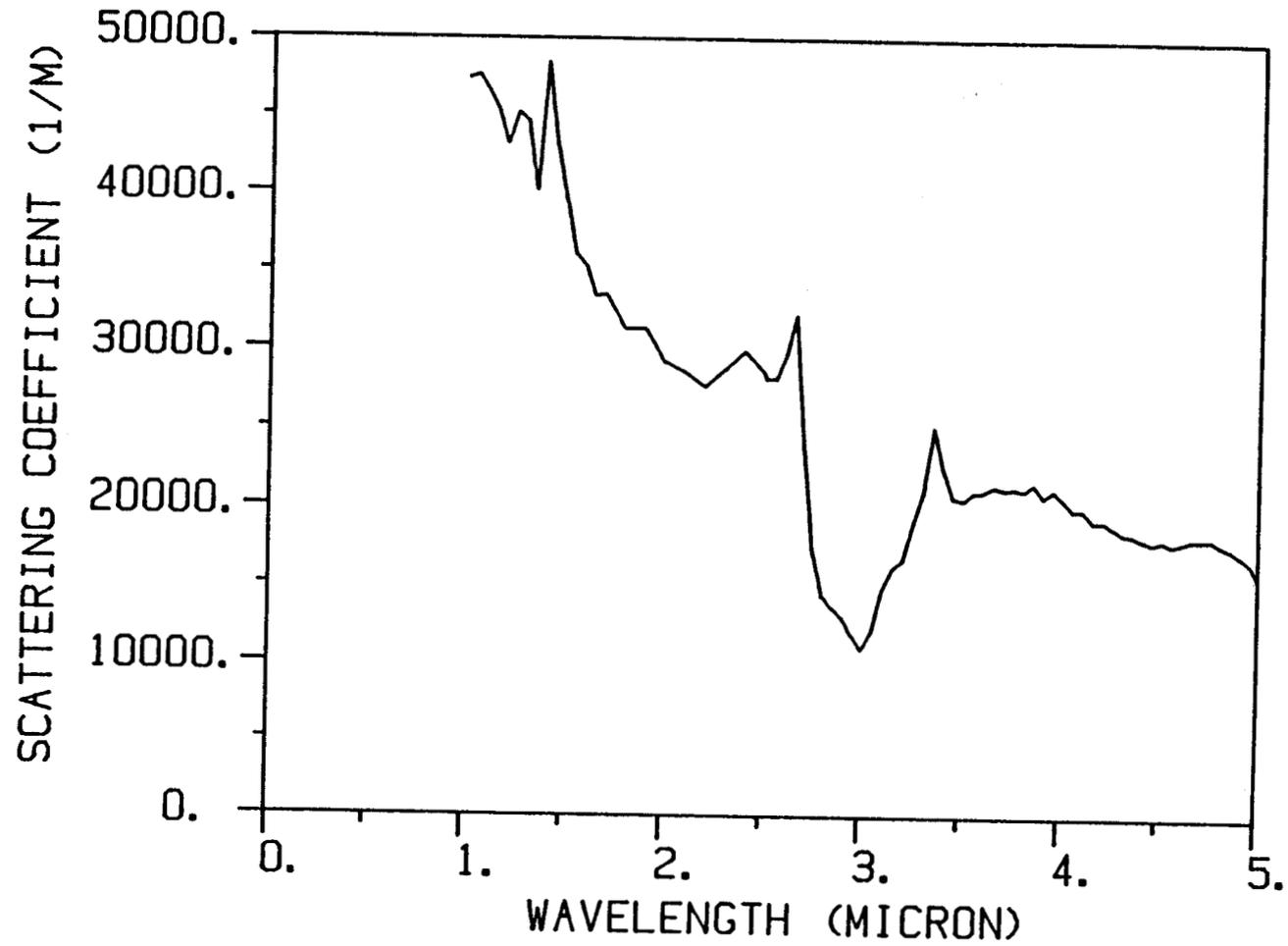


Figure 12(a). Inverted scattering coefficient spectrum of material D.

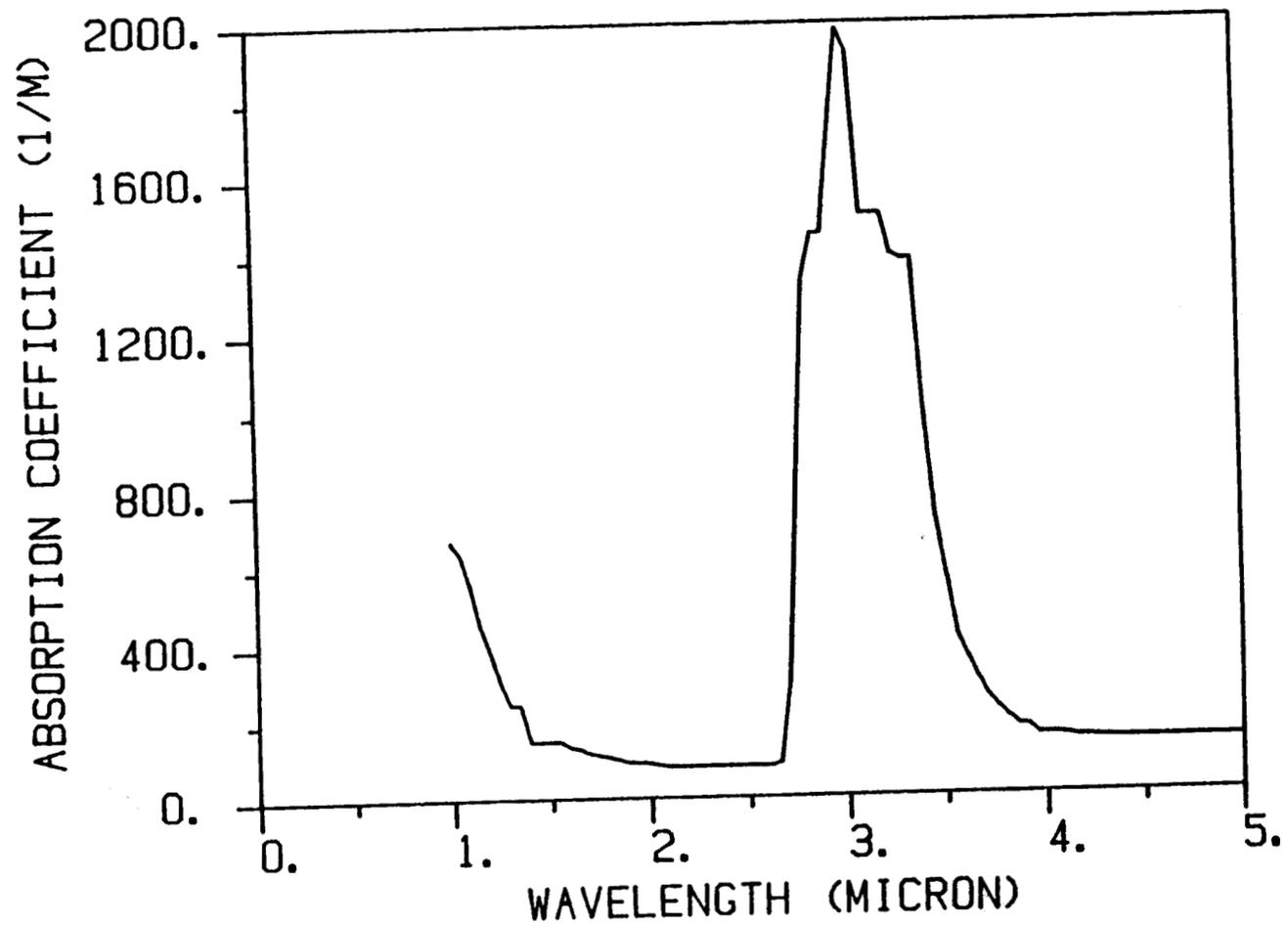


Figure 12(b). Inverted absorption coefficient spectrum of material D.

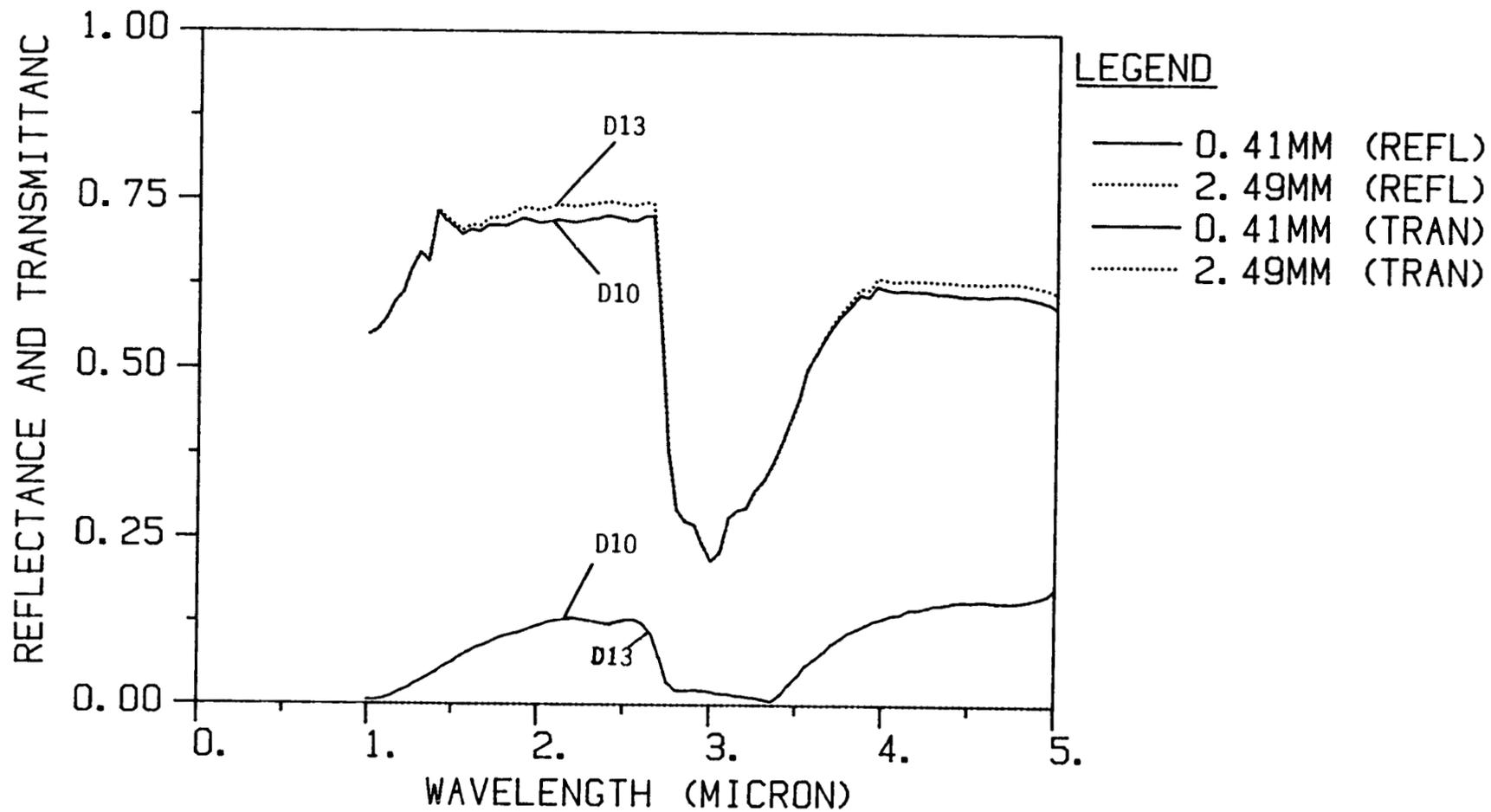


Figure 13(a). Calculated transmittance and reflectance spectra of material D.

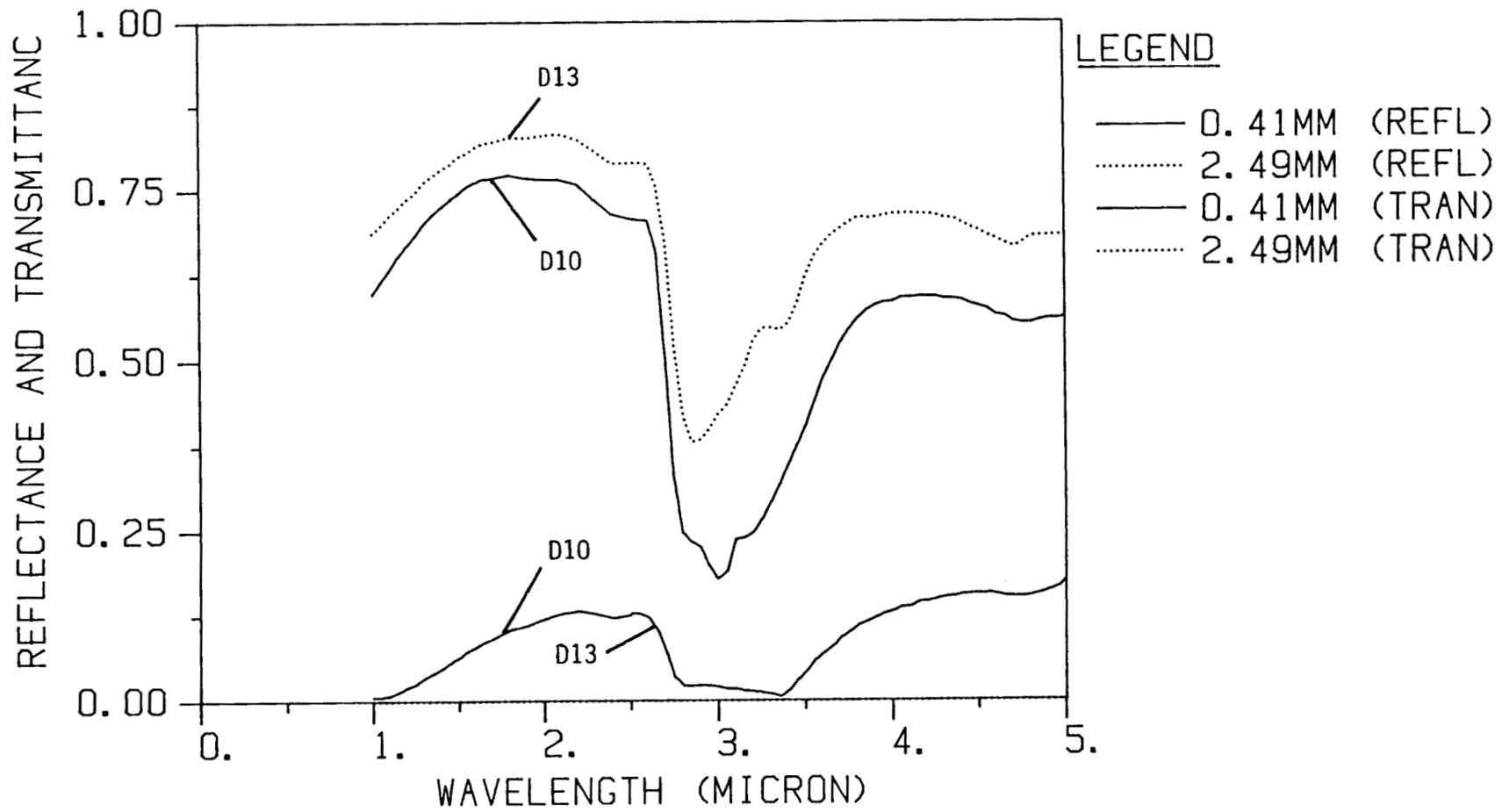


Figure 13(b). Measured transmittance and reflectance spectra of material D.

