NASA TECHNICAL NOTE

NASA TN D-5647

c. 1

NASA TN D-5647

SPECTRAL EMITTANCE OF SOOT

by Curt H. Liebert and Robert R. Hibbard

Lewis Research Center
Cleveland, Ohio

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • FEBRUARY 1970

1. Report No. NASA TN D-5647	2. Government Accession No.	3. Recipient's Catalog No.
4. Title and Subtitle SPECTRAL EMITTANCE OF SOOT		5. Report Date February 1970
		6. Performing Organization Code
7. Author(s) Curt H. Liebert and Robert R. Hibbard		8. Performing Organization Report No. E-5435
9. Performing Organization Name and Address Lewis Research Center National Aeronautics and Space Administration Cleveland, Ohio 44135		10. Work Unit No. 129-01
		11. Contract or Grant No.
12. Sponsoring Agency Name and Address National Aeronautics and Space Administration Washington, D.C. 20546		13. Type of Report and Period Covered Technical Note
		14. Sponsoring Agency Code
15. Supplementary Notes		

16. Abstract

The spectral emittances of thin layers of soot were experimentally determined over the wavelength range of 0.35 to 14 μm and at the two temperatures of 300 and 670 K. The soot was deposited from a smoky flame on transparent and reflective substrates, and soot layer thicknesses and emittances were measured on these substrates. The experimental results are compared with the predictions of simplified classical radiation absorption theory and with Hottel's widely used empirical relation. The results indicate that good assessments of the emittances of soot over broad ranges of temperature, thickness, and wavelength can be obtained by use of either the classical theory or the empirical theory.

17. Key Words (Suggested by Author(s)) Emittance, Soot, Smoke, Mie theory		18. Distribution Statement Unclassified - unlimited	
19. Security Classif. (of this report) Unclassified	20. Security Classif. (of this page) Unclassified	21. No. of Pages 12	22. Price* $3.00

*For sale by the Clearinghouse for Federal Scientific and Technical Information Springfield, Virginia 22151

SPECTRAL EMITTANCE OF SOOT

by Curt H. Liebert and Robert R. Hibbard

Lewis Research Center

SUMMARY

The spectral emittances of thin layers of soot were experimentally determined over the wavelength range of 0.35 to 14 micrometers and at the two temperatures of 300 and 670 K. The soot was deposited from a smoky flame on transparent and reflective substrates, and soot layer thicknesses and emittances were measured on these substrates. The experimental results are compared with the predictions of simplified classical radiation absorption theory and with Hottel's widely used empirical relation. The results indicate that good assessments of the emittances of soot over broad ranges of temperature, thickness, and wavelength can be obtained by use of either the classical theory or the empirical theory.

INTRODUCTION

There are two reasons for wanting to relate the spectral emittance of soot with its quantity in a system. First, the amount of heat radiated by luminous flames is strongly dependent on radiation from the soot contained in these flames. The radiation from the nonluminous gaseous combustion products can be calculated by using well-established methods (e.g., ref. 1), but the added contribution from soot is very important because it can double or triple the heat that would be radiated by the gaseous products alone. Second, emittance measurements may be used to estimate soot concentrations in combustion systems. This would be of considerable interest to turbine engine designers who are concerned with the amount of smoke discharged by these engines. The use of emittance measurements deserves attention because no other methods are known by which soot concentrations can be measured in the flame zones of combustors operating at high pressures (ref. 2). In both cases it would be helpful to have quantitative relations between flame emittance and soot concentration. Then, depending on engineering needs, the radiant energy could be estimated from known soot concentrations, or the concentration of soot could be estimated from emittance data.