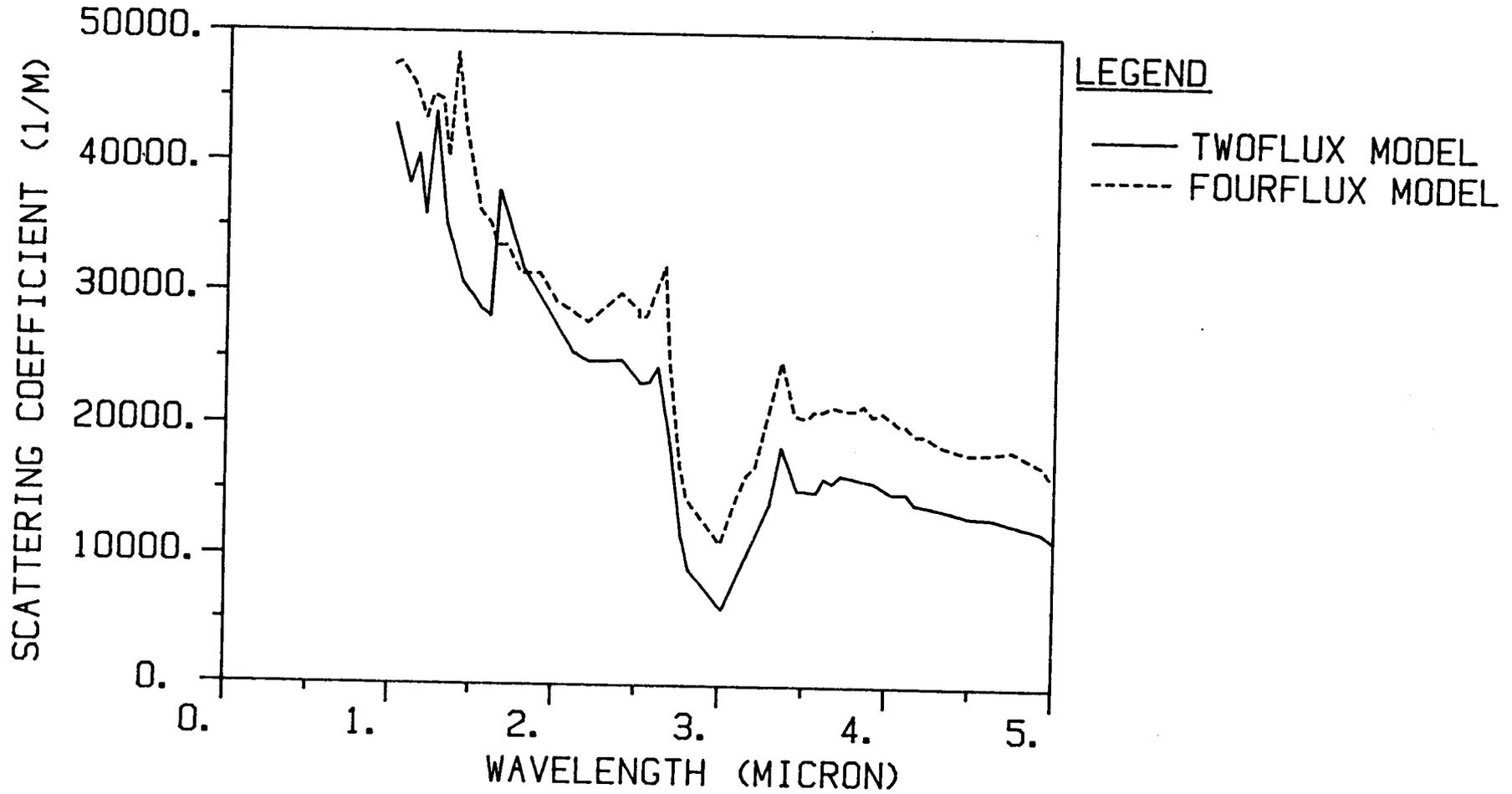


Figure 14(a). Comparison of inverted scattering coefficient spectrum of material D for four flux and two flux models.

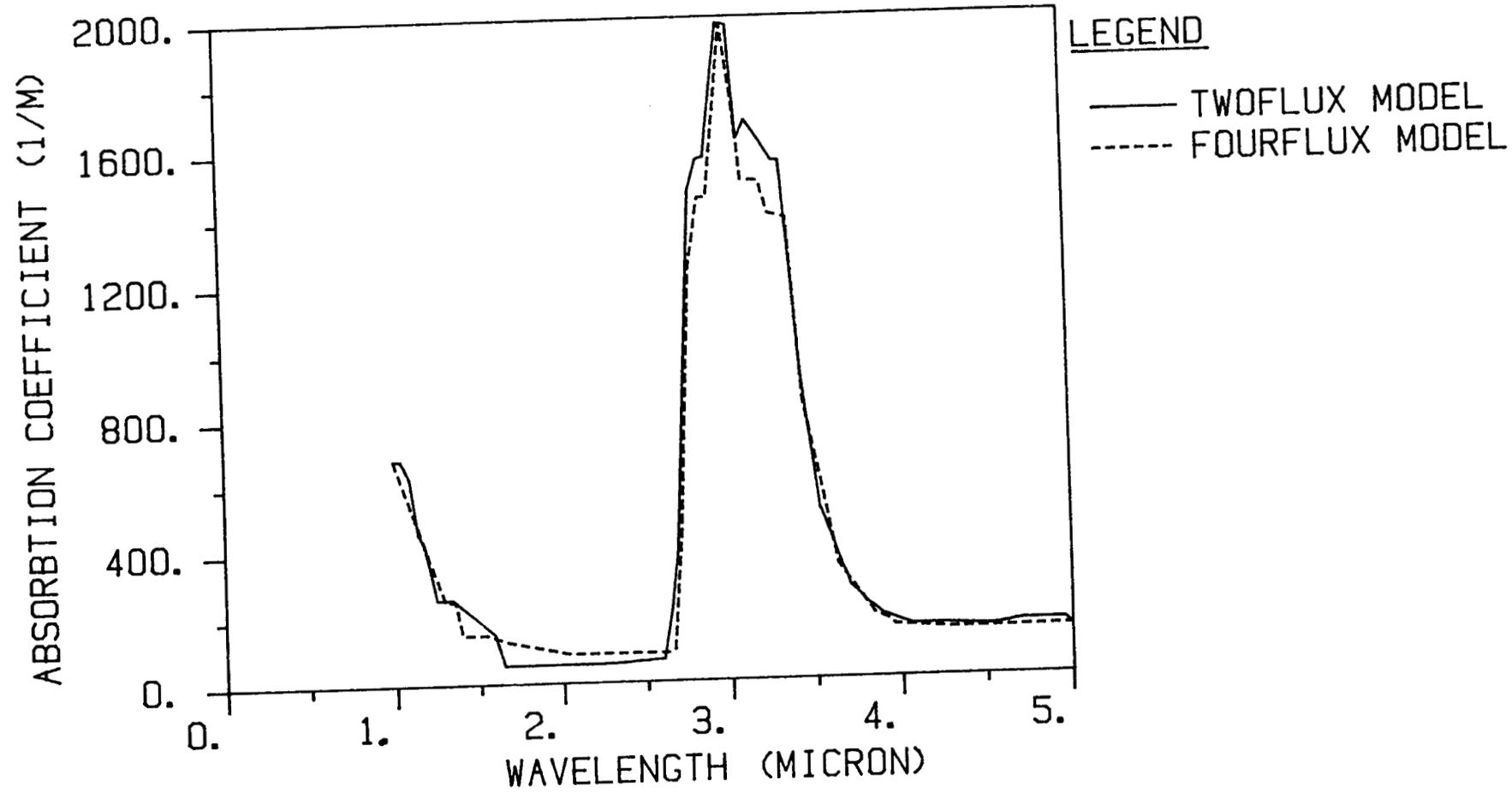


Figure 14(b). Comparison of inverted absorption coefficient spectrum of material D for four flux and two flux models.

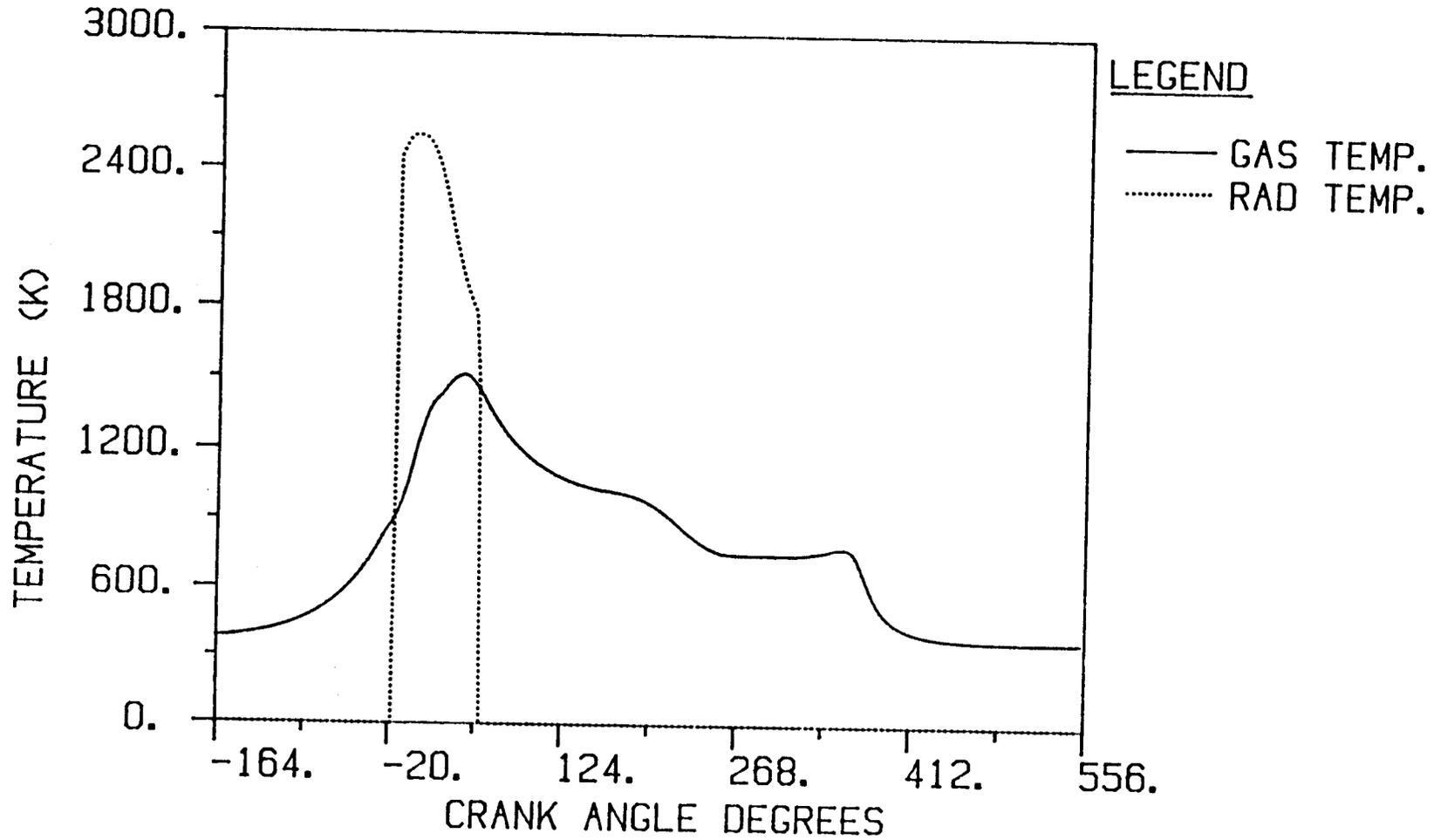


Figure 15(a). Gas and radiation temperature history used as gas side boundary condition on the heat barrier coating.

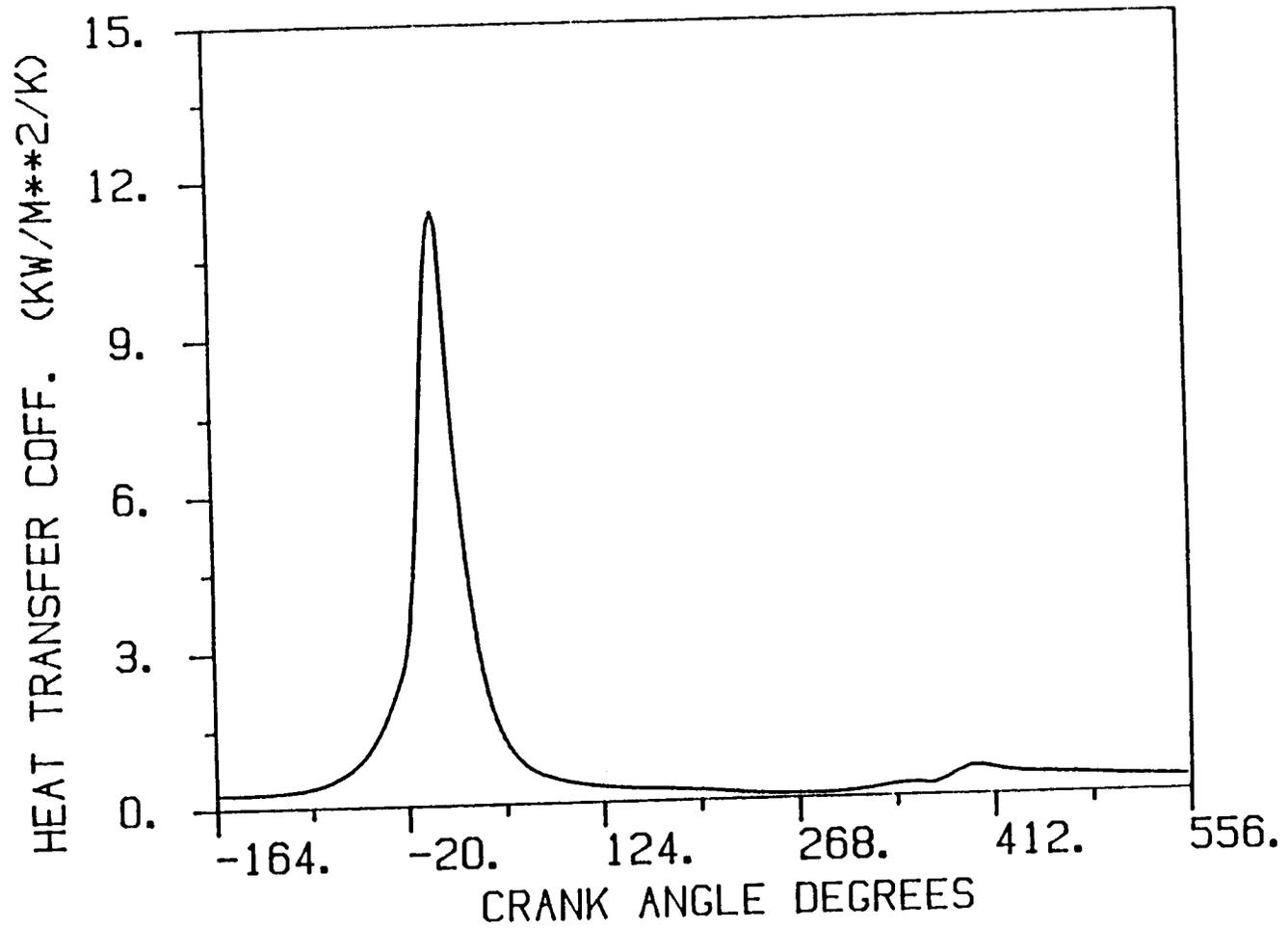


Figure 15(b). Heat transfer coefficient history used as gas side boundary condition on the heat barrier coating.

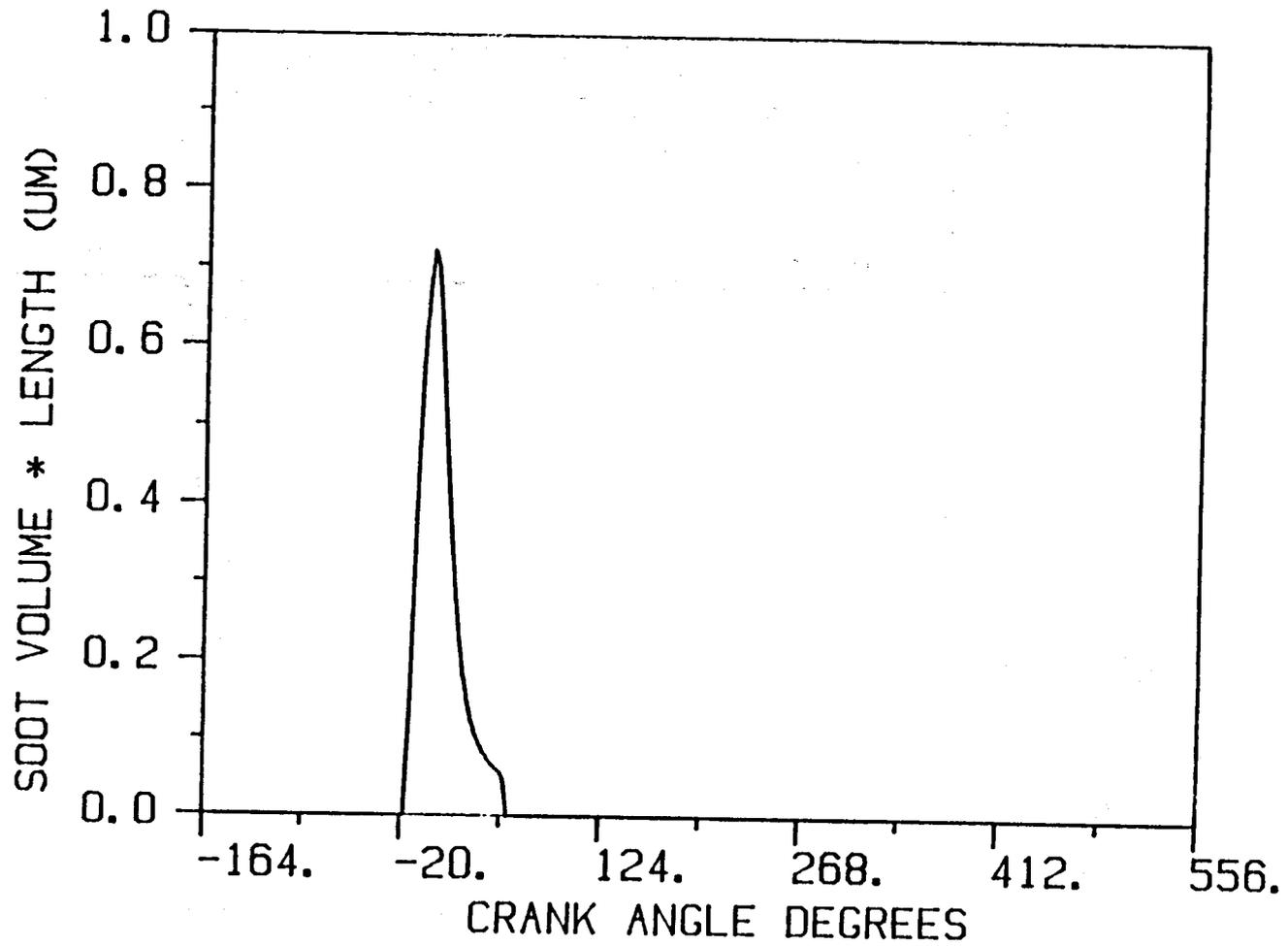


Figure 15(c). History of soot volume fraction times radiation path length used as gas side boundary condition on the heat barrier coating.

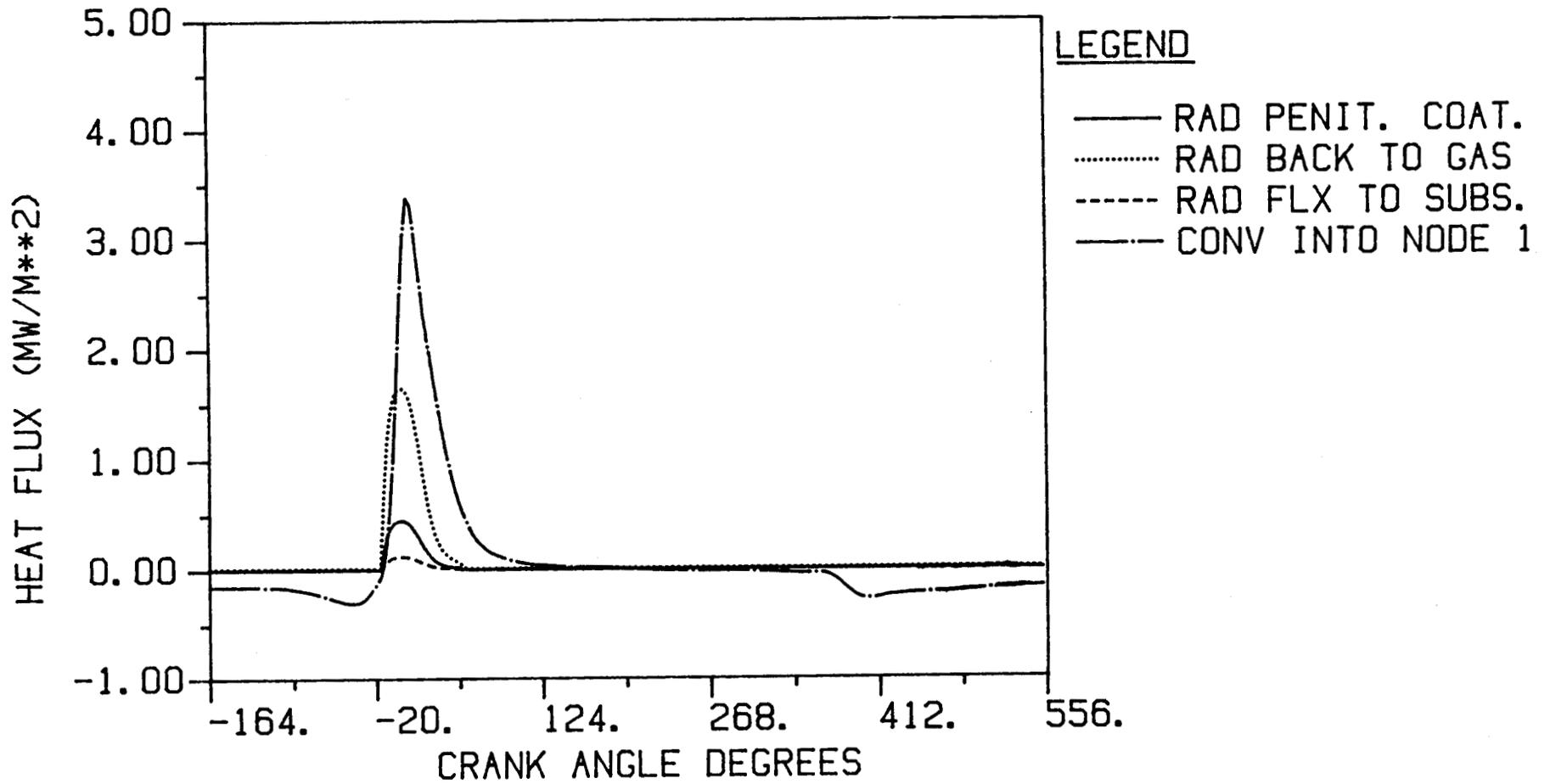


Figure 16(a). Time dependent fluxes computed for 2.5 mm heat barrier coating using radiative properties of material C.

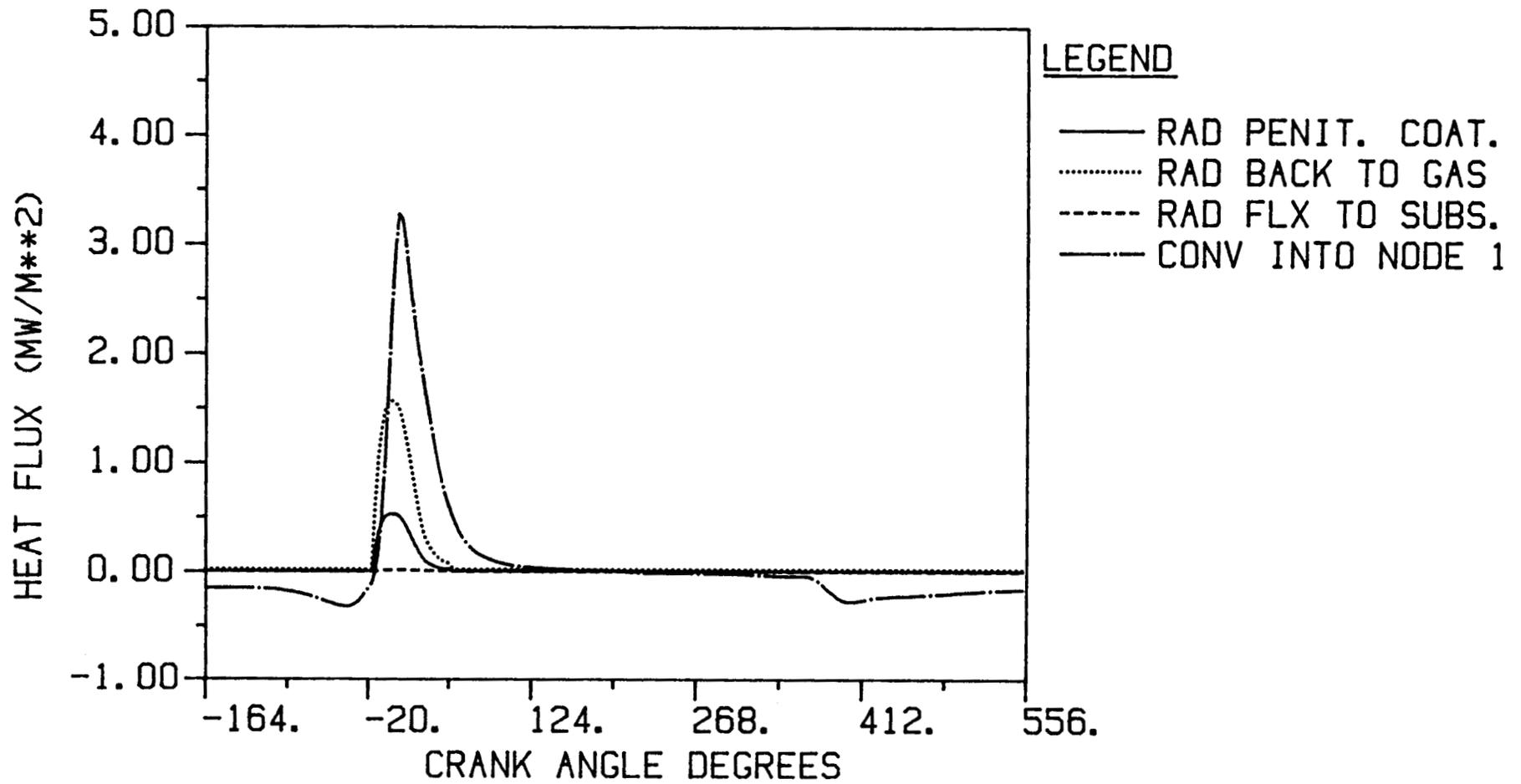


Figure 16(b). Time dependent fluxes computed for 2.5 mm heat barrier coating using radiative properties of material D.

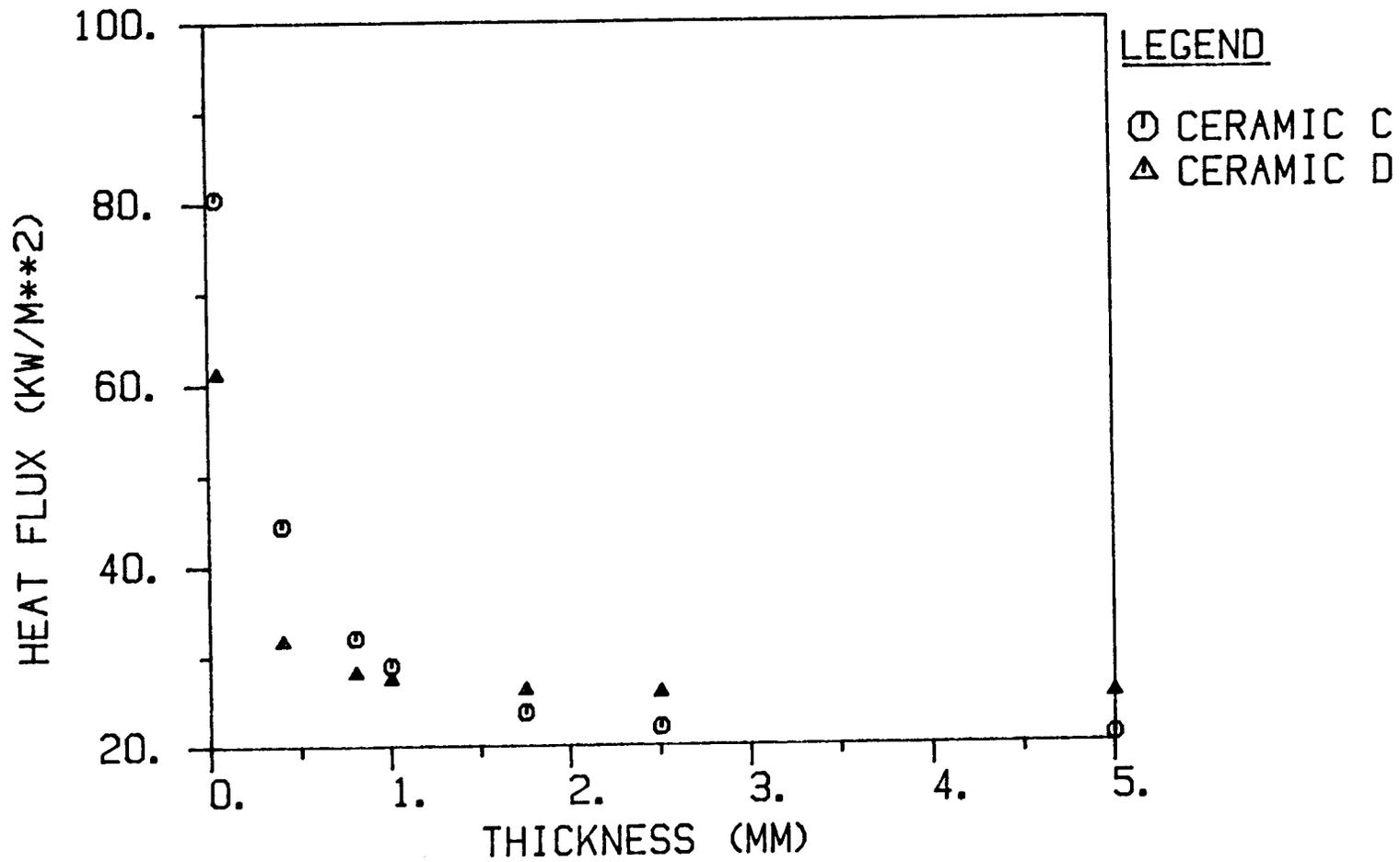


Figure 17(a). Time averaged radiative heat flux for ceramics C and D as a function of thickness.

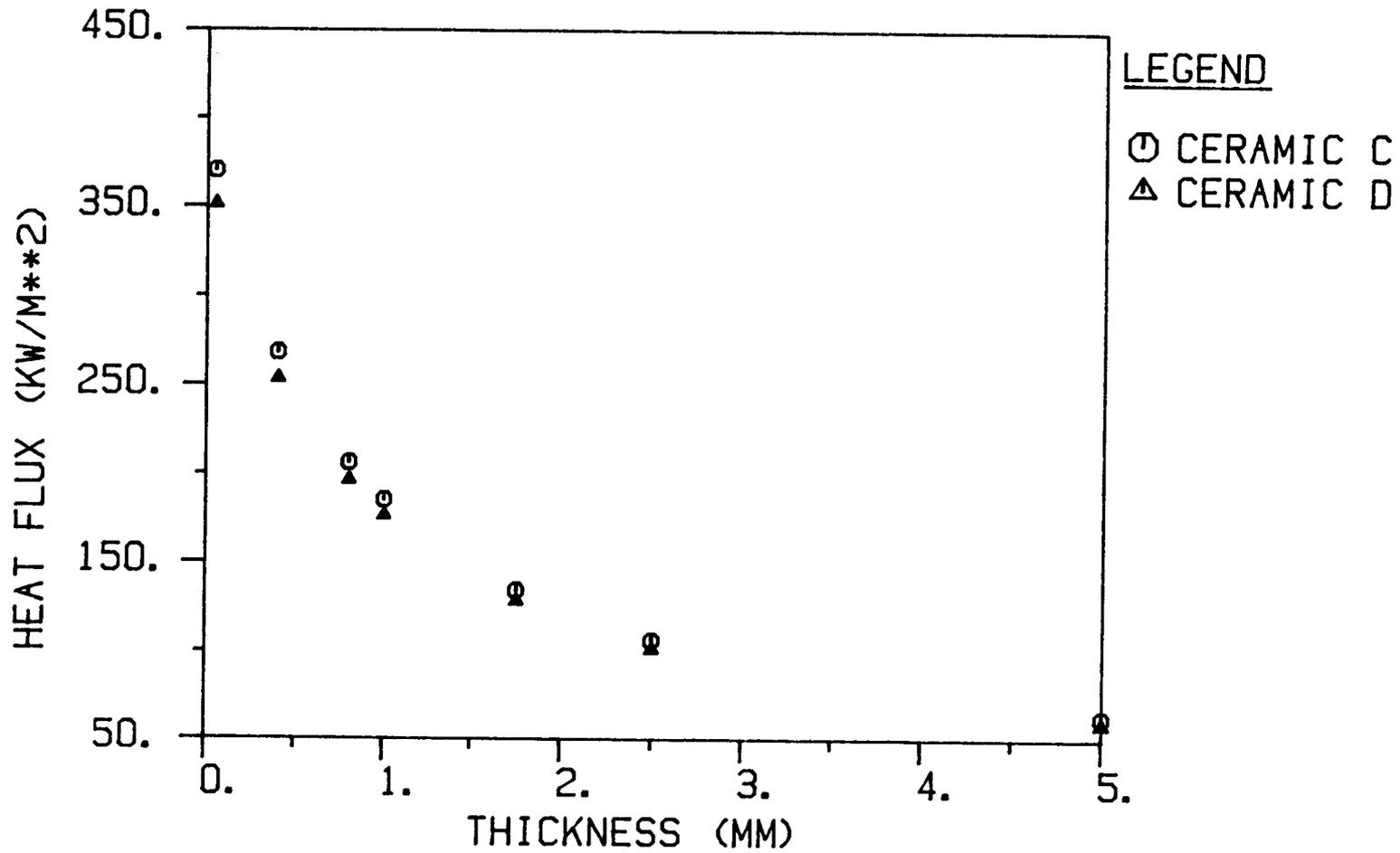


Figure 17(b). Time averaged total heat flux for ceramics C and D as a function of thickness.