The emittance and heat flux radiated from the soot in a luminous flame can be approximately calculated from the Mie theory (ref. 3) if the soot concentration, flame thickness, and temperature are known. This theory is based on the interaction of an electromagnetic wave with a spherical particle. Other techniques which are used to relate flame emittance and flame temperature are based on the empirical formulation proposed by Hottel (ref. 4). From data in the literature and his own measurements of the transmittance of soot deposits on substrates, Hottel found that the spectral absorption coefficient of soot is inversely proportional to the wavelength raised to a power. This relation, when substituted into Wien's formula (an approximation to Planck's blackbody distribution) yields simple, convenient relations for calculating luminous flame temperatures from optical pyrometry data.

Svet (ref. 5) presents an extensive bibliography of the spectral reflectance and transmittance of layers of soot deposited on transparent substrates and measured at room temperature. This reference shows that the reflectance of soot is only 0.006 to 0.05. It follows from the law of conservation of energy that reflectance plus absorptance plus transmittance equals 1. Thus, because reflectance is negligible, the emittance of soot at room temperature may be calculated directly from the transmittance by assuming that spectral emittance equals 1 minus spectral transmittance.

With zero reflectance, the emittance of soot must increase with increasing thickness. However, no quantitative data were found in the literature in which soot emittance was determined as a function of thickness or mass path length. Also, no data were found on measurements of soot emittance as a function of thickness at elevated temperatures.

Reported herein are data on the normal spectral emittances measured for five thicknesses of deposited soot varying from 0.05 to 1 micrometer (0.05×10^{-4} to 10^{-4} cm) and at two temperatures, 300 K (room temperature) and 670 K. The soot was produced from a stearin candle and deposited on polished sodium chloride (NaCl) and on polished aluminum substrates. The room-temperature spectral emittance of soot was calculated from transmittance measurements of deposits of soot on NaCl substrates taken at wavelengths of 0.35 to 14 micrometers. Spectral emittances at 670 K of the soot deposits on the aluminum substrates were directly measured at wavelengths of 2 to 14 micrometers. The surface of the aluminum substrate was highly reflective and thus contributed negligible emitted energy to the measurement. The experimental data are compared with calculated values of spectral emittance based on the Mie theory and with Hottel's formulation.

PROCEDURE

Sample Preparation and Thickness Measurement

Soot from the smoky flame of a stearin candle was deposited on polished aluminum and on polished NaCl substrates by passing the surfaces through the flame at a given height above the wick and at a controlled rate. This was done by hand and, after considerable practice, deposits with nominal thicknesses of 0.05, 0.14, 0.27, 0.40, and 1.0 micrometer could be repeated over the area viewed by the emittance measuring instruments. This area was about 1 square centimeter. One pass was required to deposit the smallest thickness, five passes were needed for the largest thickness. The deposits adhered well to the substrates.

These thicknesses were measured with a microscope which uses the interference of light to measure thickness. The measuring range of this instrument was about 0.03 to 2.0 micrometers, and its accuracy is within ±0.01 micrometer.

The interference microscope technique could be used to measure deposit thickness only on the highly reflective aluminum substrates. The thickness on the NaCl substrates was controlled to that measured on the aluminum substrates by making nearly identical passes of the surfaces through the flame.

The soot thickness varied somewhat over the surface area of each aluminum substrate. The variation of thickness in micrometers about a nominal thickness measured on a given sample was ±0.01 at thicknesses of 0.05 and 0.14, ±0.02 at thicknesses of 0.27 and 0.40, and ±0.04 at a thickness of 1.0. Slight variations of soot deposit thickness were also visually noted on the NaCl substrates.