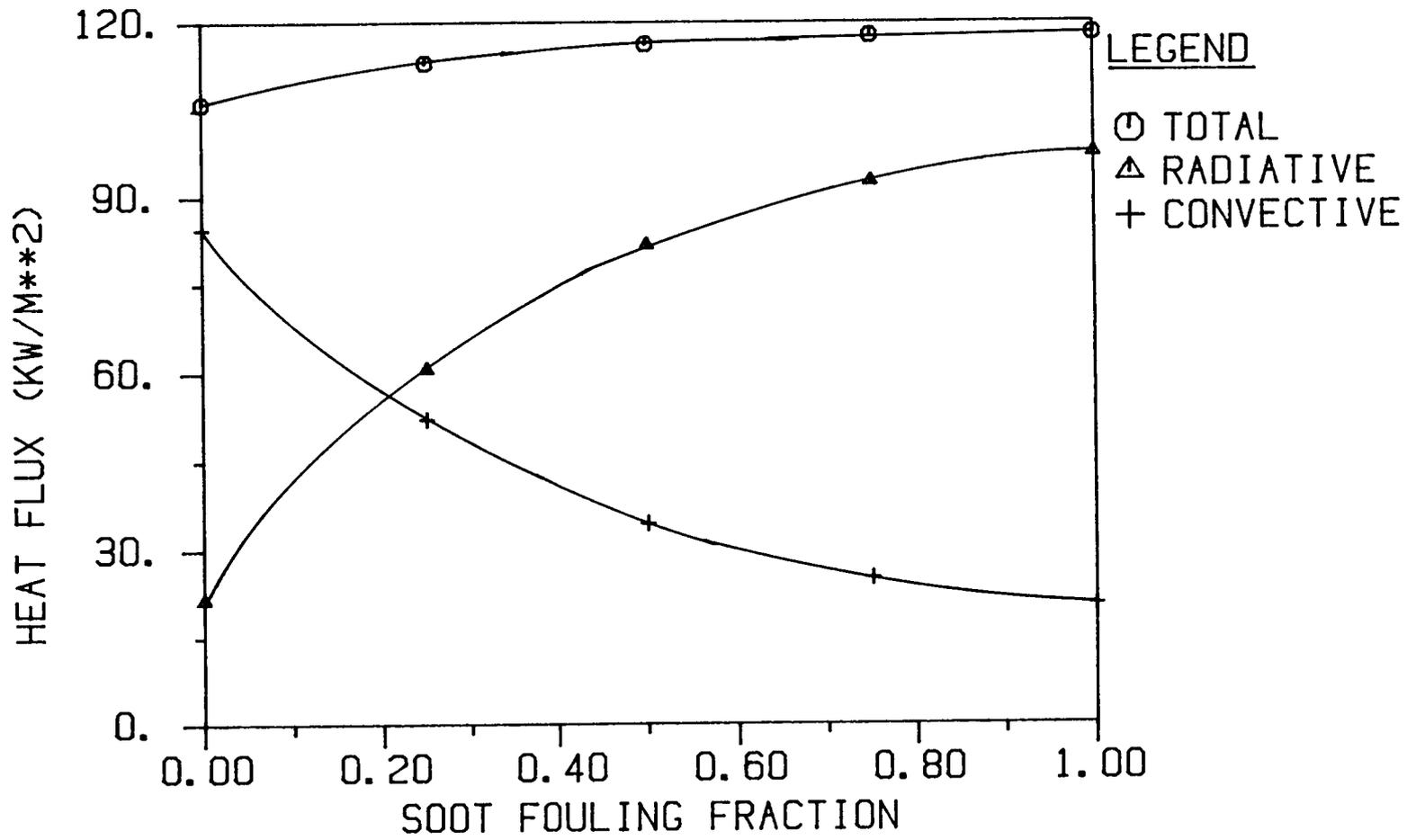


Figure 18. Effect of soot fouling for 2.5 mm Ceramic C coating used as heat barrier on time averaged fluxes.

## APPENDIX

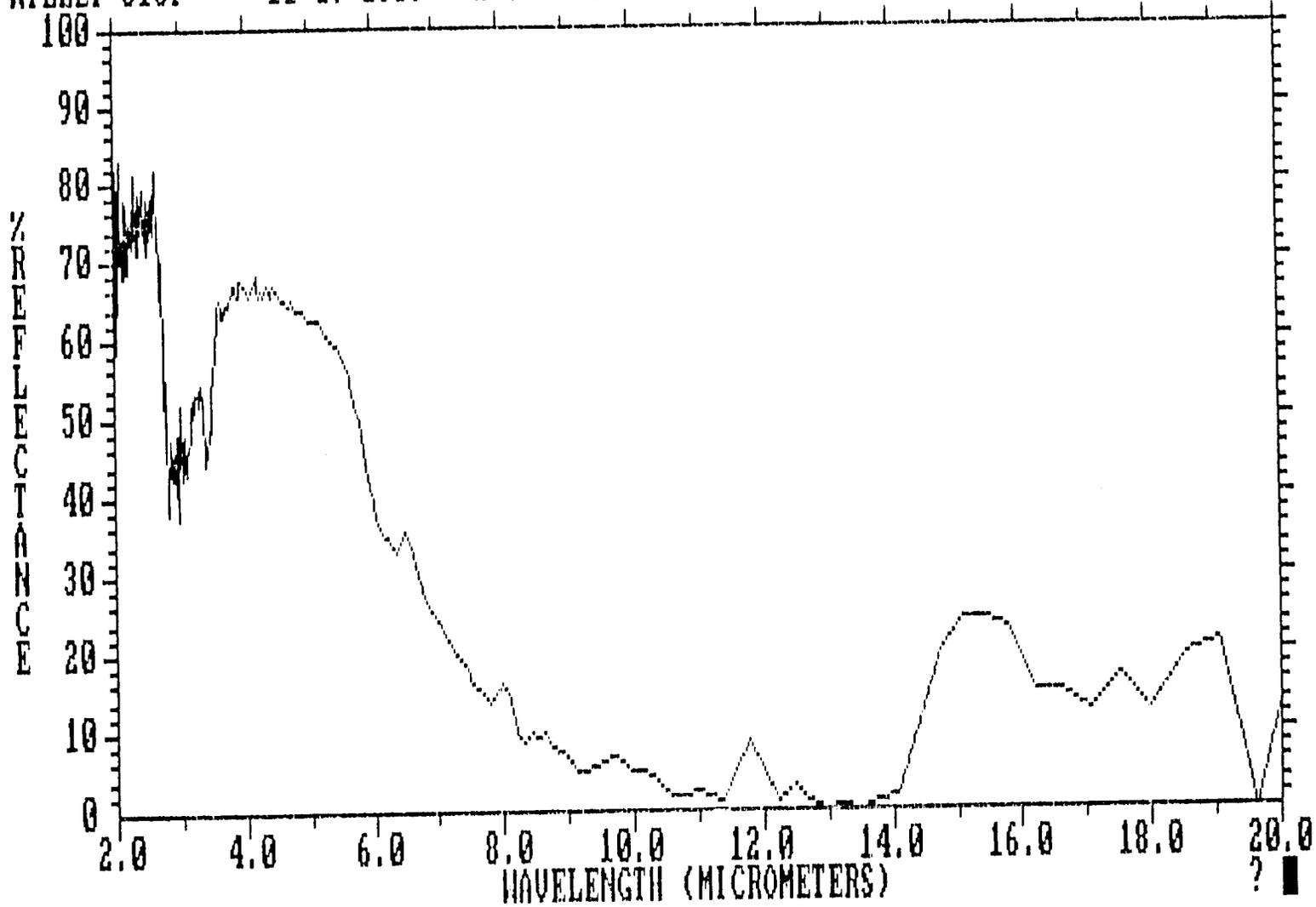
The reflectance and transmittance measurements at short to long infrared regions ( $2\ \mu\text{m}$  to  $20\ \mu\text{m}$ ) were measured at the Willey Corporation. The data obtained were quite noisy at shorter wavelengths and necessitated smoothing. The smoothed profiles shown in the text have been used in the analysis. However, for reference purposes, the unsmoothed data is also presented in the following figures. (Note: Per instruction from the Willey Corporation the values in the reflectance spectra need to be divided by 0.95). The corresponding NBS measurements carried out in the range  $0.25\ \mu\text{m}$  to  $2.5\ \mu\text{m}$  were furnished in tabular form and plots obtained from these tables are presented without modification in the general text of this report and will not be repeated here.

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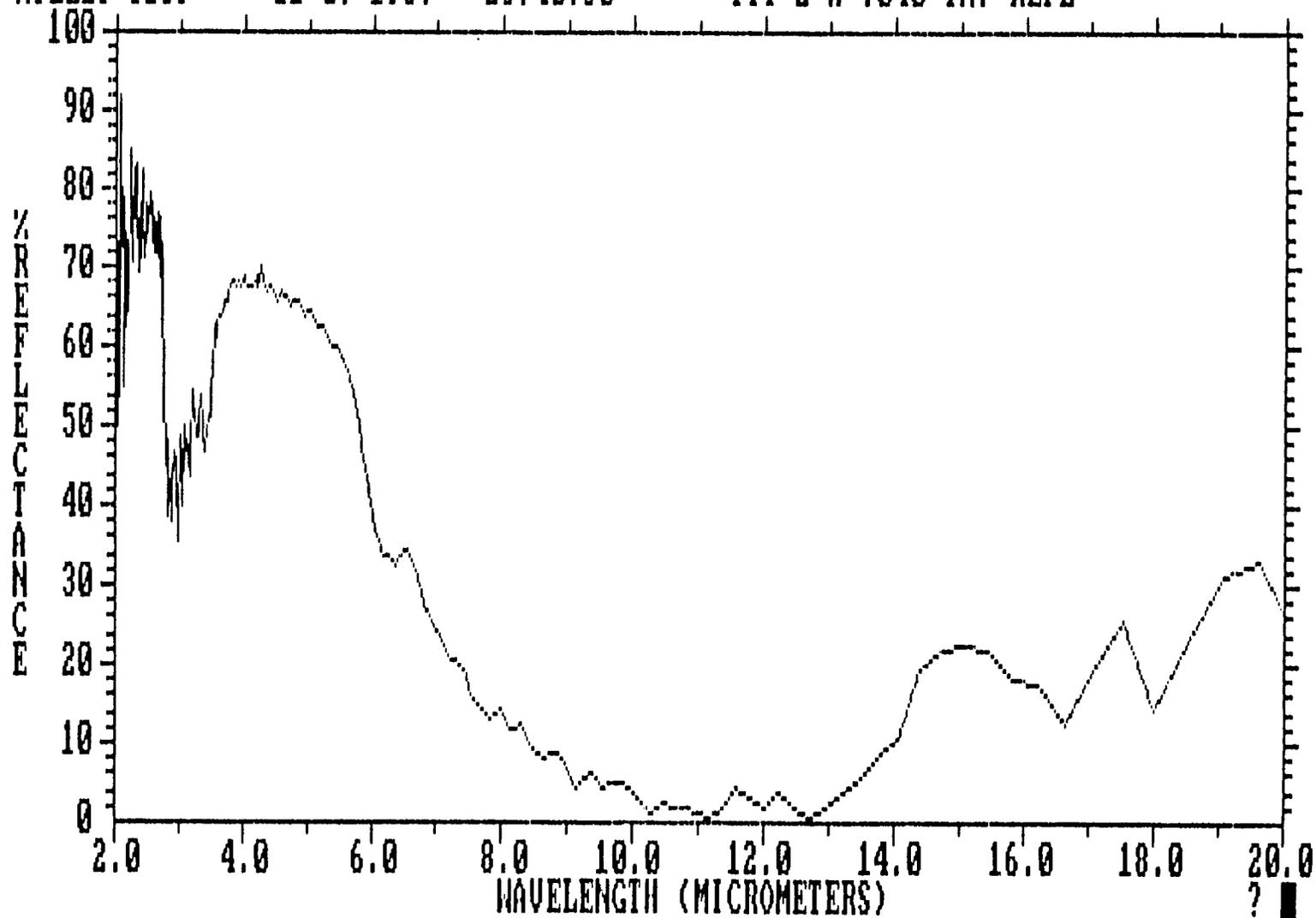
Figure A-1 Unsmoothed Reflectance Spectra of Sample A1

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Figure A-2 Unsmoothed Reflectance Spectra of Sample A2

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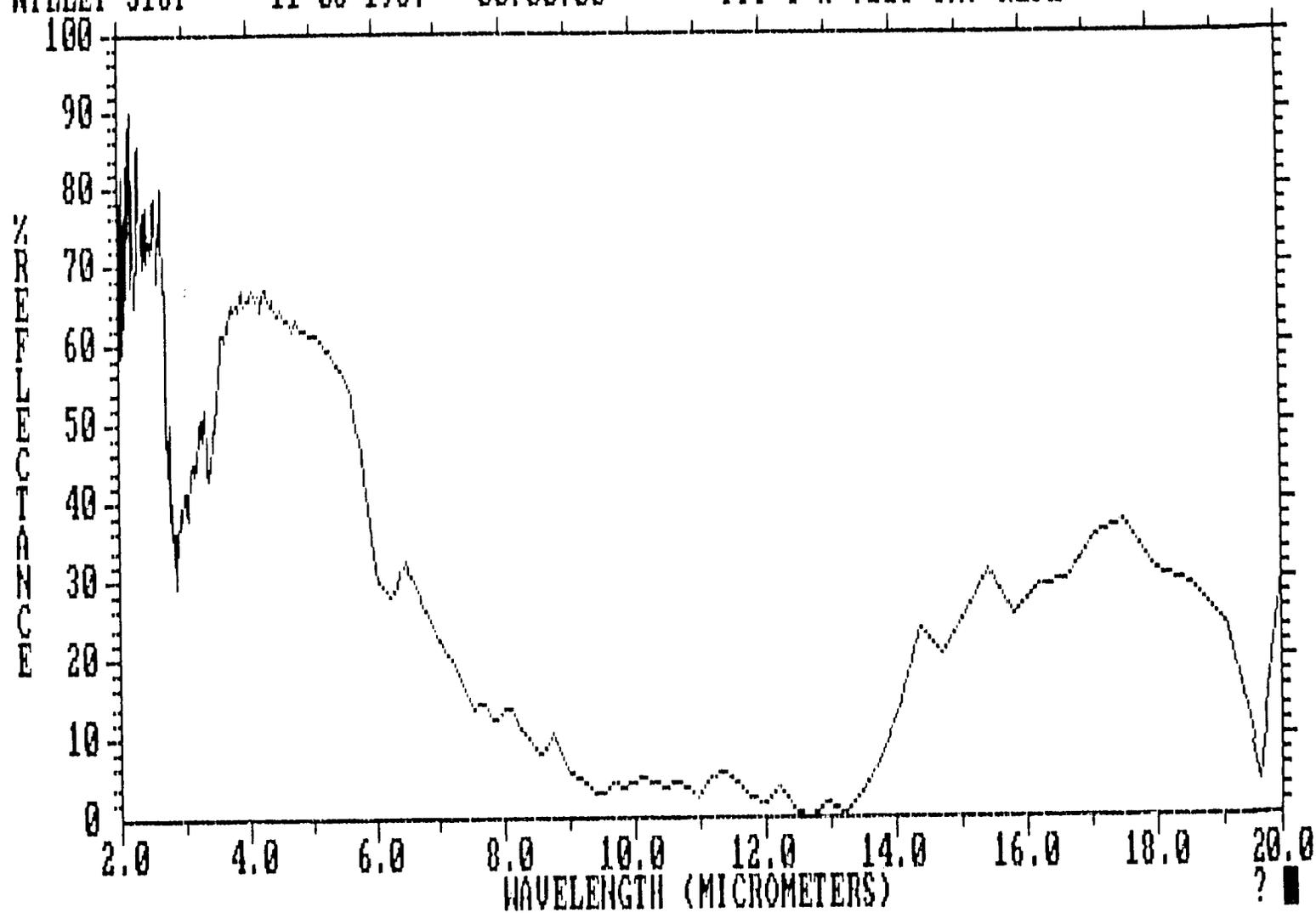


Figure A-3 Unsmoothed Reflectance Spectra of Sample A3

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18:54:39

ITI 4 B 0.5MM REFL

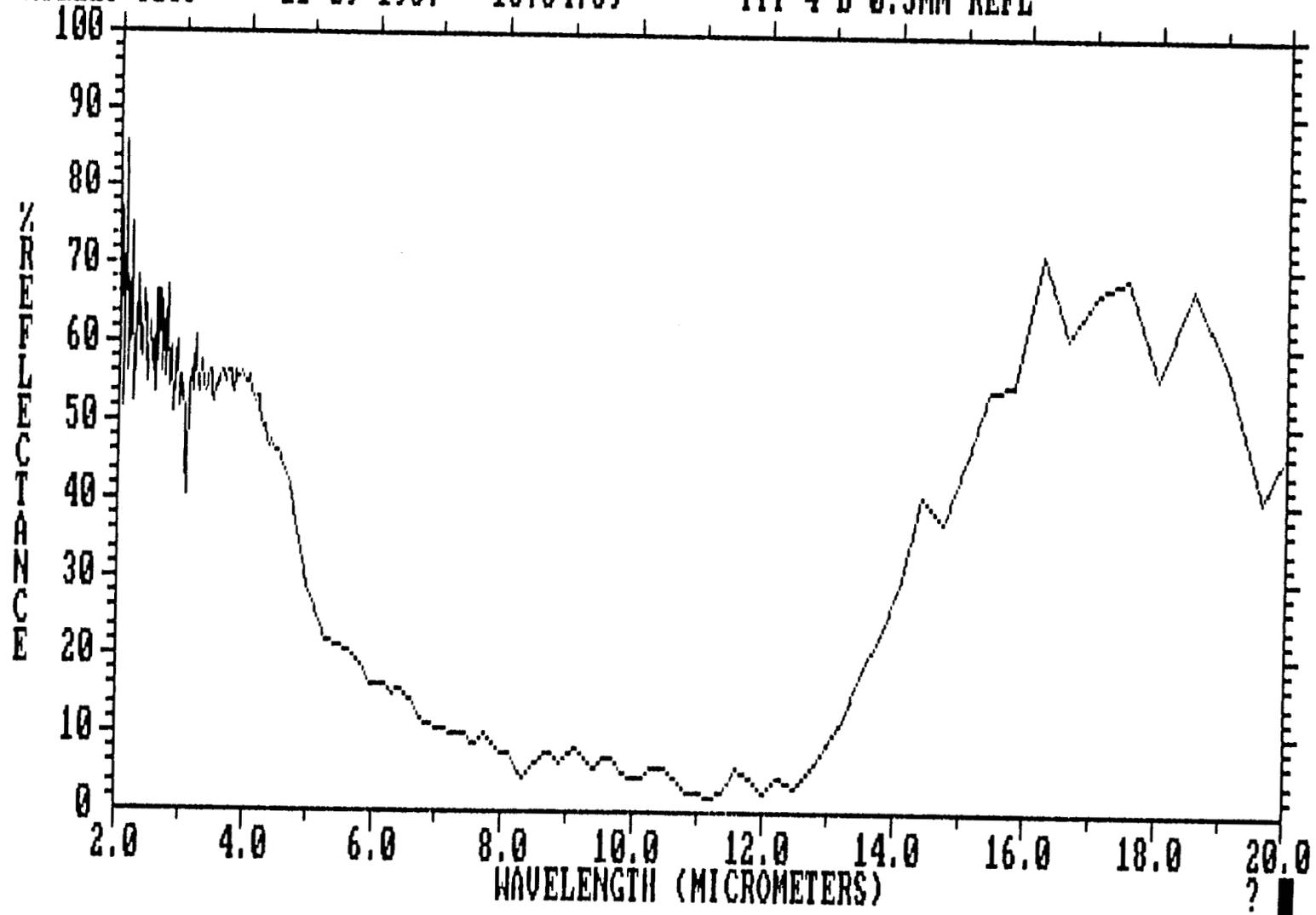


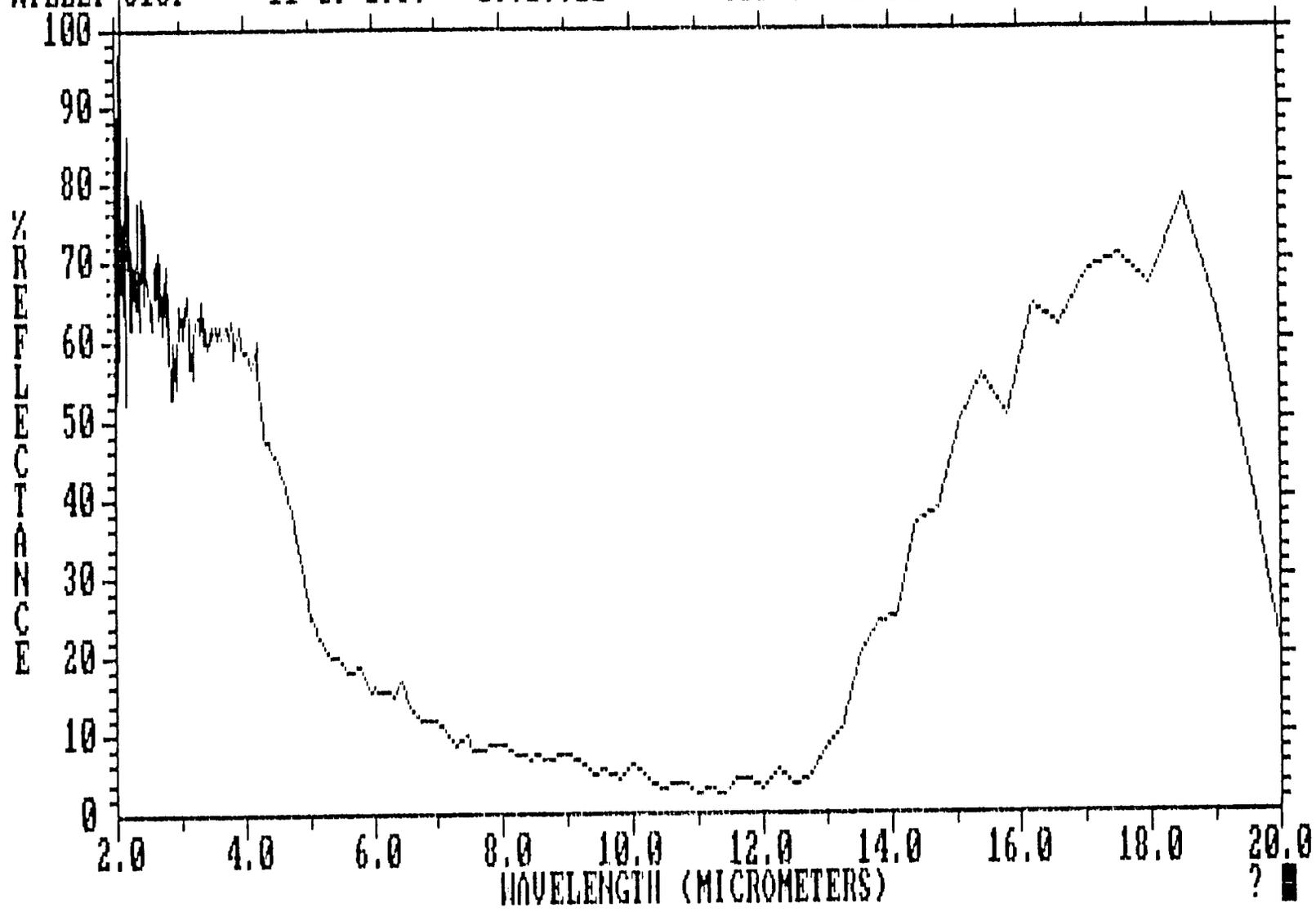
Figure A-4 Unsmoothed Reflectance Spectra of Sample B4

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19:27:11

ITI 5 B 1.0MM REFL



74

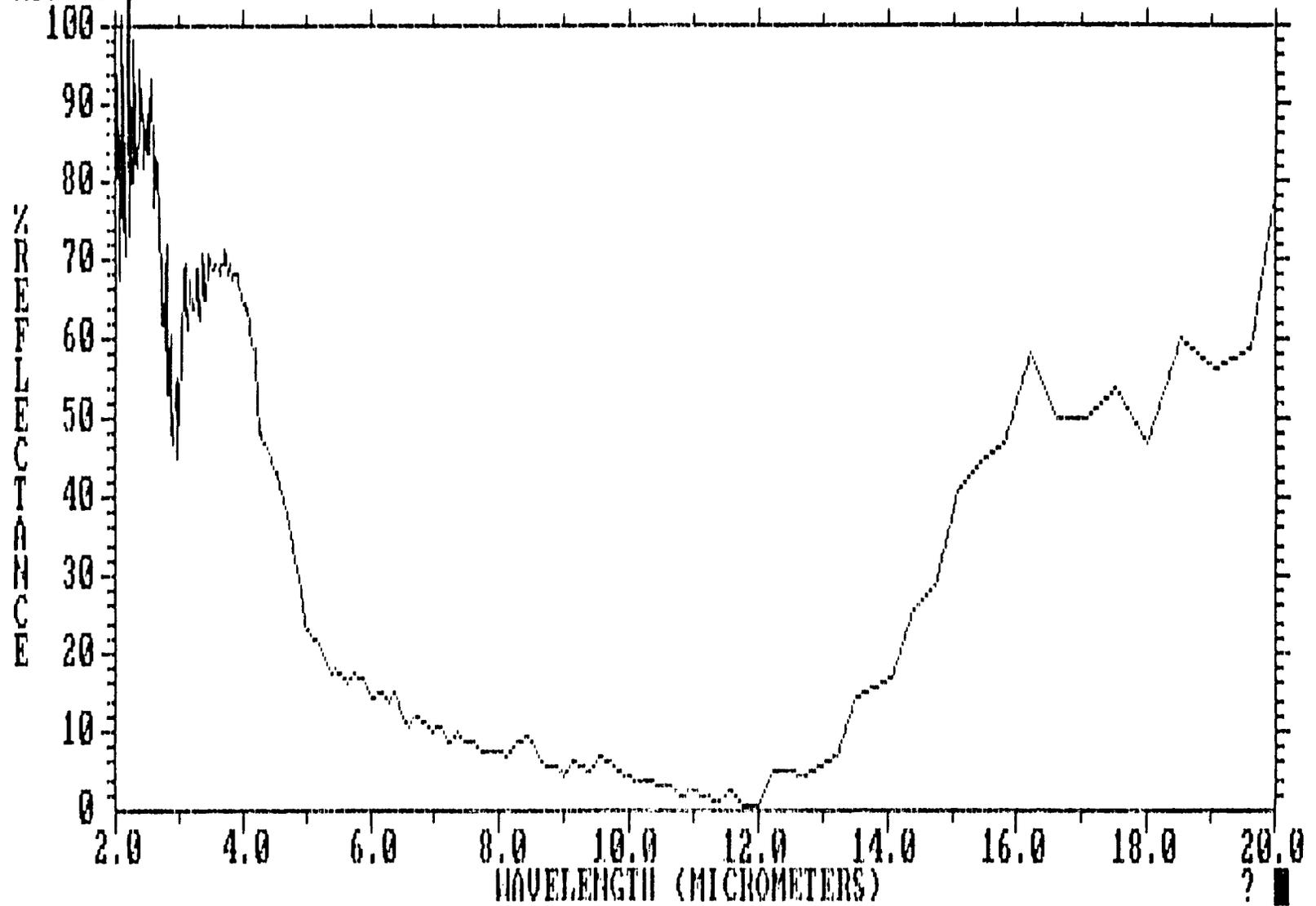
Figure A-5 Unsmoothed Reflectance Spectra of Sample B5

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20:12:02

ITI 6 B 10.0MM REFL



75

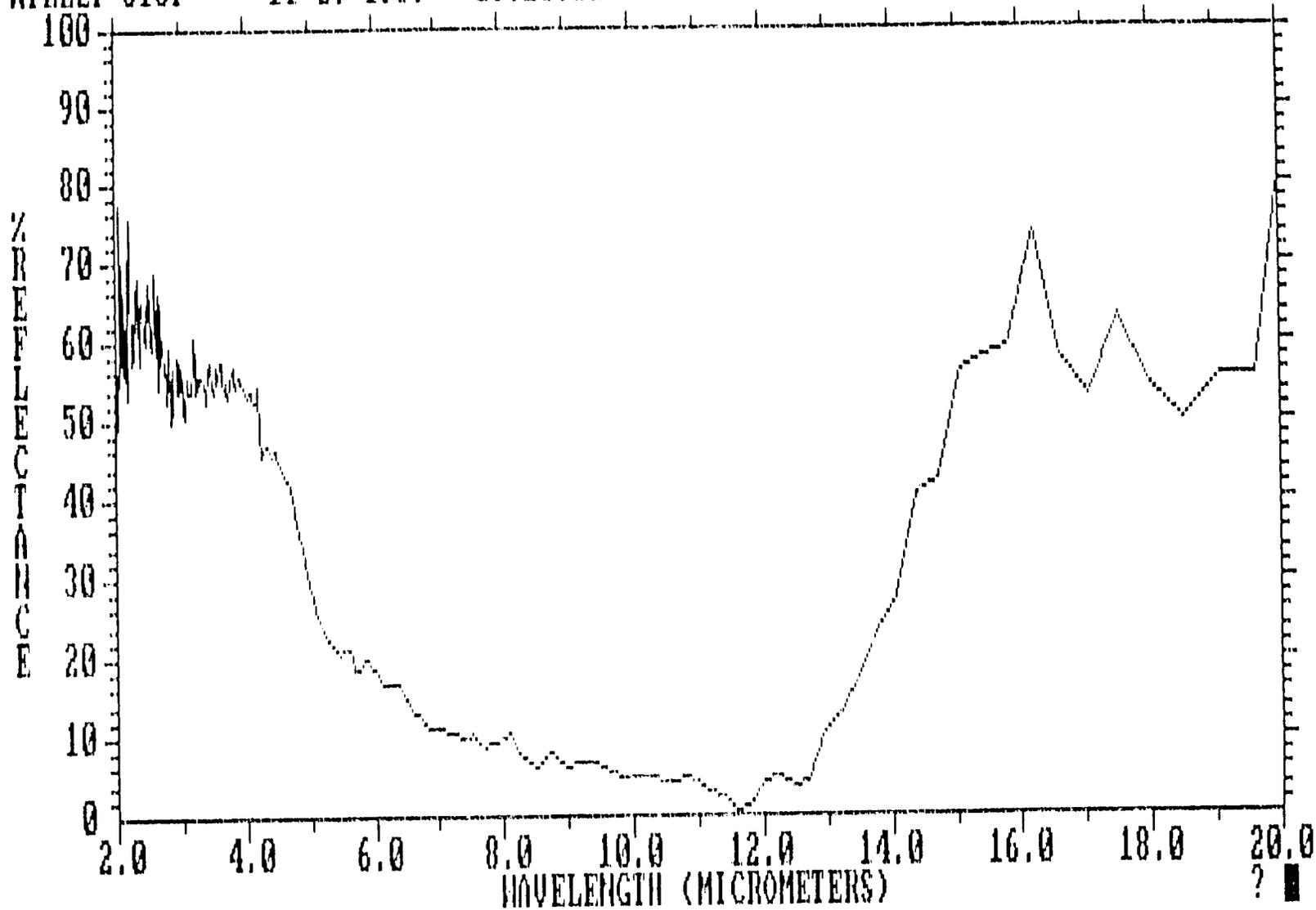
Figure A-6 Unsmoothed Reflectance Spectra of Sample B6