Emittance Measurements

From room-temperature transmittance data. - Two spectrophotometers with different spectral ranges were used to measure the nearly normal transmittance at room temperature of the soot deposited on NaCl at wavelengths between 0.35 to 2 micrometers and between 2.5 to 14 micrometers. The experimental error of the instruments is ±3 percent, but the sample-to-sample transmittance measurements through soot coating and substrate could only be repeated within ±15 percent. Since the transmittance of the soot varies greatly with thickness, the range of repeatability is mostly associated with the inability to reproduce a nominal thickness closer than ±0.01 to ±0.04 micrometer. The room-temperature emittance was calculated from the transmittance measurements by

$$\epsilon_\lambda = 1 - \tau_\lambda \tag{1}$$

where ϵ_λ is the spectral emittance and τ_λ is the spectral transmittance. The transmittance of the uncoated NaCl window over the wavelength regions investigated was 0.90 at wavelengths of 0.35 to 2 micrometers and varied from 0.90 to 0.96 at longer wavelengths. The soot transmittance was corrected for this effect.

From high-temperature emittance data. - A recording infrared spectrophotometer with an emittance-reflectance attachment was used to directly measure the nearly normal spectral emittance of soot on optically polished aluminum at 670 K over a wavelength range of 2 to 14 micrometers. The polished aluminum did not oxidize in the few hours required to collect the data, and its measured emittance was less than 0.03. The data were not corrected for this effect.

At 670 K the oxidation rate of the soot was also slow enough so that there was no measurable change in emittance over several hours. However, soot was rapidly oxidized at 50 K higher temperature so that data could not be obtained. At 50 K lower temperature there was insufficient energy for satisfactory instrument measurements. The two limitations fixed the 670 K temperature that was used.

The uncertainty associated with the emittance measurements made with this spectrophotometer is discussed in reference 6. This reference shows that experimental errors will arise because of temperature gradients in the blackbody source used as a reference and because of heat loss through the thermocouple wires attached to the specimen. Using the procedures described in reference 6, the uncertainty of spectral emittance presented herein was determined as ±0.05.

Using fresh deposits of soot, emittance measurements could be repeated within ±0.10 at given nominal thicknesses. The uncertainty of the emittance measurement and the inability to precisely reproduce a desired thickness can account for this range.

RESULTS AND DISCUSSION

Figure 1 presents the variation of the normal spectral emittance for the various thicknesses of soot. Except for the curve corresponding to 0.05-micrometer thickness, which was obtained at room temperature only, the smooth curves in figure 1 were faired through the values of emittances obtained at the two temperatures; all data and the curves agreed within ±15 percent. The vertical lines at selected wavelengths indicate the highest and lowest values of emittance obtained.

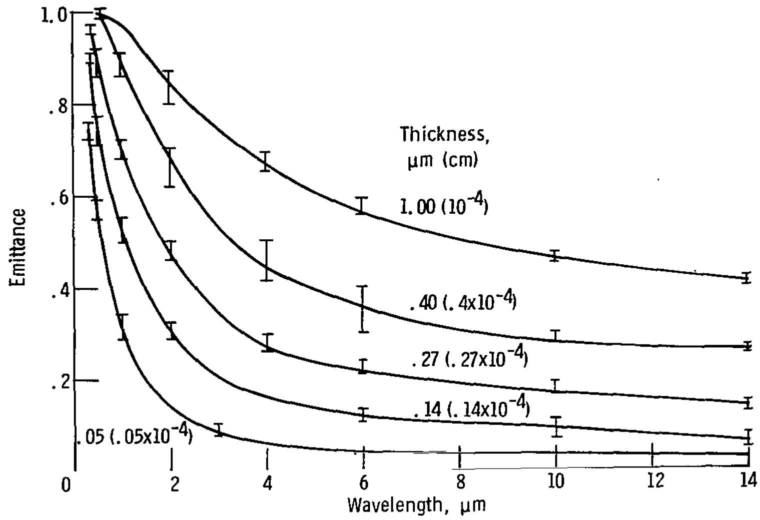


Figure 1. - Normal spectral emittance at various thicknesses of soot layers. (Data taken at 300 and 670 K.)

Within a range of ±15 percent, the normal spectral emittances at comparable thickness were the same at 300 K or 670 K. Thus, within the ability to produce identical coating thickness on the various substrates, the emittances are temperature invariant in this range of temperatures. This result is not surprising because reference 5 shows that the emittance of graphite, another form of carbon, varies less than 3 percent over a temperature range of 1200 to 2100 K and at a wavelength of 0.665 micrometer. Therefore, the two measuring techniques provide a good check on each other.

At all wavelengths, the emittance varies greatly with thickness. At small thicknesses, the soot is strongly spectrally selective, with the largest emittance occurring at the shorter wavelengths. As thickness increases, the spectral emittance also increases.

Mie Absorption Theory

Stull and Plass (ref. 7) and Foster (ref. 8) have derived simplifications to the Mie theory for predicting soot emittance. Both reduce the Mie equations to useful but different forms. To express their results, Stull and Plass use the number density per unit area, and Foster uses the mass of carbon per unit volume and unit path length. When converted to the same units, both give the same results for particles having diameters of 0.05 micrometer or less. It was easier to compare the experimental spectral emittance data with the Mie theory by using the Foster calculations and the relation

$$\epsilon_\lambda = 1 - \exp(-K_\lambda \bar{c} L) \tag{2}$$

where

K_λ spectral attenuation coefficient, cm^2/g

$\bar{c}$ soot concentration, g/cm^3

L soot thickness, cm

The Mie-Foster theory assumes that the particles are round, randomly distributed, small with respect to wavelength, and separated from each other by distances that are large compared with wavelength. In addition, the theory assumes that the complex refractive index of graphite determined experimentally in the literature at 2250 K applies also to soot and that the density of a single particle of soot is 2 grams per cubic centimeter. Reference 9 gives the single particle density of soot as 1.9 grams per cubic centimeter.

Some of these conditions are not met in these experiments and cannot be met in most practical flame systems. Nevertheless, the practice is to compare the theory with experiment because it is derived by using basic principles and is simple to use.

Foster gives a curve relating the spectral attenuation coefficient K_λ with wavelength for use in equation (2). Foster's curve is reproduced herein as figure 2. The quantity $\bar{c}L$ (the mass path length) in equation (2) is given in terms of mass per unit area

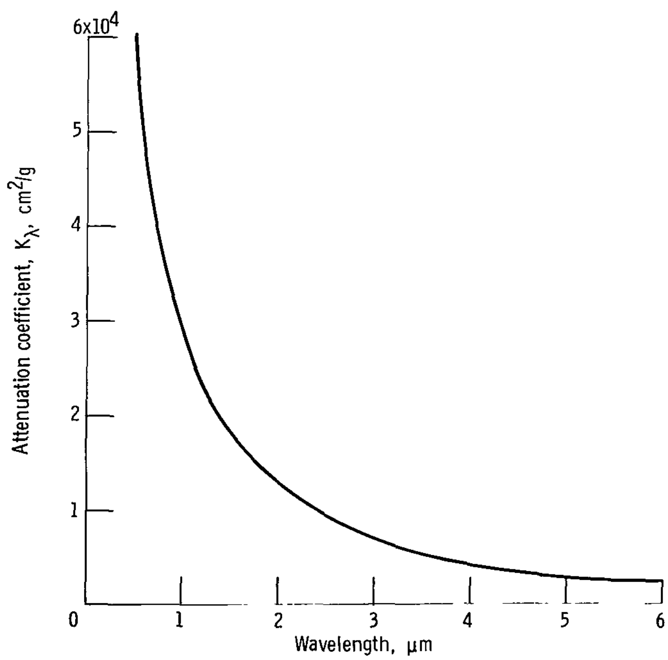
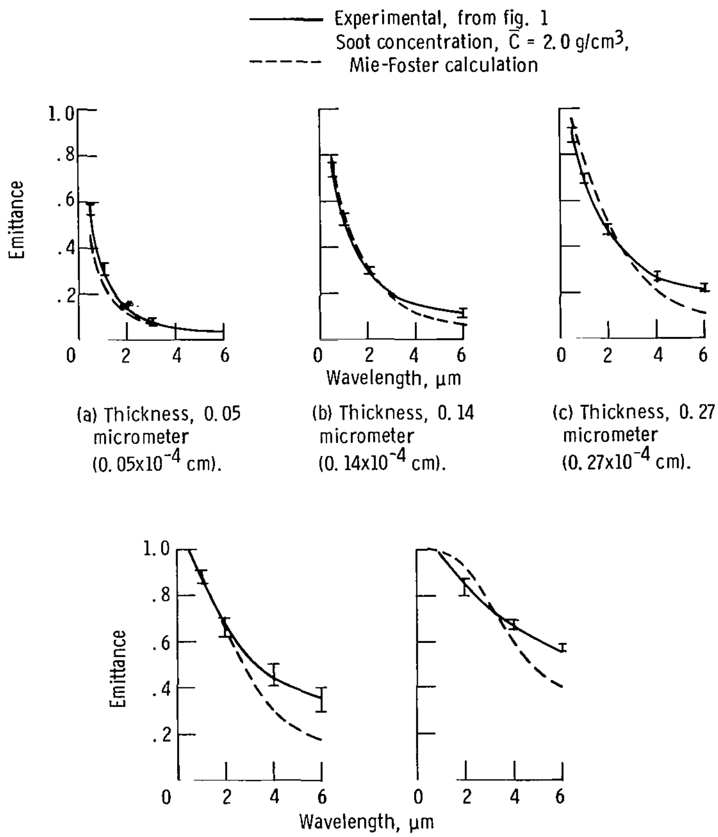