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19:26:50

ITI 7 C 0.5MM REFL



76

Figure A-7 Unsmoothed Reflectance Spectra of Sample C7

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19:48:16

ITI 8 C 1.0MM REFL

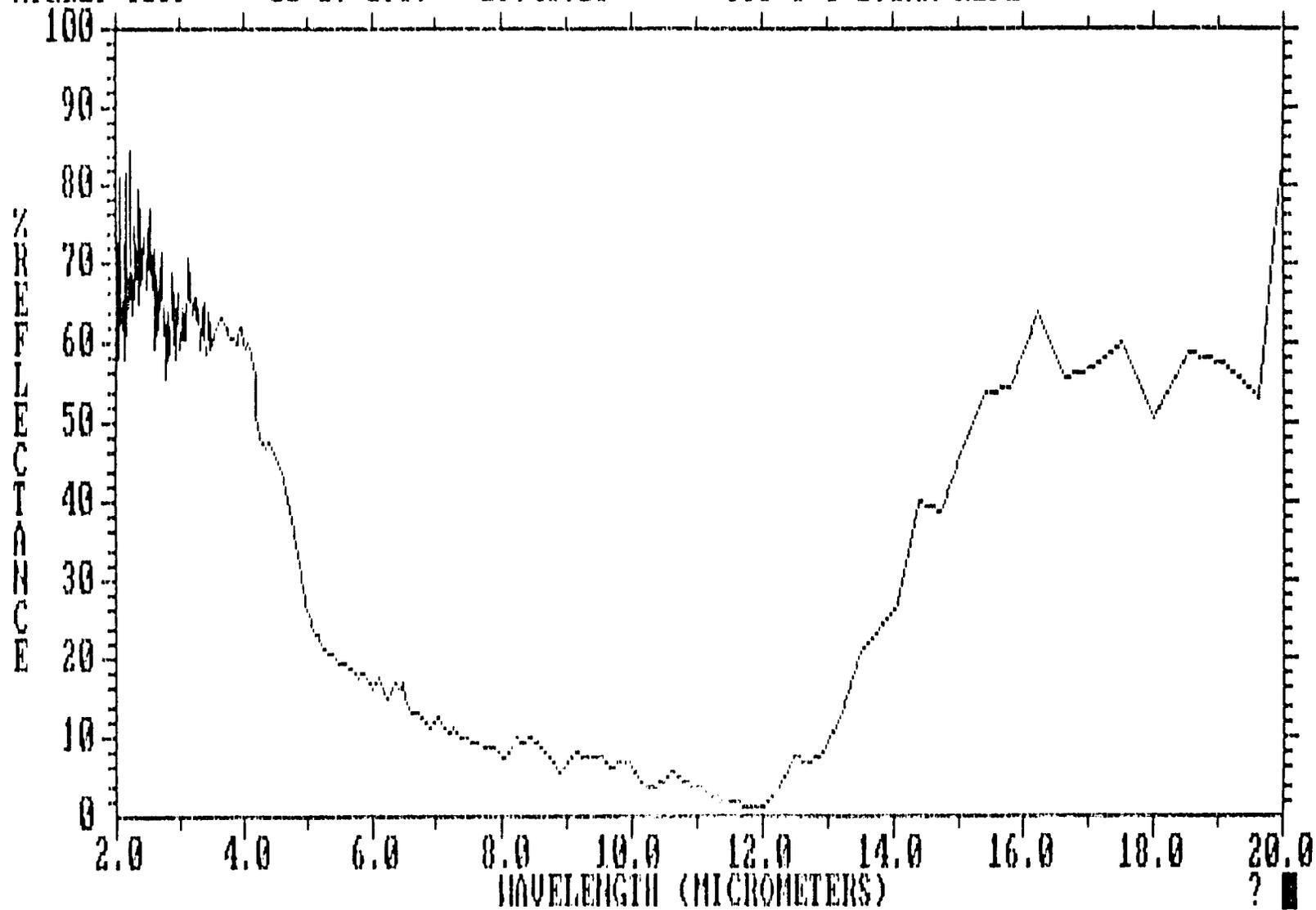


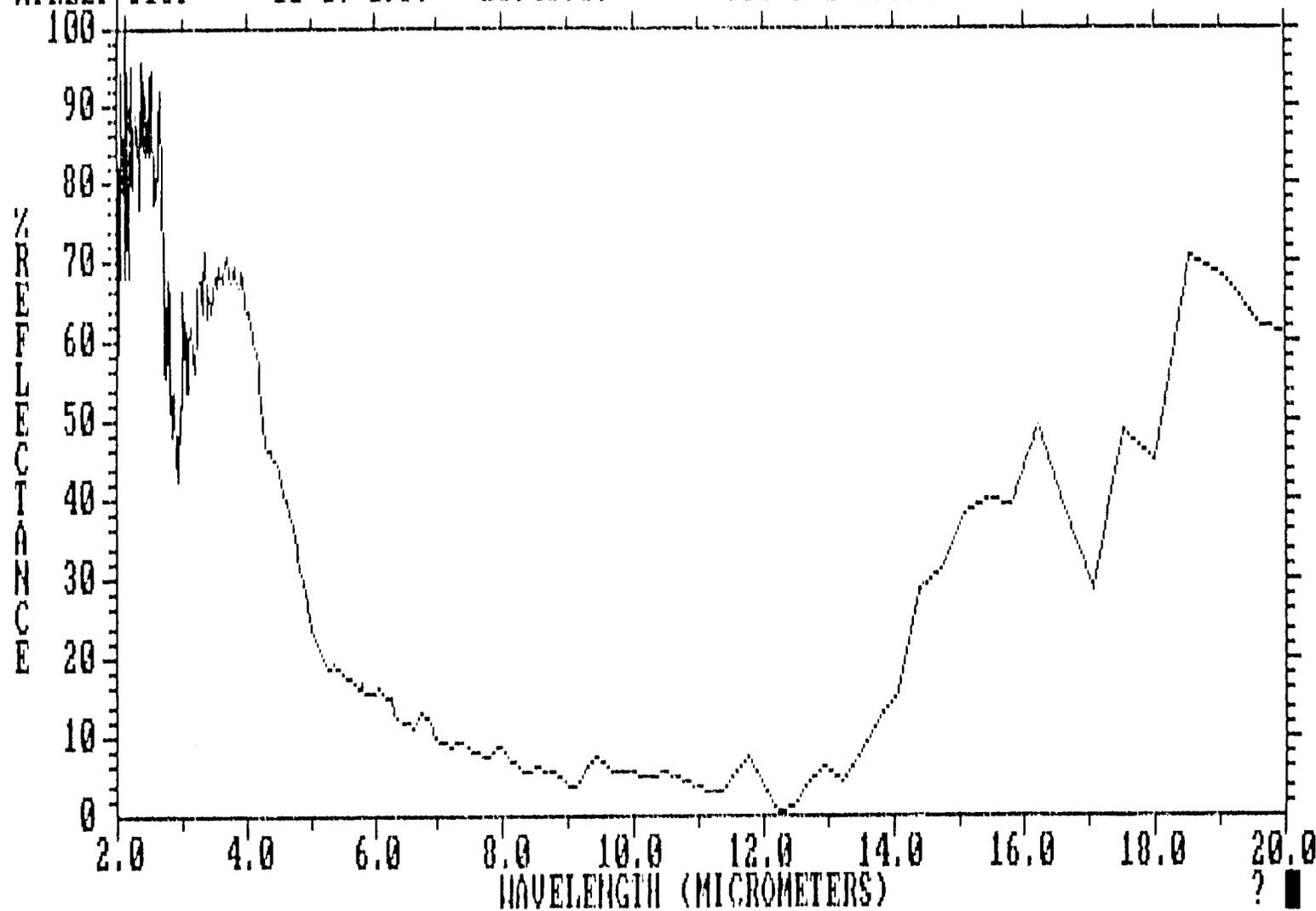
Figure A-8 Unsmoothed Reflectance Spectra of Sample C8

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20:43:37

ITI 9 C 10.0MM REFL



78

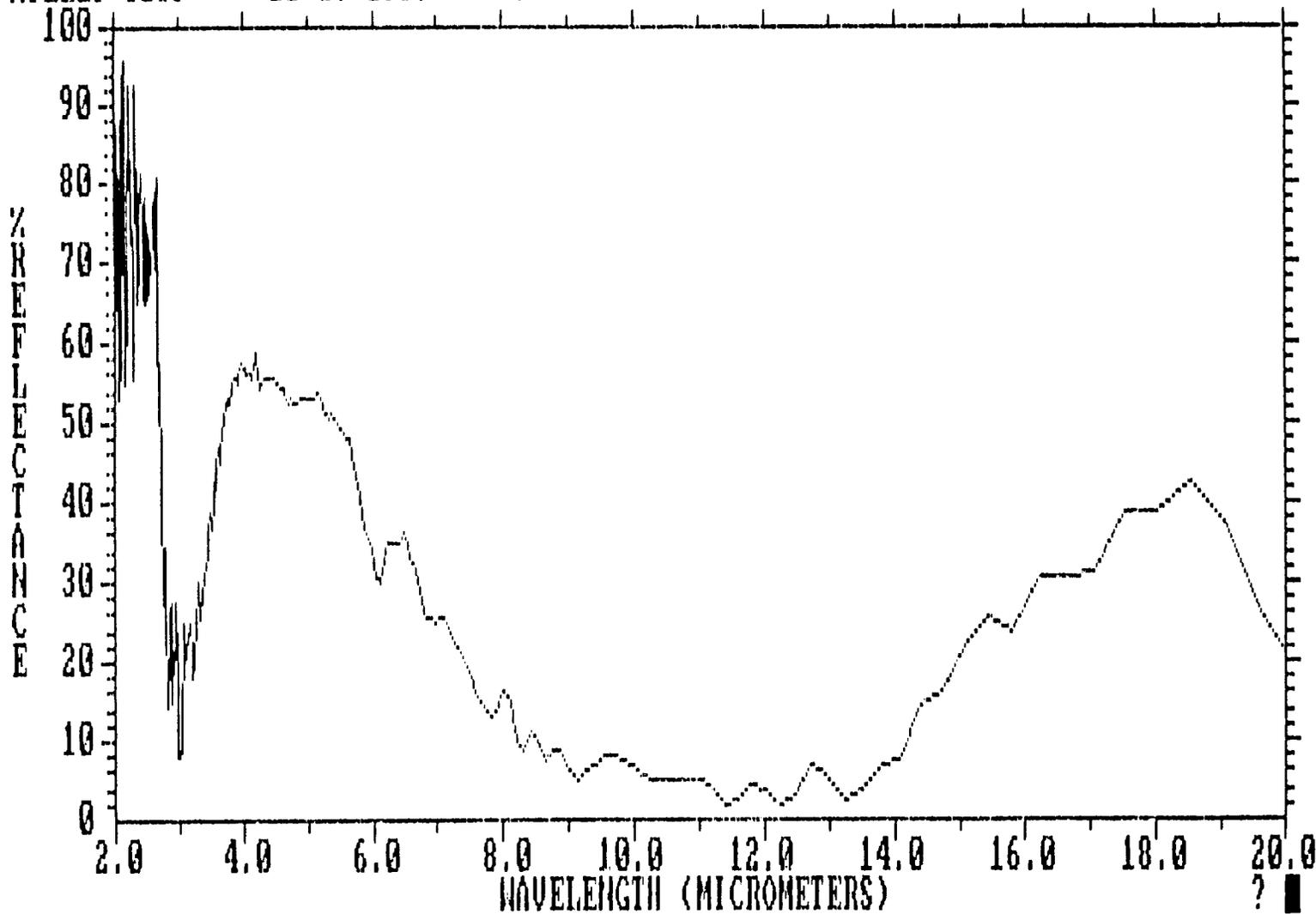
Figure A-9 Unsmoothed Reflectance Spectra of Sample C9

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21:12:18

ITI 10 D .016 IN. REFL



79

Figure A-10 Unsmoothed Reflectance Spectra of Sample D10

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22:13:47

ITI 11 D .023 IN. REFL

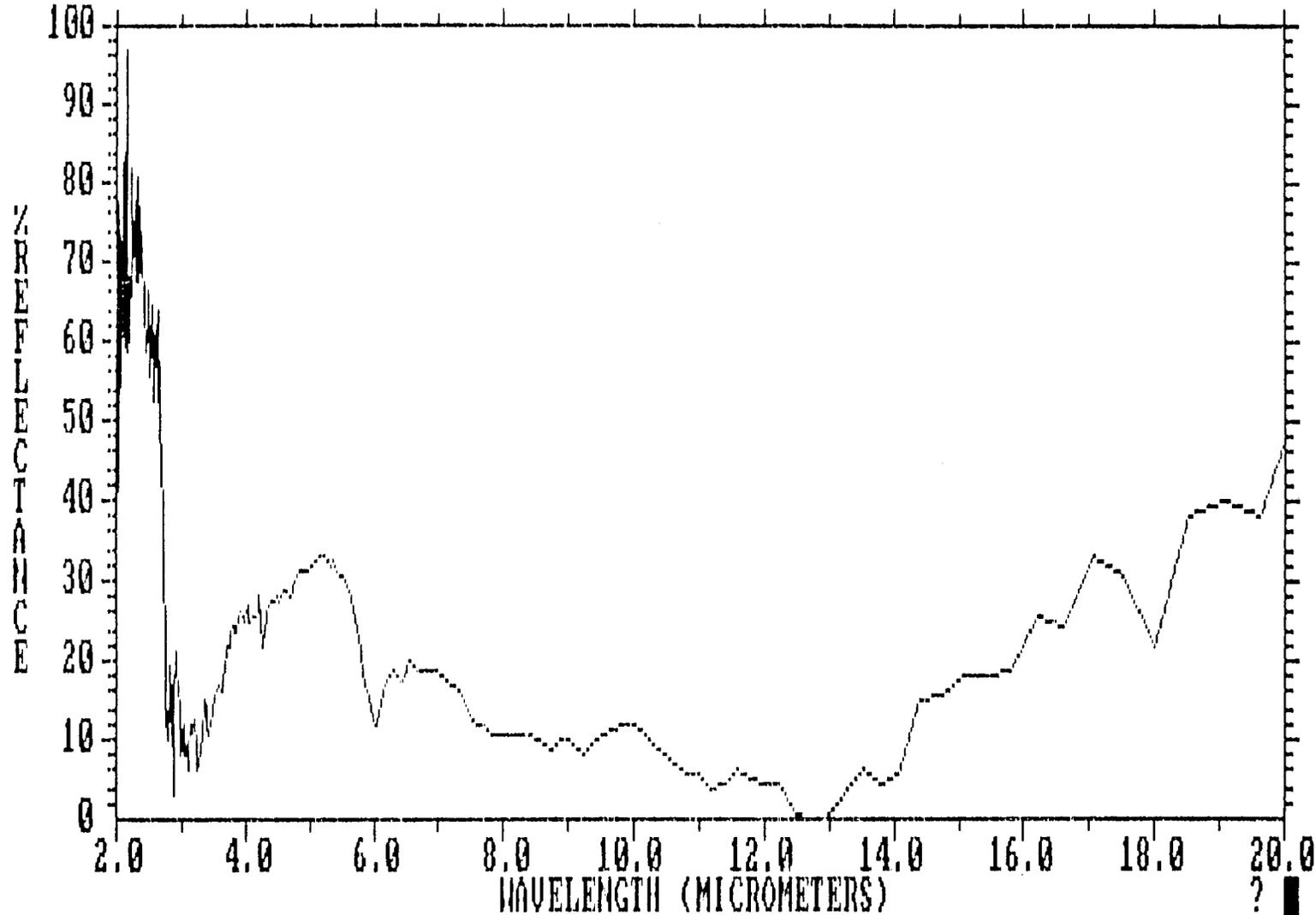


Figure A-11 Unsmoothed Reflectance Spectra of Sample D11

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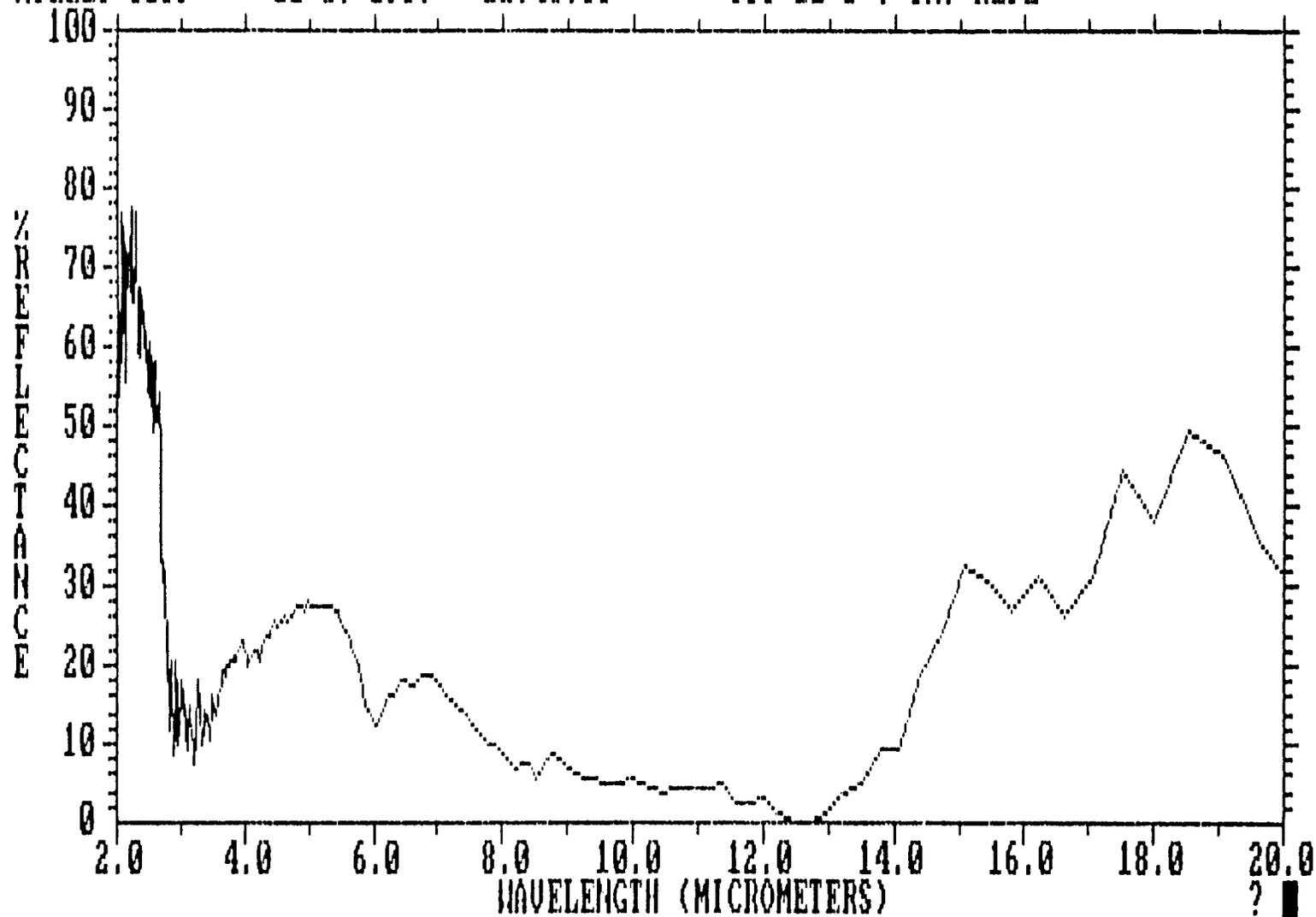