Figure 2. - Calculated attenuation coefficient of soot. (From ref. 8.)

so that comparison between experiment and theory can be made for soot films of given thickness provided the density of the film is known.

The comparison between experimental and calculated spectral emittance at thicknesses between 0.05 and 1.0 micrometer is given in figure 3. The solid line in each part of this figure represents the experimental results redrawn from figure 1. The dashed lines are calculated from equation (2) using the thicknesses shown, a value of $\overline{c}$ of 2.0 grams per cubic centimeter, and coefficients taken from figure 2. The $\overline{c}$ value is for the pure solid; the bulk density of the films of soot will be less. Electron micrographs of the stearin candle soot deposited on an aluminum substrate show that the particles are uniform in size, random and closely packed, and nearly round, with diameters of 0.025 micrometer. The thinnest film (0.05 μm) would be only two particles deep, and the thickest film (1.0 μm) only 40 particles deep. The densities of these films would de-



(a) Thickness, 0.05 micrometer (0.05×10^{-4} cm).

(b) Thickness, 0.14 micrometer (0.14×10^{-4} cm).

(c) Thickness, 0.27 micrometer (0.27×10^{-4} cm).

(d) Thickness, 0.40 micrometer (0.40×10^{-4} cm).

(e) Thickness, 1.00 micrometer (1.00×10^{-4} cm).

Figure 3. - Comparison between experimental and calculated normal spectral emittance of soot.

pend on how the spheres are packed; it is certainly less than 2.0 grams per cubic centimeter. Nevertheless, good agreement is obtained using this density of the pure solid.

The correspondence between experiment and theory obtained herein indicates that the simple mathematical model derived by Foster is suitable for describing the variation of normal spectral emittance of soot over broad ranges of temperature and thickness.

The preceding comparison with the Mie theory is for data from soot deposits, but it is more important to compare the theory with data from sooty flames. Since the soot is less agglomerated in flames than on substrates, the Mie theory, which assumes a fully dispersed system, should be even more successful in predicting the radiant properties of flames. This comparison has been made in reference 3 using Foster's derivation. Total radiation data from small and large flames were compared with theoretical predictions on the basis of the overall (i.e., integrated over all wavelengths) attenuation coefficients. The agreement is generally good. The present work showing good agreement with spectral (i.e., wavelength dependent) properties lends further credence to the conclusion that the Mie theory can be applied to estimate the radiation from the smoke particles in luminous flames.

Other Correlations

The Mie theory and derivations thereof have been used to relate the absolute value of spectral emittance to soot concentration or thickness. However, there are applications where only the wavelength dependence and not the absolute value of the emittance need be known. Examples are the determination of the true temperatures of luminous flames by two-color pyrometry or the estimation of the total emittance of a flame for use in radiant-heat-transfer calculations by using pyrometry data.

Hottel (ref. 4), in developing equations for calculating temperatures and charts for estimating emittance, reviewed a considerable amount of earlier spectral transmittance data on soot layers. He also made measurements of the transmittance of soot. Hottel found that plots of ln [ln (1/transmittance)] against ln of the wavelength in micrometers gave straight lines of (negative) slope α. He also found that α varied between 0.5 and 1.75. Hottel suggested that α has two values in all soot or smoke systems, one for shorter wavelengths and another for longer. From consideration of all the data, he also suggested the use of $\alpha = 1.39$ for wavelengths less than 0.8 micrometer and $\alpha = 0.95$ for longer wavelengths (ref. 1). The former value was recommended for calculating temperatures from two-color pyrometry data, and the latter value for estimating total emittance. Hottel observed no systematic change in α with absorptive strength and therefore recommended that the same numerical values be used for flames of all sizes.

Siddall and McGrath (ref. 10) made a similar study using soot deposited from the flames of a variety of fuels. They found about the same range in α as did Hottel but believed that there was a systematic variation in the numerical value of α as a function of the carbon-hydrogen ratio of the fuel. Siddall and McGrath did not measure soot thicknesses, nor did Hottel.

In the present study the thickness of the soot was varied and measured. Therefore, to compare the wavelength dependence of spectral emittance with the earlier work, the curves of figure 1 were replotted as ln [ln (1/transmittance)] against ln of wavelength. The results are shown in figure 4.

Figure 4 shows essentially straight lines (constant value of α) over much of the wavelength range for all five soot thicknesses. It does not show a change of slope at wavelengths of about 0.8 micrometer as suggested by Hottel, although there appears to be some reduction of slope at wavelengths around 7 micrometer. There is also an indication of mild variation in α with thickness, the thinner films having the greater value of α.