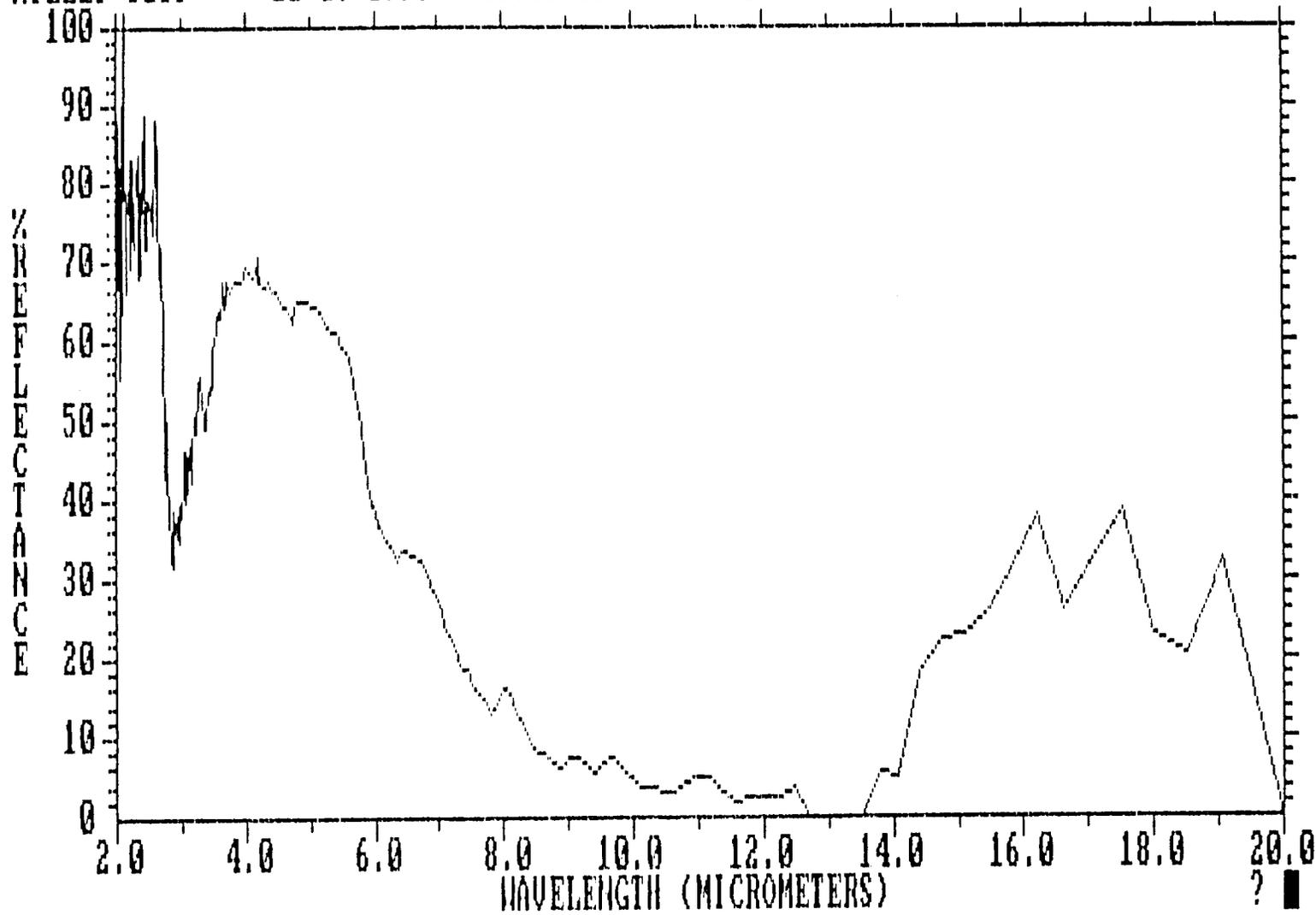
Figure A-12 Unsmoothed Reflectance Spectra of Sample D12