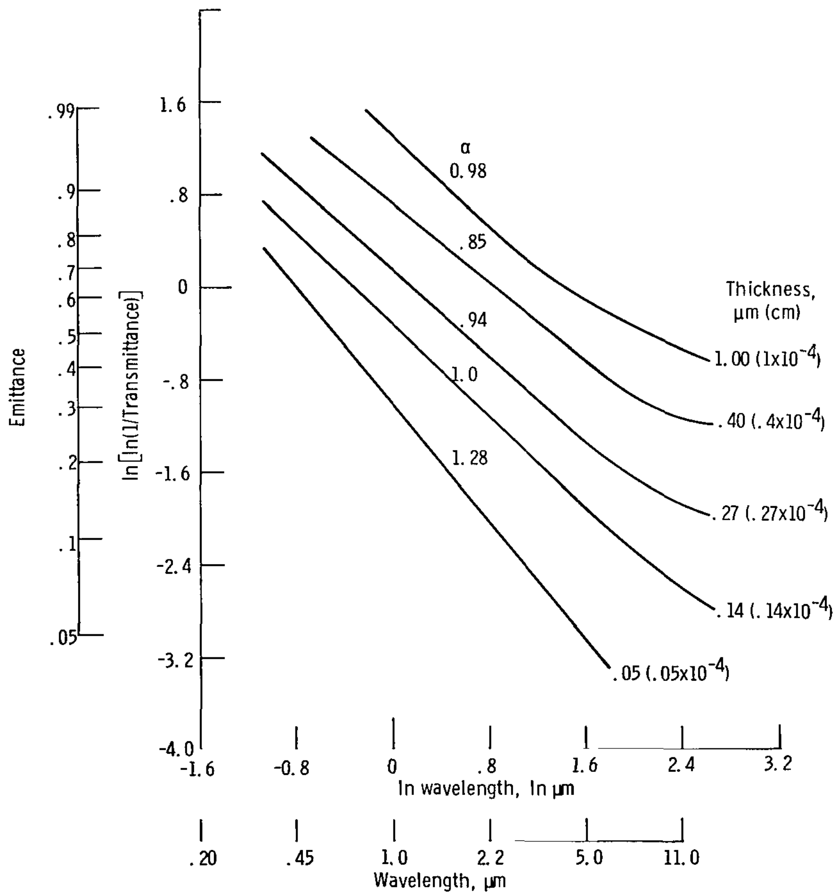


Figure 4. - Normal spectral emittance and transmittance at various thicknesses of soot layers. (Data taken at 300 and 670 K.)

Except for the thinnest film, these data suggest a value of $\alpha = 0.95 \pm 0.10$ for all wavelengths. This is the same as the Hottel recommendation at longer wavelengths but differs considerably from the 1.39 value recommended at shorter wavelengths. The values of α found herein are also substantially the same as those for most of the data of Siddall and McGrath. However, the latter authors interpreted a change in α from 0.9 to 1.15 as the effect of changing the carbon-hydrogen ratio of the fuels, independent of the soot layer thickness, which was not measured. The present work suggests that the same range of α values can be found with a single fuel when the thickness varies. A dimensional analysis argument along with the experimental value of 0.95 ± 0.10 suggests that the true value of α might be unity.

In general, the present data confirm the earlier work on the wavelength dependence of the spectral emittance of soot. They also confirm within reasonable limits the numerical value of the exponent describing this wavelength dependence. However, this work questions the recommendation made in reference 4 that the exponent varies with wavelength and also questions the suggestion in reference 10 that the exponent varies with fuel source.

Finally, the agreement between experiment and both the Mie and Hottel theories suggests that the contribution of soot to radiant heat transfer can be calculated with confidence provided that the soot concentration is known. This has already been established (e.g., ref. 3), although there is the practical problem of estimating soot concentrations in flames. A more important application may be in the estimation of soot concentrations in jet engine combustors by using spectral emittance data. Such data could be obtained through windows in wavelength regions where there is little interfering radiation from carbon dioxide and water vapor. Then, knowing path length L and taking K_λ from figure 2, the concentration of soot $\bar{c}$ can be calculated from equation (2).

CONCLUSIONS

The normal spectral emittances of soot deposited from a stearin candle onto substrates were experimentally determined at measured thicknesses varying from 0.05 to 1 micrometer (0.05×10^{-4} to 10^{-4} cm). Data were obtained at wavelengths of 0.35 to 14 micrometers at 300 K and from 2 to 14 micrometers at 670 K. The emittance of the soot layer did not vary with temperature but varied greatly with thickness and wavelength.

The results indicate that good assessments of the emittance of soot deposited on substrates can be obtained by use of the Mie-Foster absorption theory over wide ranges of temperature, thickness, and wavelength when the density of the soot deposit is assumed to be the same as that of the pure solid. Since the soot is more agglomerated when placed on substrates than when present in luminous flames, the Mie-Foster theory, which assumes zero agglomeration, should also be valid in predicting the radiant heat flux from the luminous flames.

The good agreement between theory and experiment also suggests that soot concentrations in turbojet combustors can be estimated with considerable confidence from radiation data by using Mie theory.

The empirical relation that absorption coefficient is inversely proportional to wavelength raised to a power is valid over large ranges of temperature, thickness, and wavelength. The data obtained herein suggest that the wavelength exponent equals 0.95±0.10 when this relation is used in Hottel's method for obtaining flame temperatures.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, December 5, 1969,
129-01.

REFERENCES

1. McAdams, William H.: Heat Transmission. Third ed., McGraw-Hill Book Co., Inc., 1954.
2. Toone, B.: A Review of Aero Engine Smoke Emission. Combustion in Advanced Gas Turbine Systems. I. E. Smith, ed., Pergamon Press, 1968.
3. Thring, M. W.; Foster, P. J.; McGrath, I. A.; and Ashton, J. S.: Prediction of the Emissivity of Hydrocarbon Flames. International Developments in Heat Transfer. ASME, 1963, pp. 796-803.
4. Hottel, H. C.; and Broughton, F. P.: Determination of True Temperature and Total Radiation from Luminous Gas Flames. Ind. Eng. Chem., Anal. Ed., vol. 4, no. 2, Apr. 15, 1932, pp. 166-175.
5. Svet, Darĭ Ia.: Thermal Radiation. Consultants Bureau, 1965.
6. Liebert, Curt H.; and Thomas, Ralph D.: Spectral Emissivity of Highly Doped Silicon. NASA TN D-4303, 1968.
7. Stull, V. Robert; and Plass, Gilbert N.: Emissivity of Dispersed Carbon Particles. J. Opt. Soc. Am., vol. 50, no. 2, Feb. 1960, pp. 121-129.
8. Foster, P. J.: Calculation of the Optical Properties of Dispersed Phases. Comb. Flame, vol. 7, no. 3, Sept. 1963, pp. 277-282.
9. Mantell, Charles L.: Carbon and Graphite Handbook. Interscience Publ., 1968.
10. Siddall, R. G.; and McGrath, I. A.: The Emissivity of Luminous Flames. Ninth Symposium (International) on Combustion. Academic Press, 1963, pp. 102-110.

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
WASHINGTON, D. C. 20546

OFFICIAL BUSINESS

FIRST CLASS MAIL

POSTAGE AND FEES PAID
NATIONAL AERONAUTICS A
SPACE ADMINISTRATION

009 001 31 51 3DS 70033 00903
AIR FORCE WEAPONS LABORATORY /WLOL/
KIRTLAND AFB, NEW MEXICO 87117

ATT E. LOU BOWMAN, CHIEF,TECH. LIBRARY

POSTMASTER: If Undeliverable (Section 15
Postal Manual) Do Not Retu

"The aeronautical and space activities of the United States shall be conducted so as to contribute . . . to the expansion of human knowledge of phenomena in the atmosphere and space. The Administration shall provide for the widest practicable and appropriate dissemination of information concerning its activities and the results thereof."

—NATIONAL AERONAUTICS AND SPACE ACT OF 1958

NASA SCIENTIFIC AND TECHNICAL PUBLICATIONS

TECHNICAL REPORTS: Scientific and technical information considered important, complete, and a lasting contribution to existing knowledge.

TECHNICAL NOTES: Information less broad in scope but nevertheless of importance as a contribution to existing knowledge.

TECHNICAL MEMORANDUMS: Information receiving limited distribution because of preliminary data, security classification, or other reasons.

CONTRACTOR REPORTS: Scientific and technical information generated under a NASA contract or grant and considered an important contribution to existing knowledge.

TECHNICAL TRANSLATIONS: Information published in a foreign language considered to merit NASA distribution in English.

SPECIAL PUBLICATIONS: Information derived from or of value to NASA activities. Publications include conference proceedings, monographs, data compilations, handbooks, sourcebooks, and special bibliographies.

TECHNOLOGY UTILIZATION PUBLICATIONS: Information on technology used by NASA that may be of particular interest in commercial and other non-aerospace applications. Publications include Tech Briefs, Technology Utilization Reports and Notes, and Technology Surveys.

Details on the availability of these publications may be obtained from:

SCIENTIFIC AND TECHNICAL INFORMATION DIVISION

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

Washington, D.C. 20546