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23:30:35

ITI 13 D .098 IN. REFL



82

Figure A-13 Unsmoothed Reflectance Spectra of Sample D13

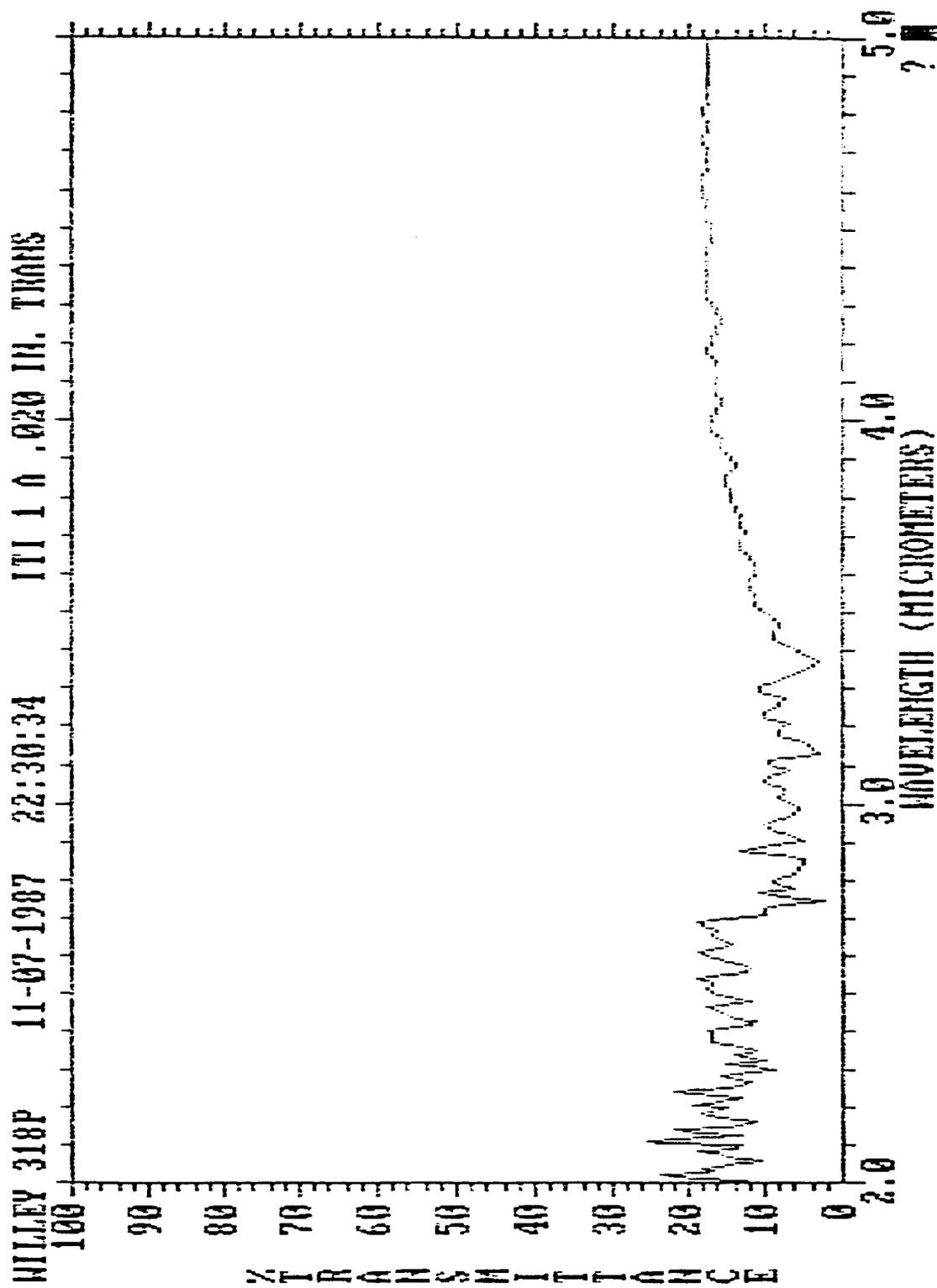


Figure A-14 Unsmoothed Reflectance Spectra of Sample A1

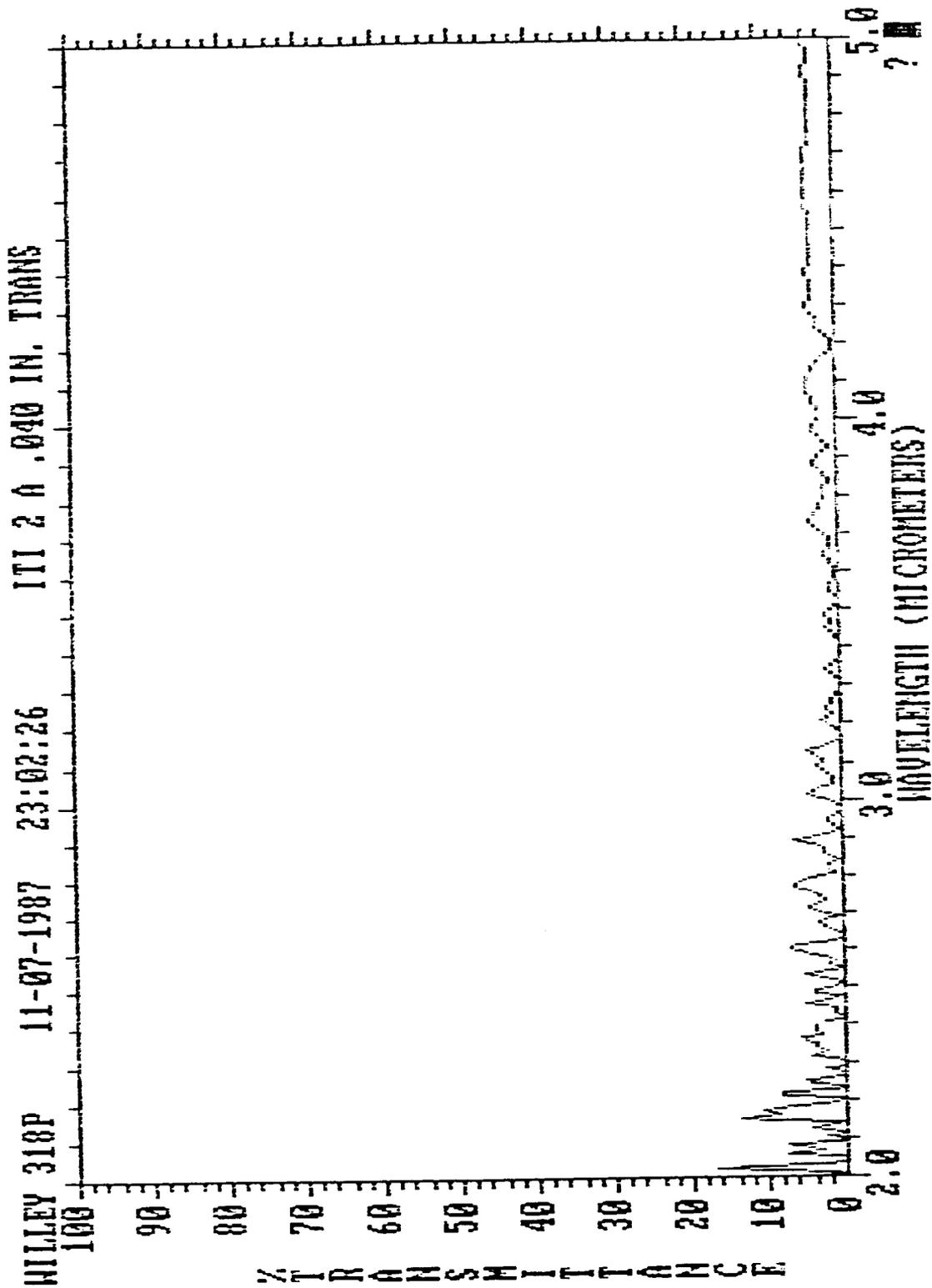


Figure A-15 Unsmoothed Reflectance Spectra of Sample A2

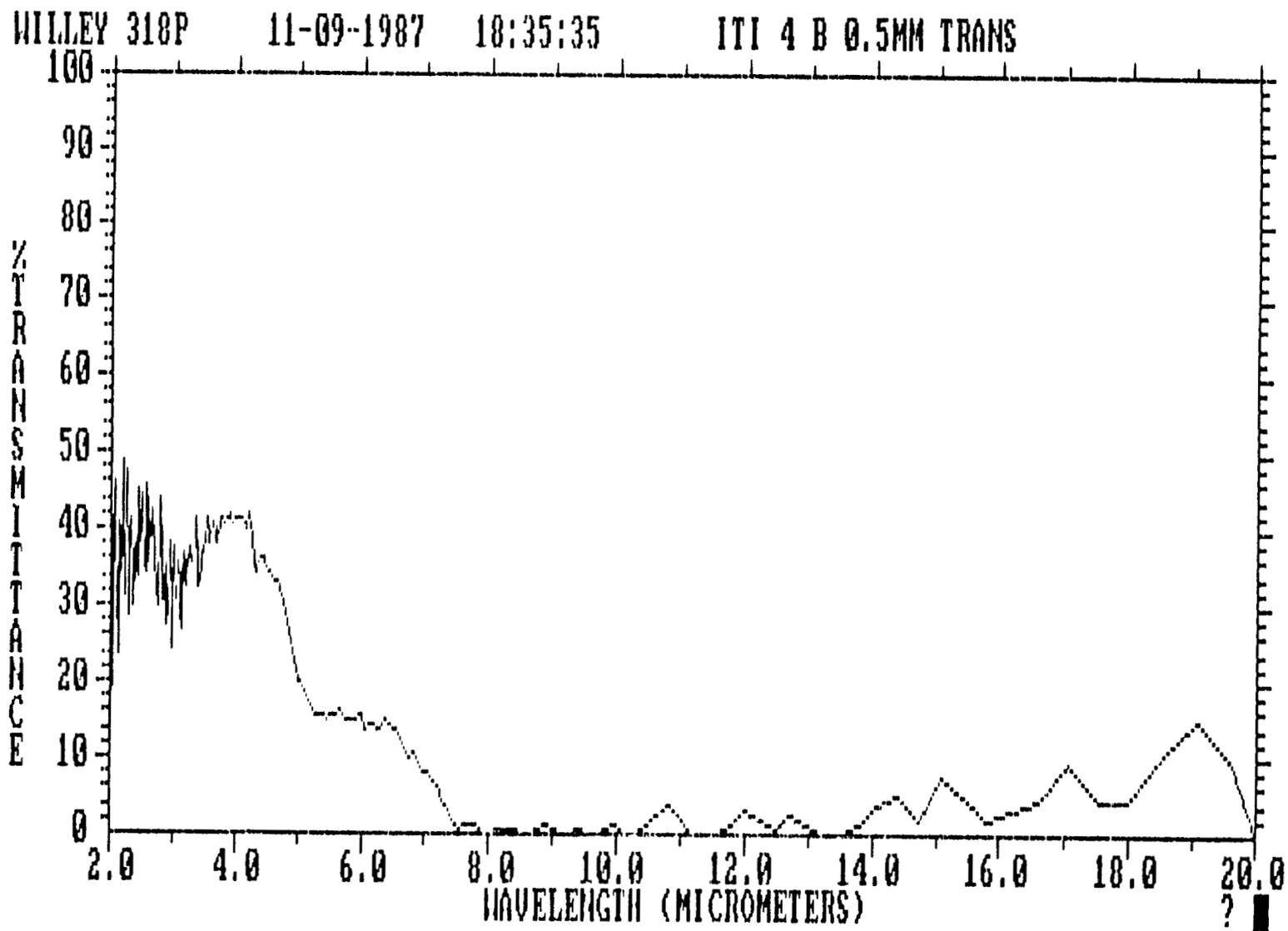


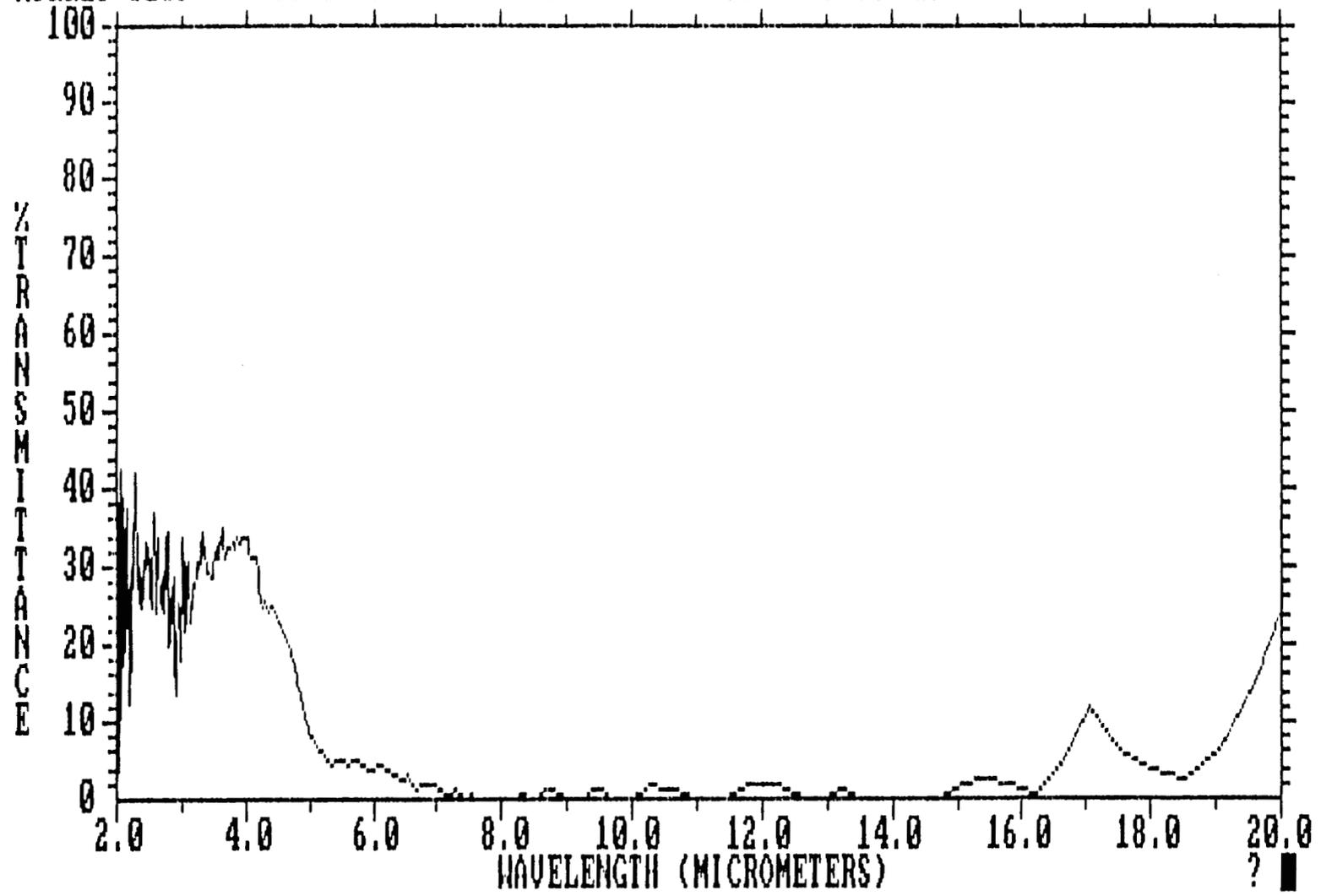
Figure A-16 Unsmoothed Reflectance Spectra of Sample B4

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19:53:27

ITI 5 B 1.0MM TRANS



86

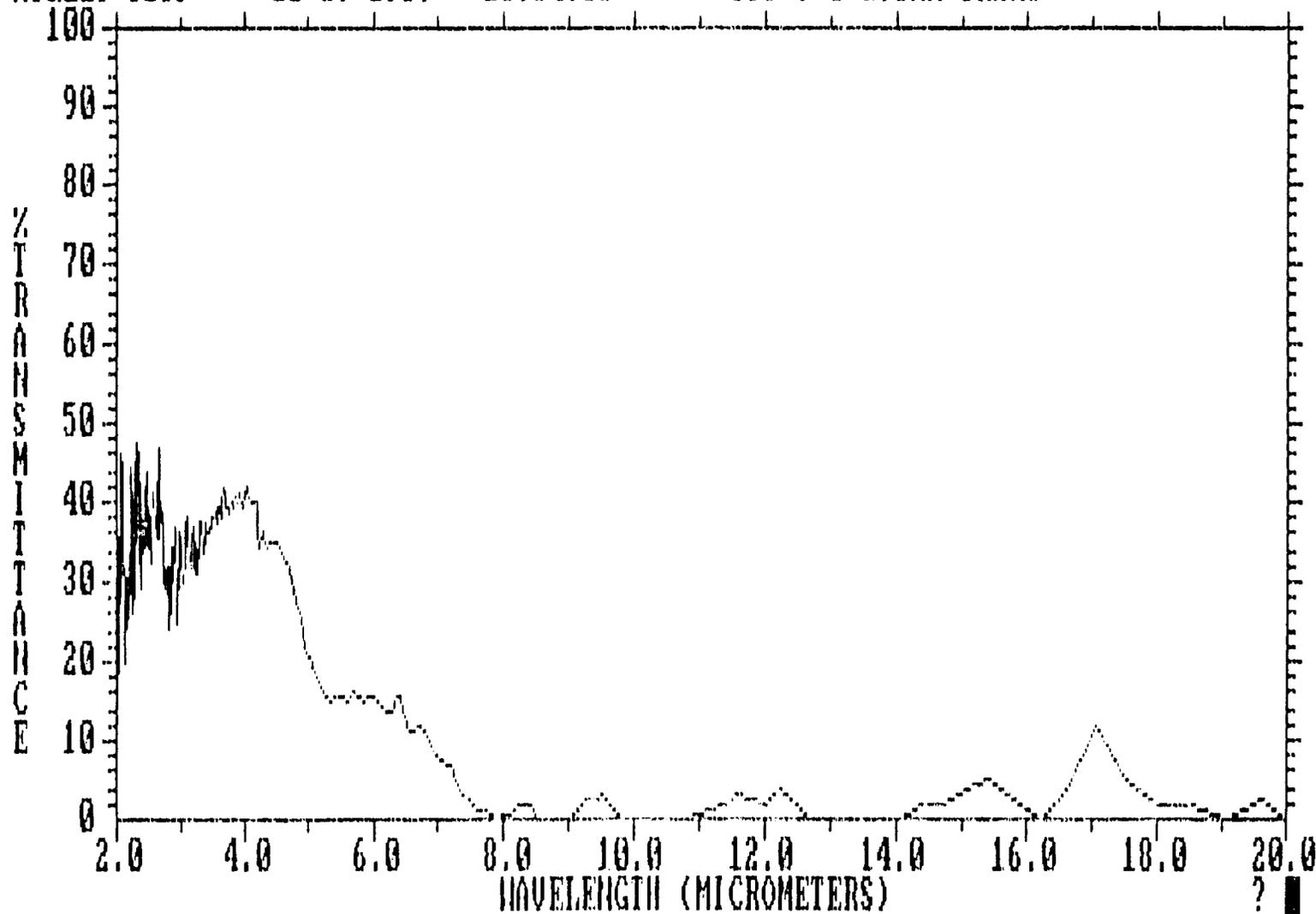
Figure A-17 Unsmoothed Reflectance Spectra of Sample B5

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18:54:50

ITI 7 C 0.5MM TRANS



87

Figure A-18 Unsmoothed Reflectance Spectra of Sample C7

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ITI 8 C 1.0MM TRANS

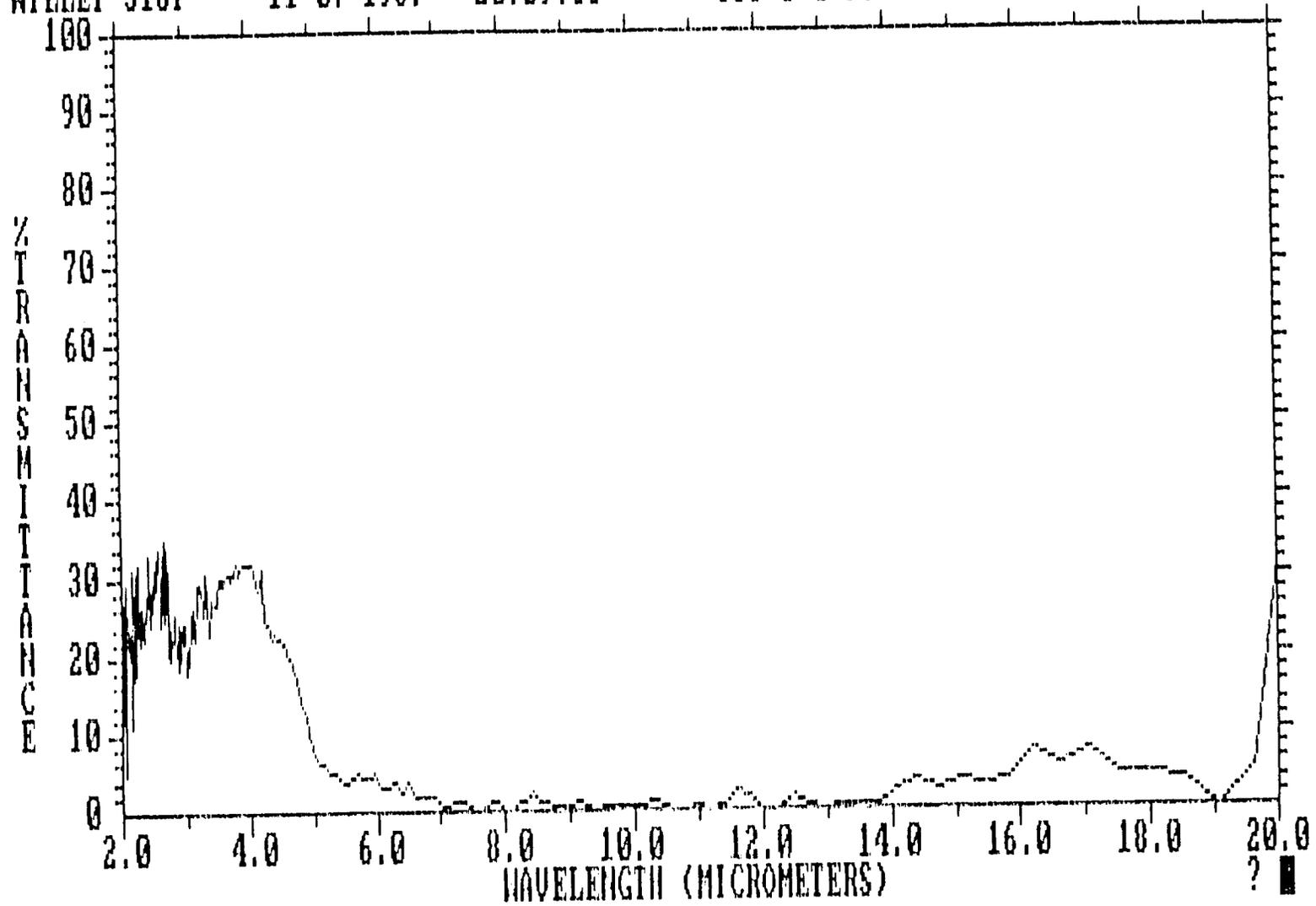


Figure A-19 Unsmoothed Reflectance Spectra of Sample C8

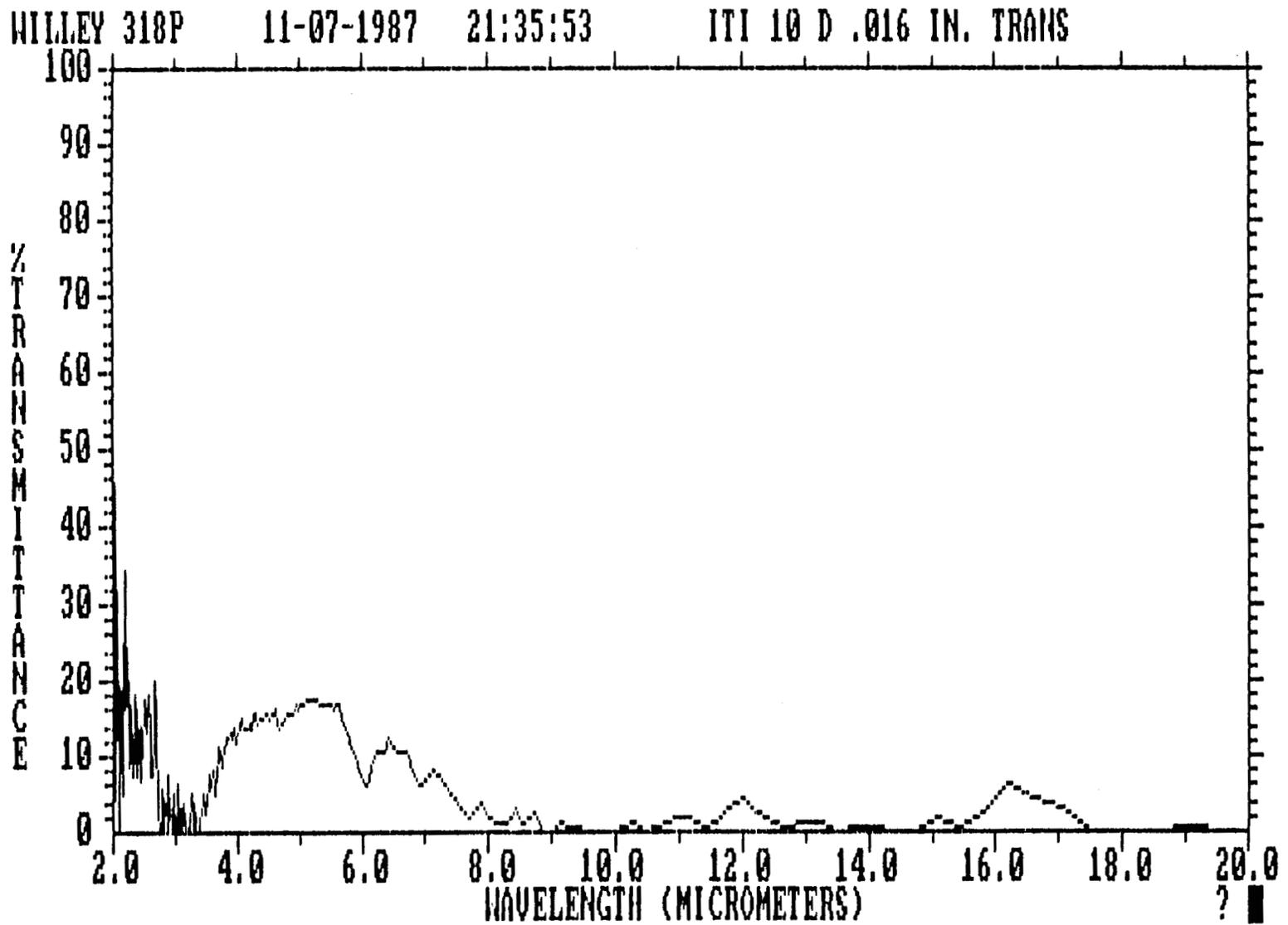


Figure A-20 Unsmoothed Reflectance Spectra of Sample D10



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